

Controlling coherent optical phonons in tellurium

JILA, University of Colorado
Boulder, CO, 20 April 2004





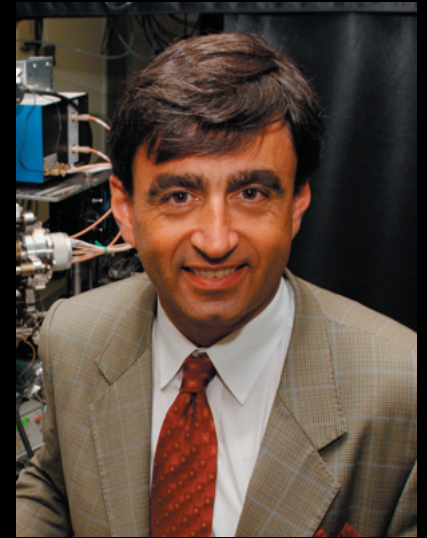
Chris Roeser



Maria Kandyla



Arantza Mendioroz



Eric Mazur

and also....

Albert Kim

Paul Callan

Eli Glezer

Li Huang

Yakir Siegal

Dr. Peter Grosse (Aachen)

Dr. Paul Tangney (Princeton)

Prof. Steven Fahy (Cork)

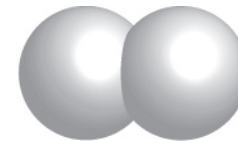
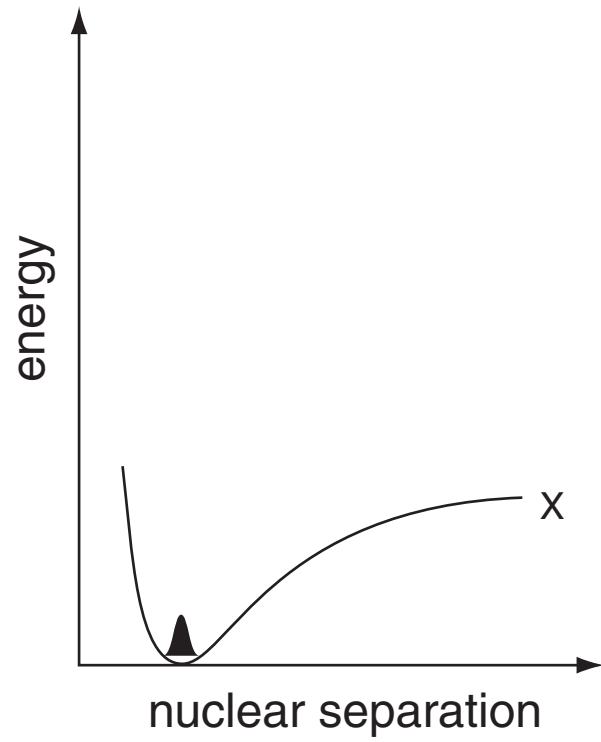
Nick Choly (Harvard University)

Prof. Tim Kaxiras (Harvard University)



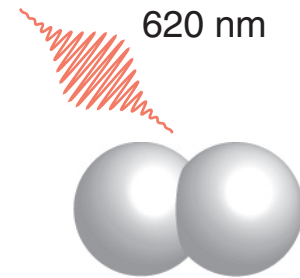
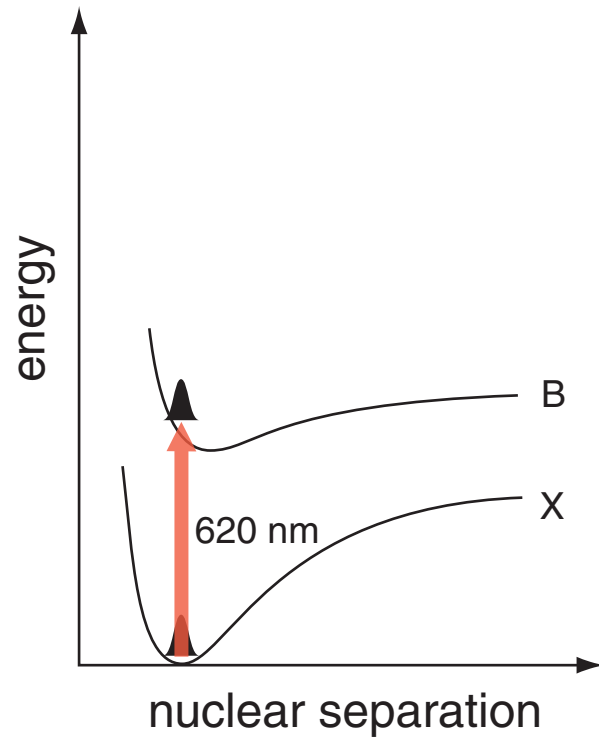
Introduction

iodine



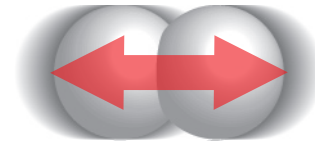
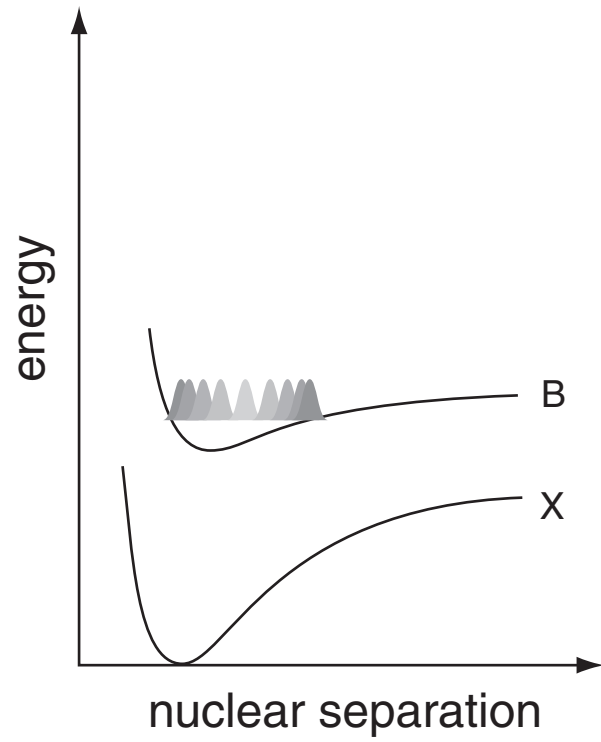
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iodine



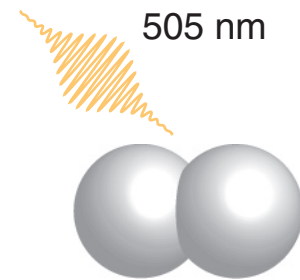
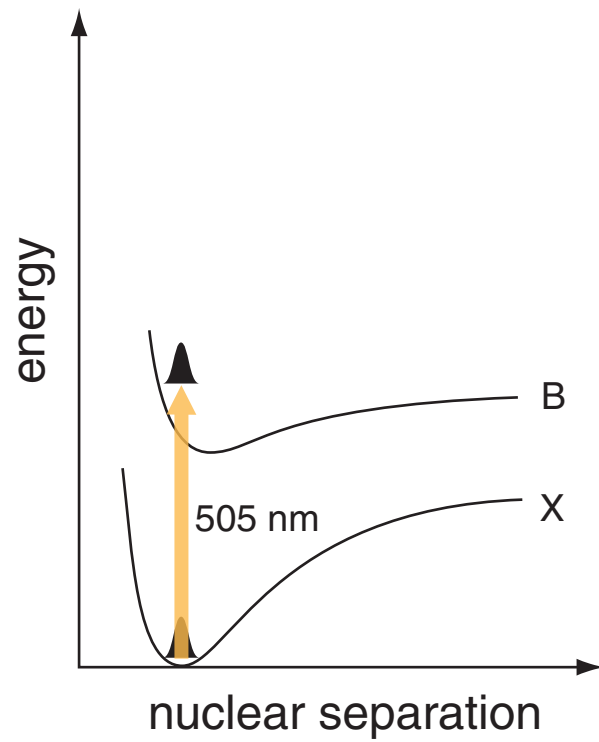
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iodine



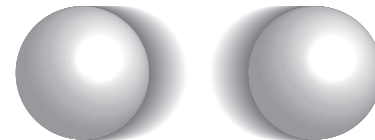
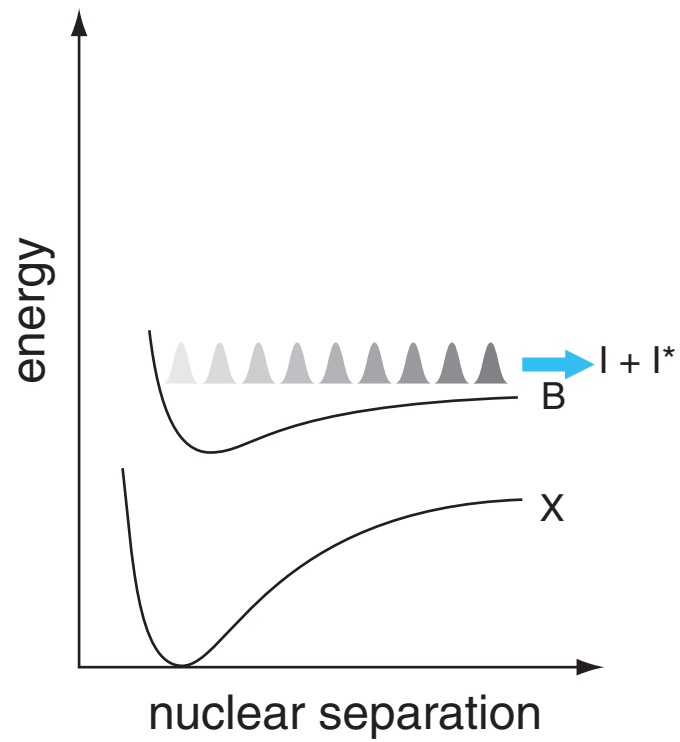
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iodine



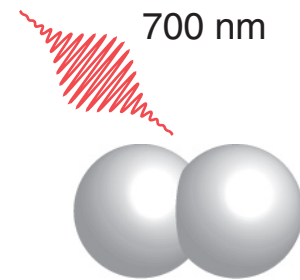
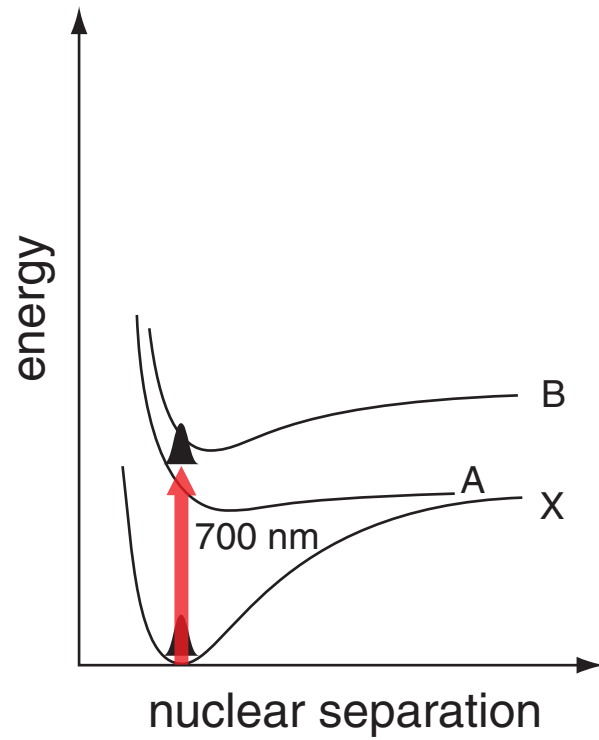
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iodine



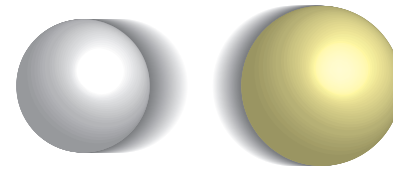
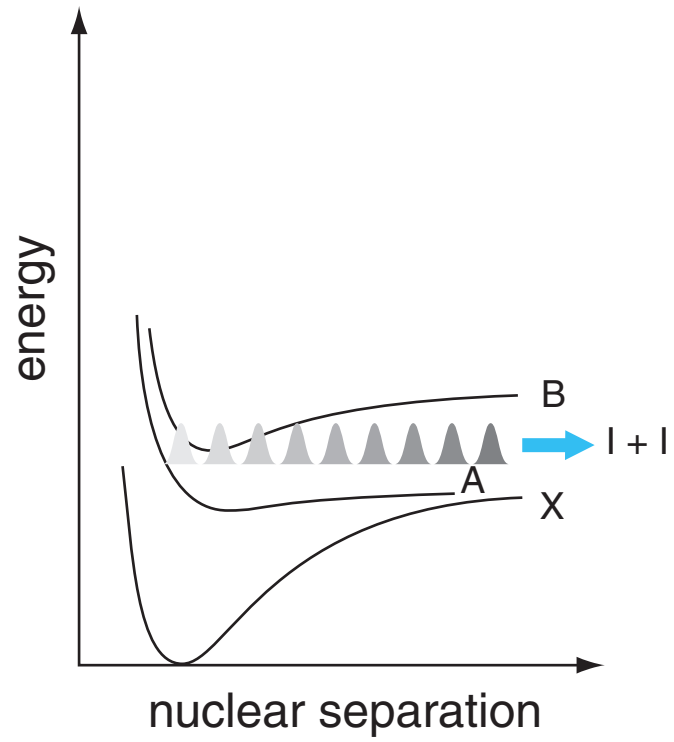
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iodine



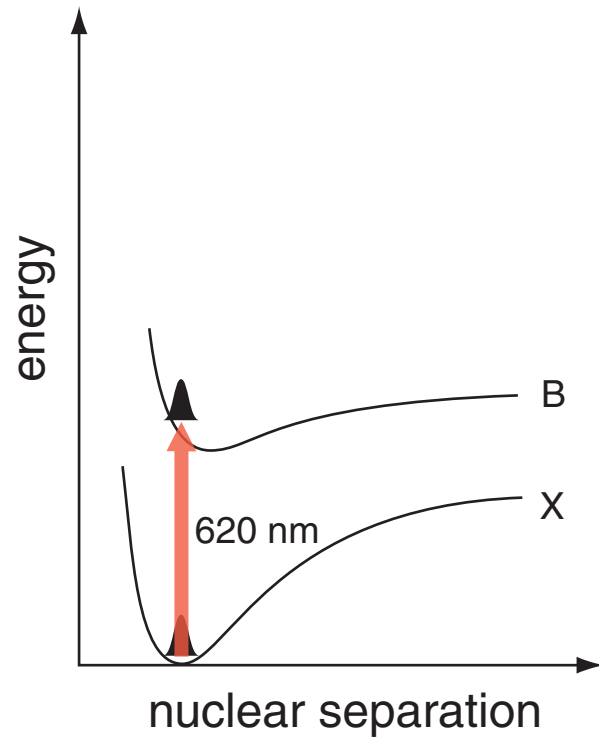
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iodine



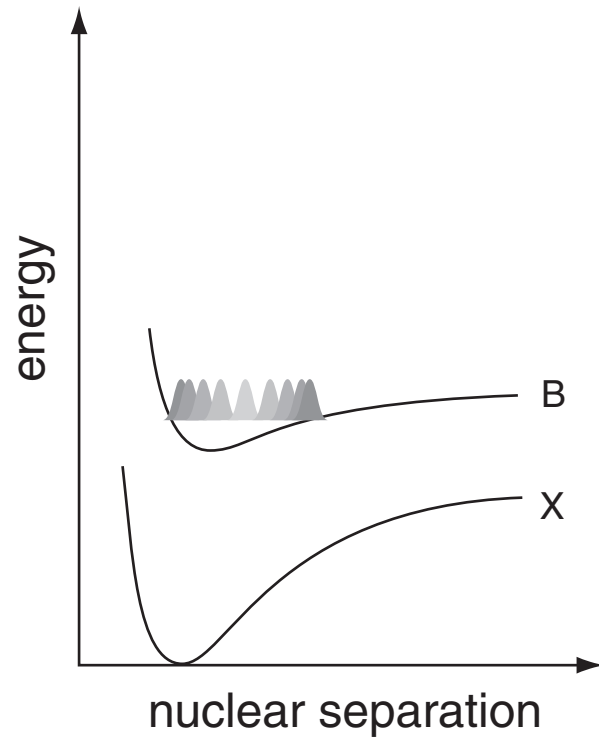
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probe wave packet dynamics



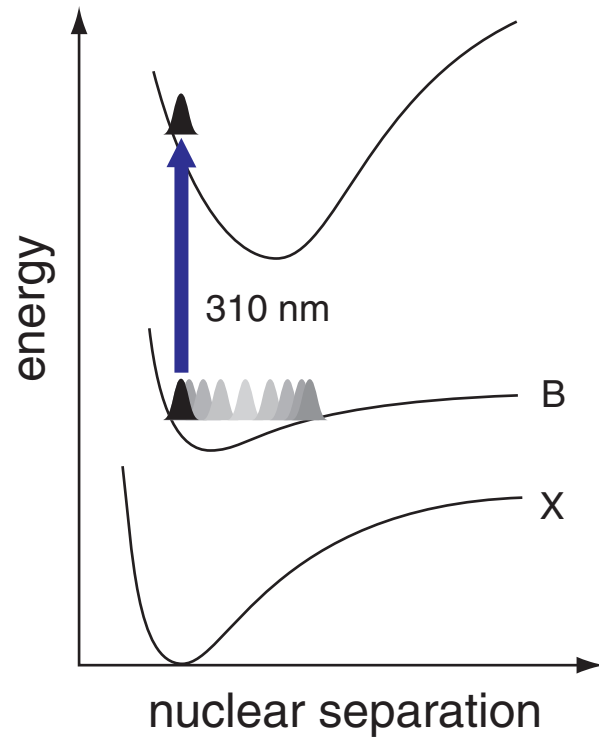
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probe wave packet dynamics



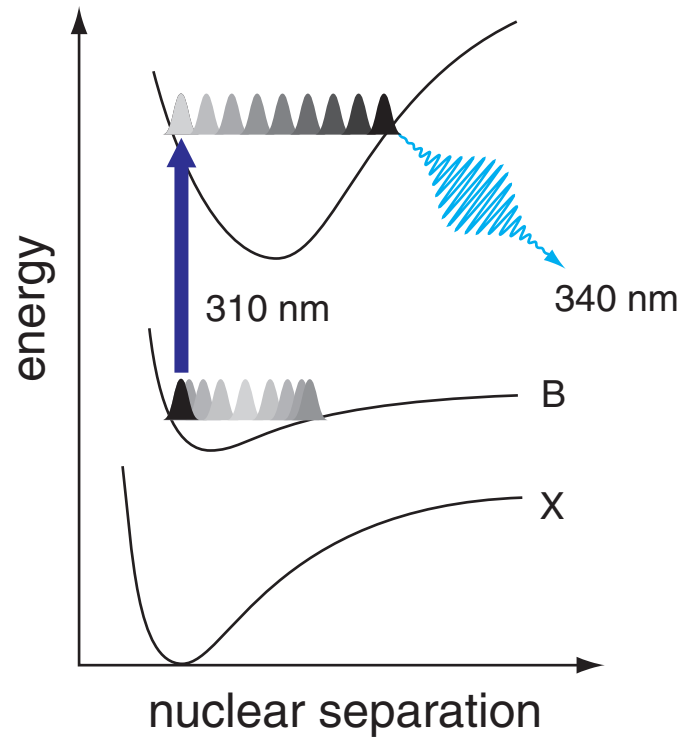
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probe wave packet dynamics



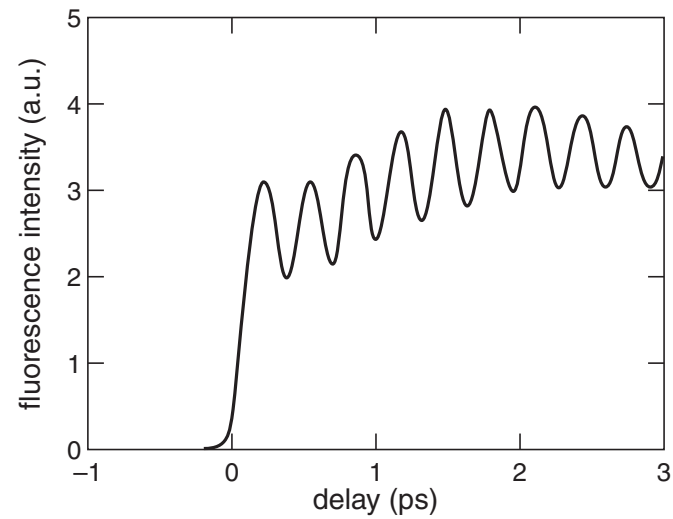
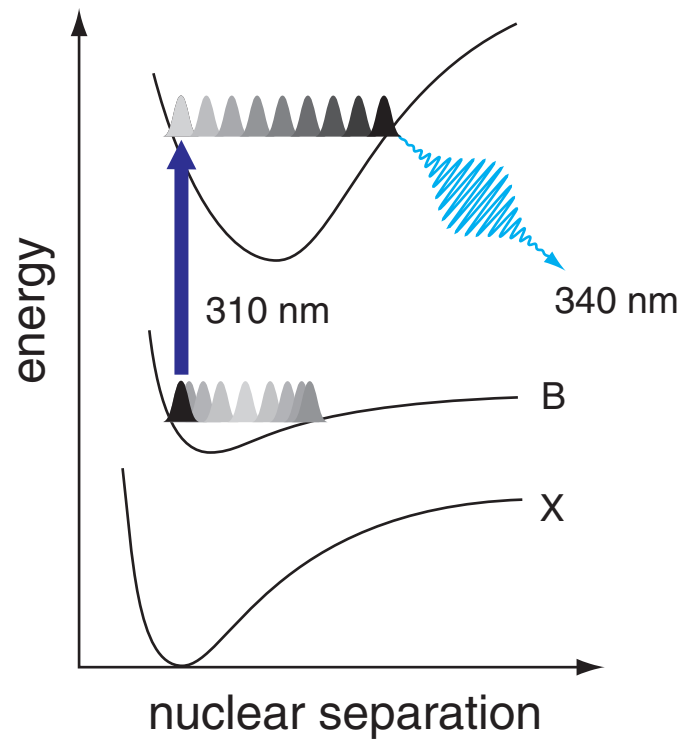
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probe wave packet dynamics



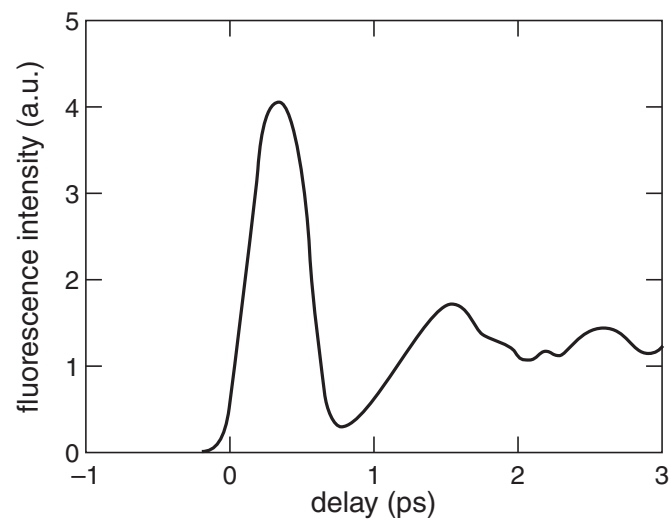
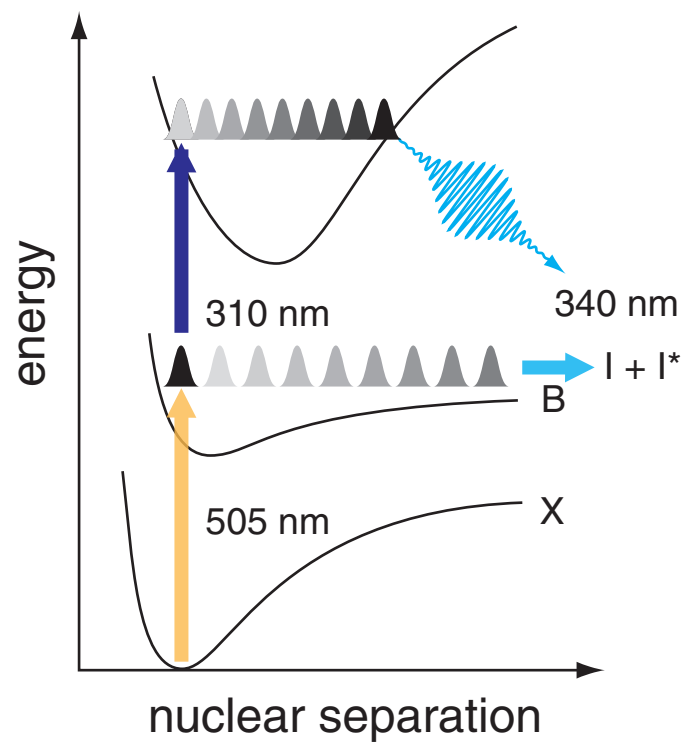
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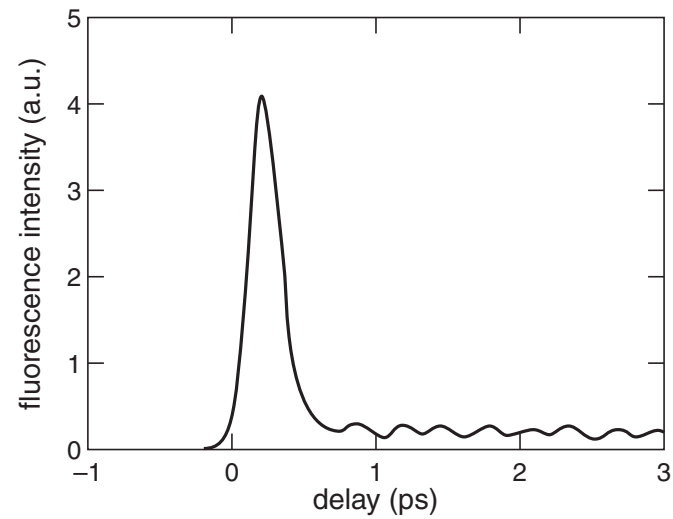
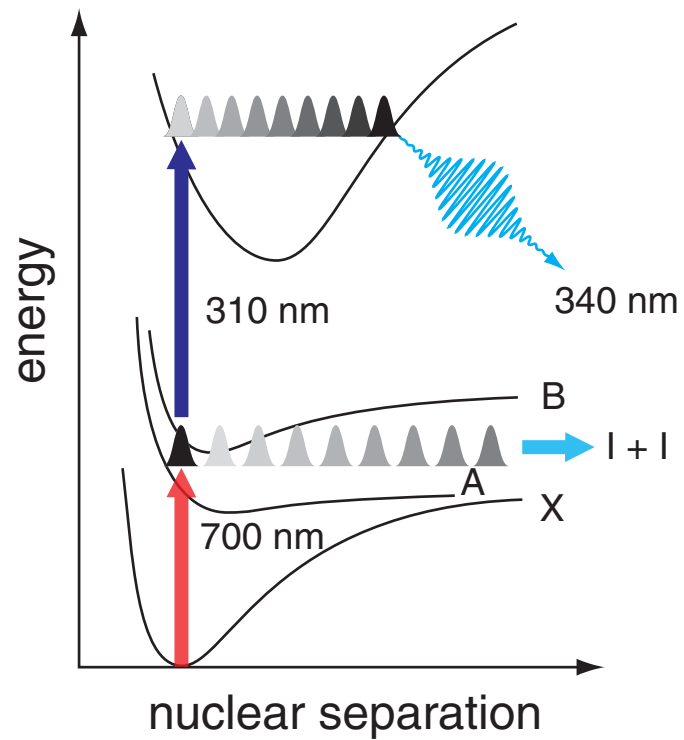
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probe wave packet dynamics



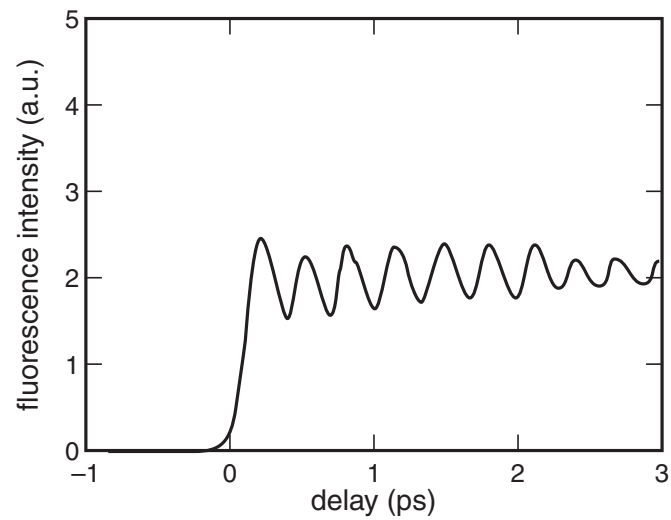
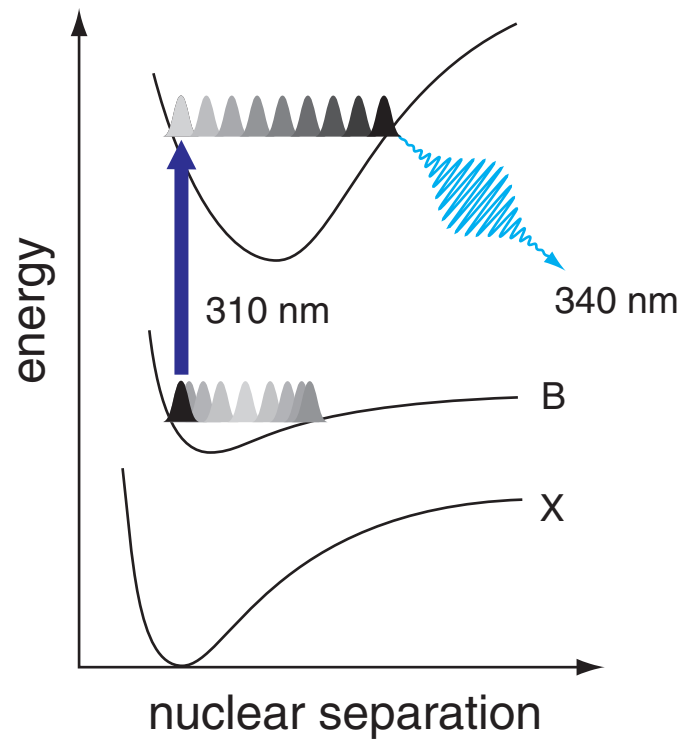
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probe wave packet dynamics



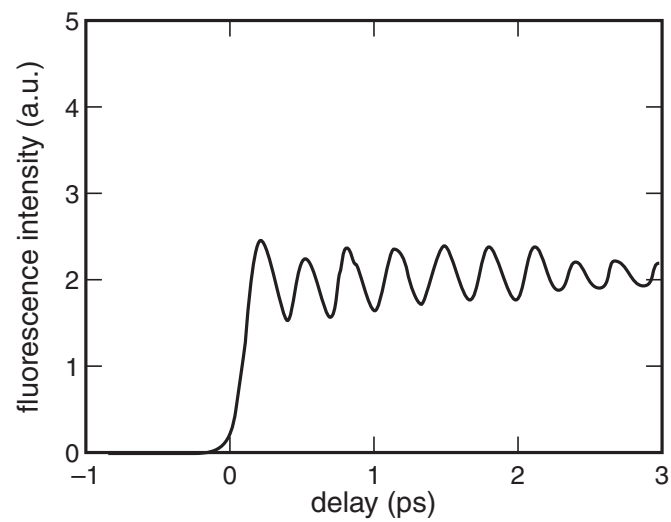
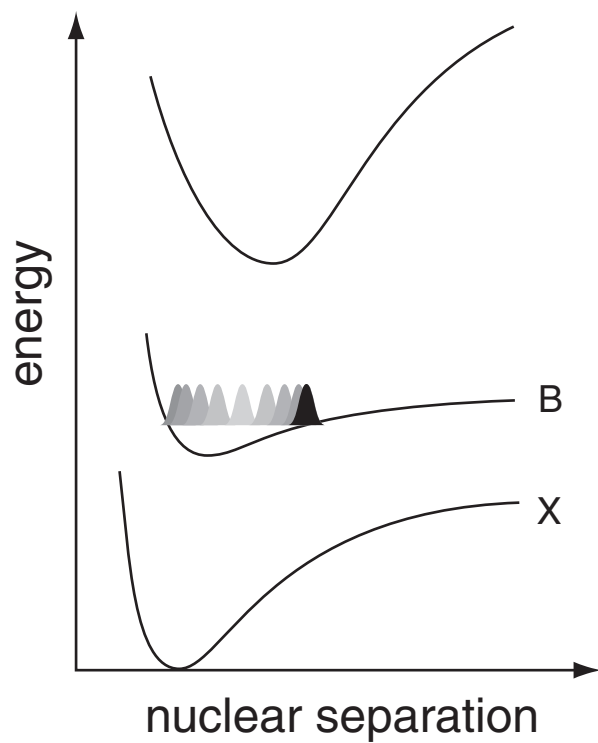
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wave packet control



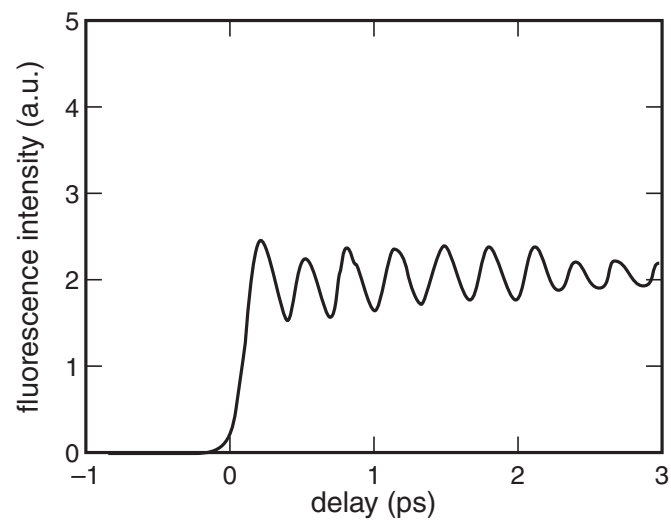
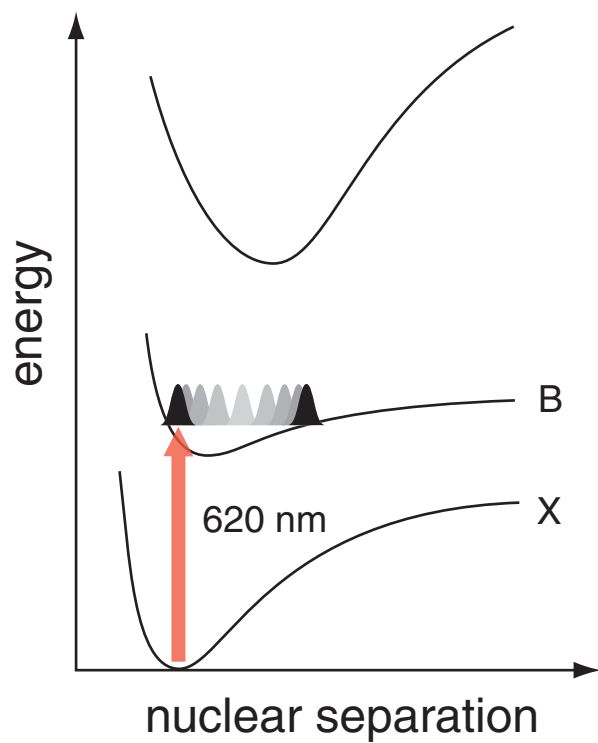
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wave packet control



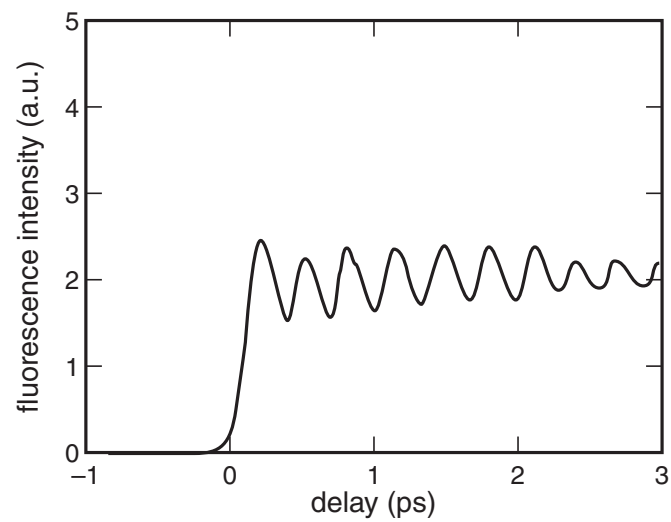
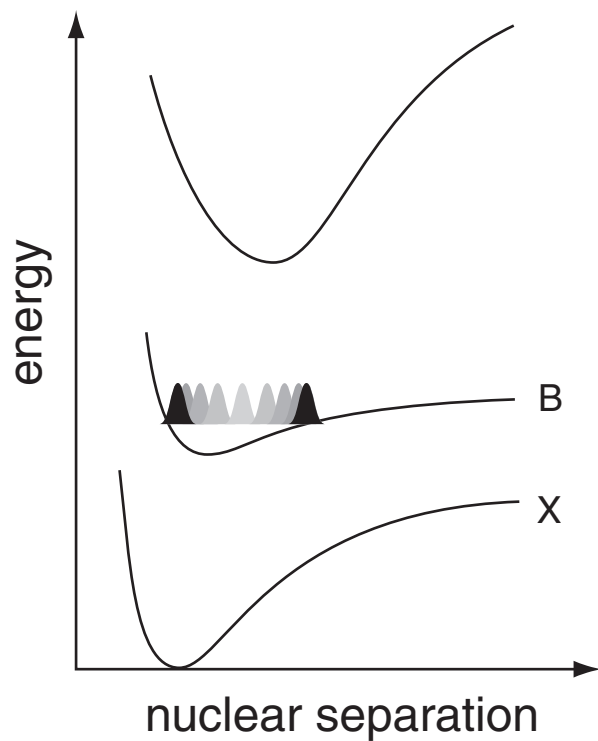
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phase delay: π



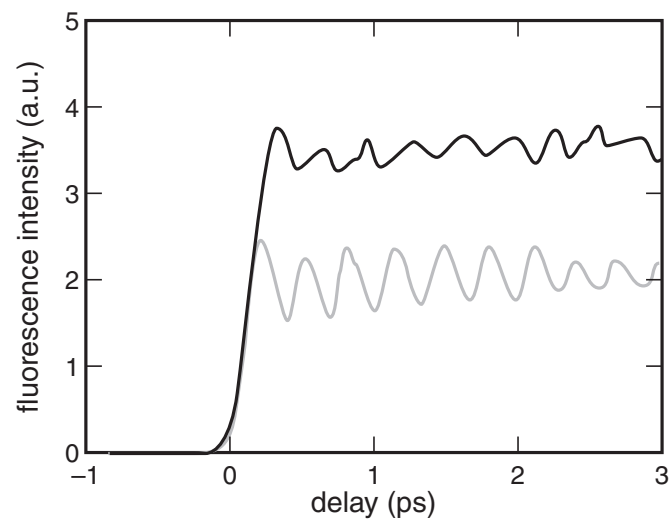
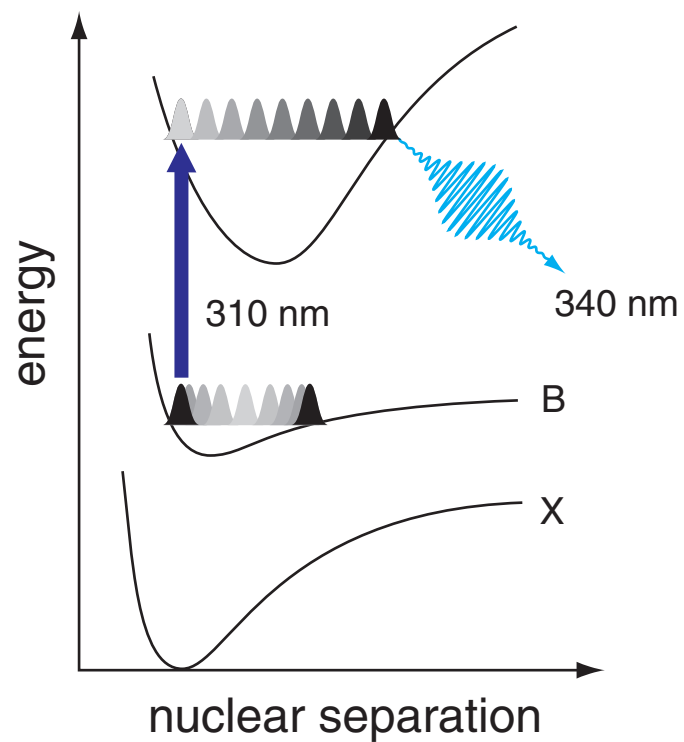
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phase delay: π



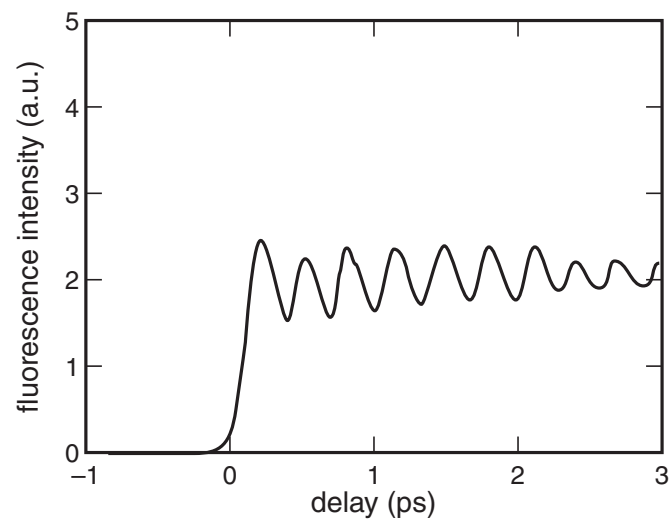
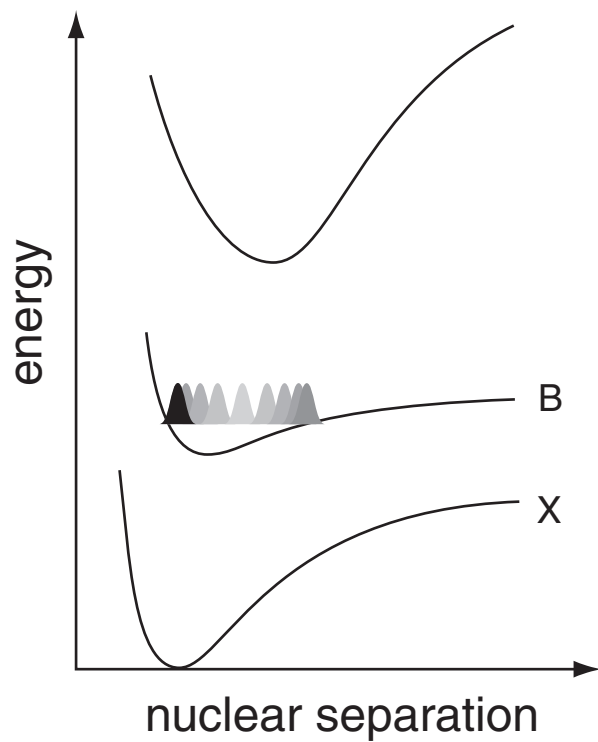
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phase delay: π



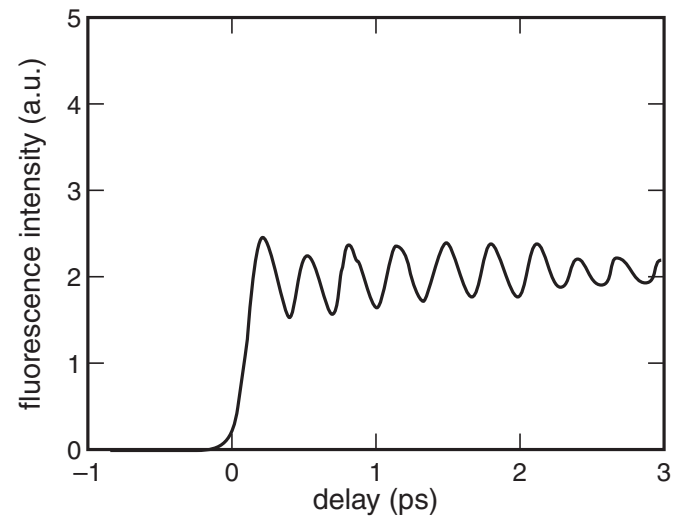
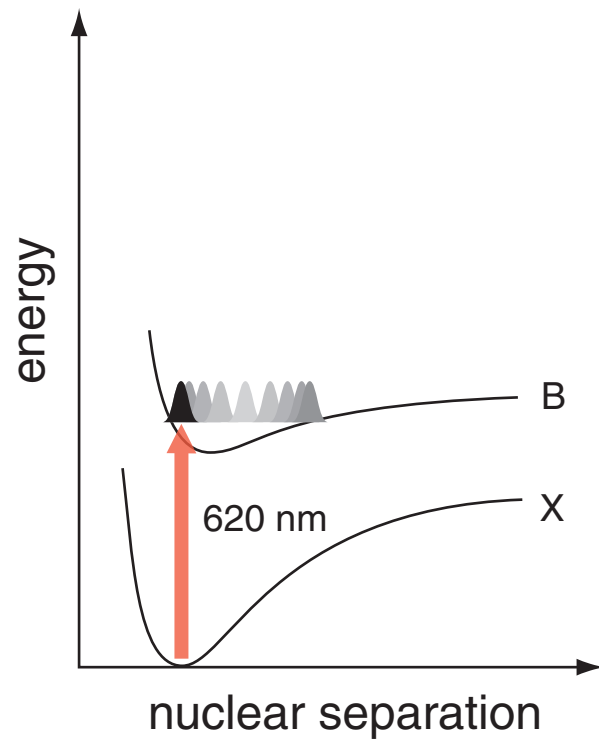
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phase delay: 2π



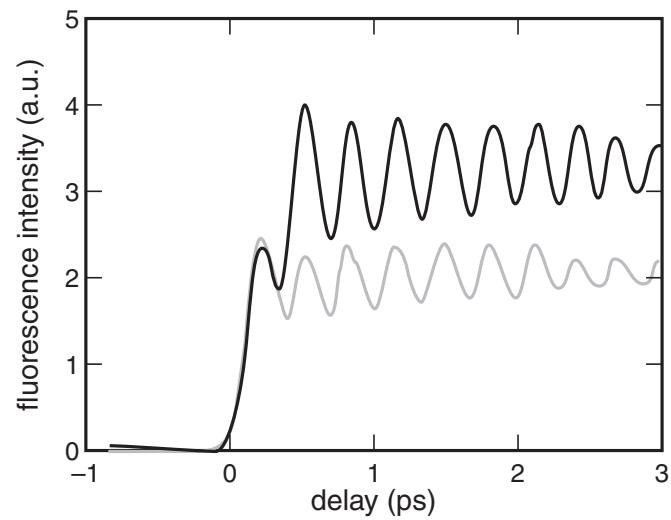
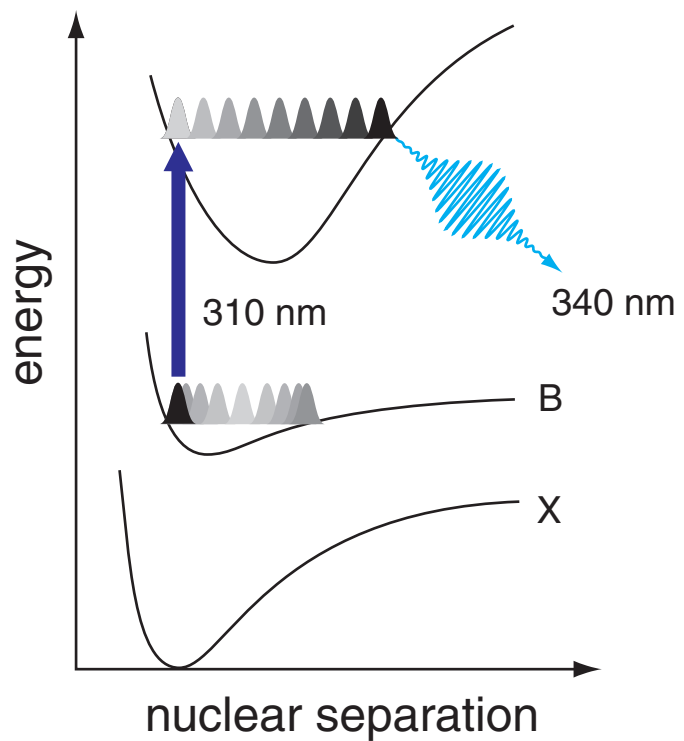
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phase delay: 2π



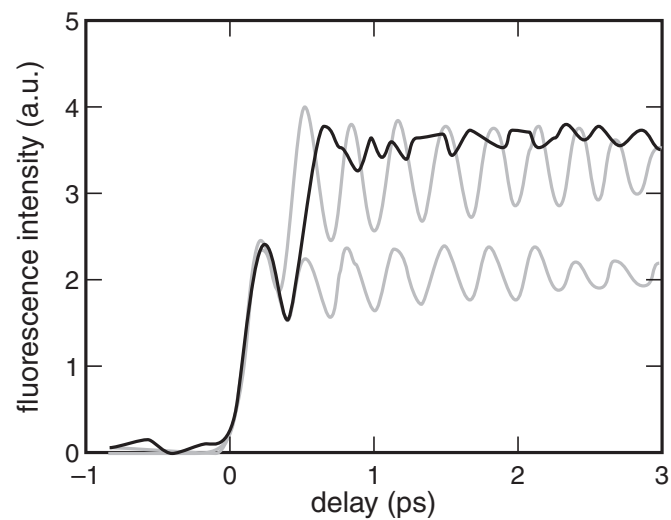
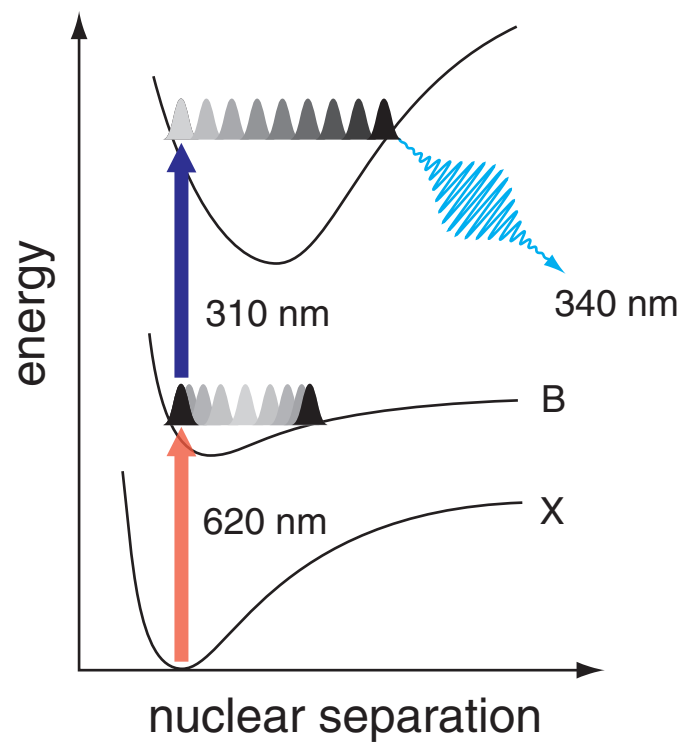
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phase delay: 2π



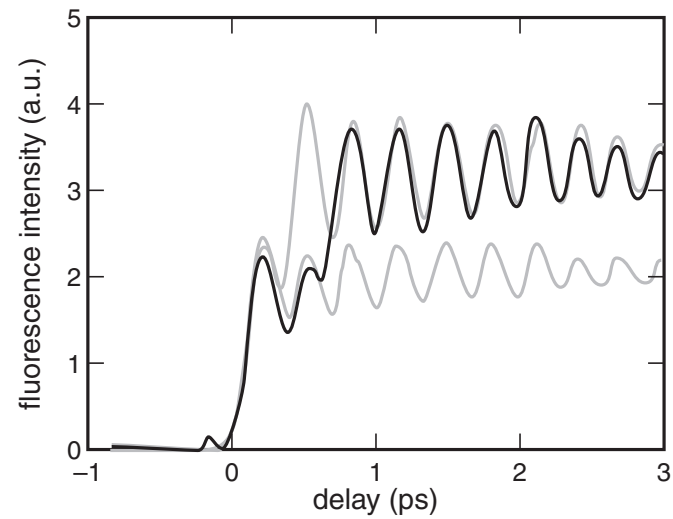
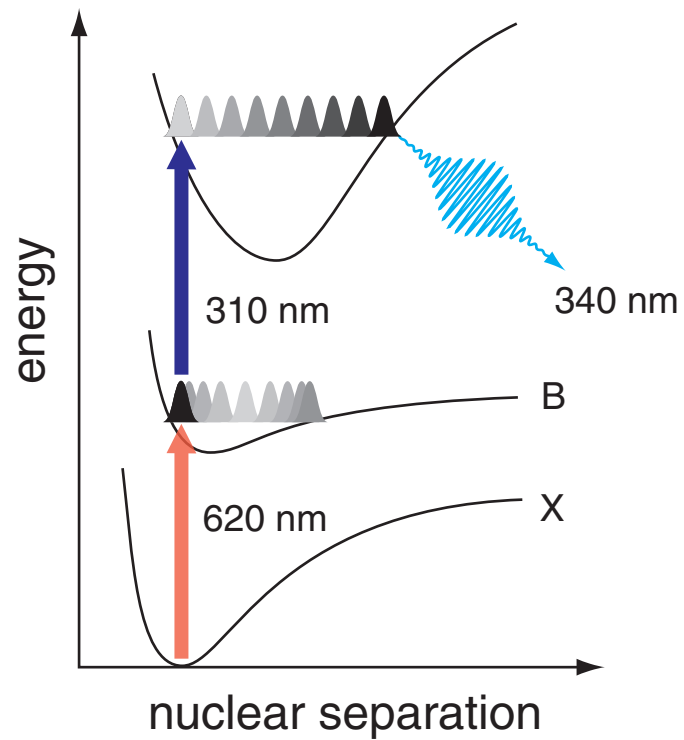
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phase delay: 3π



Introduction

phase delay: 4π

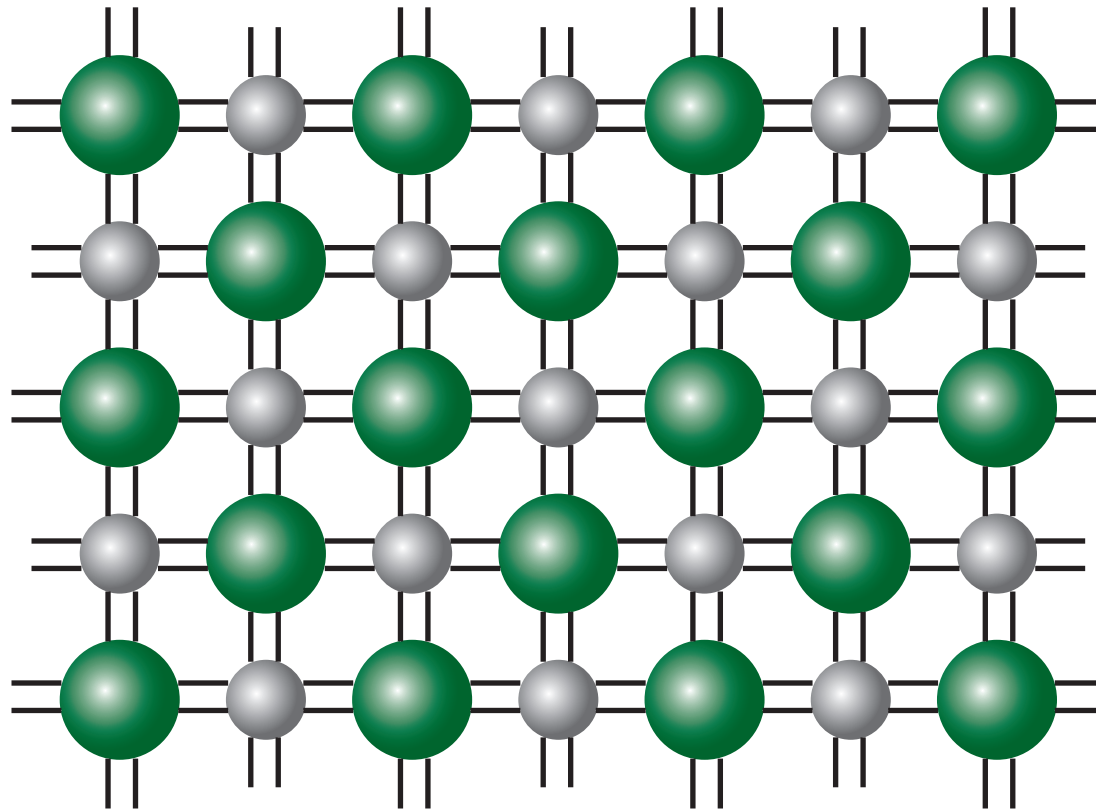


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limited control of chemical reactions demonstrated

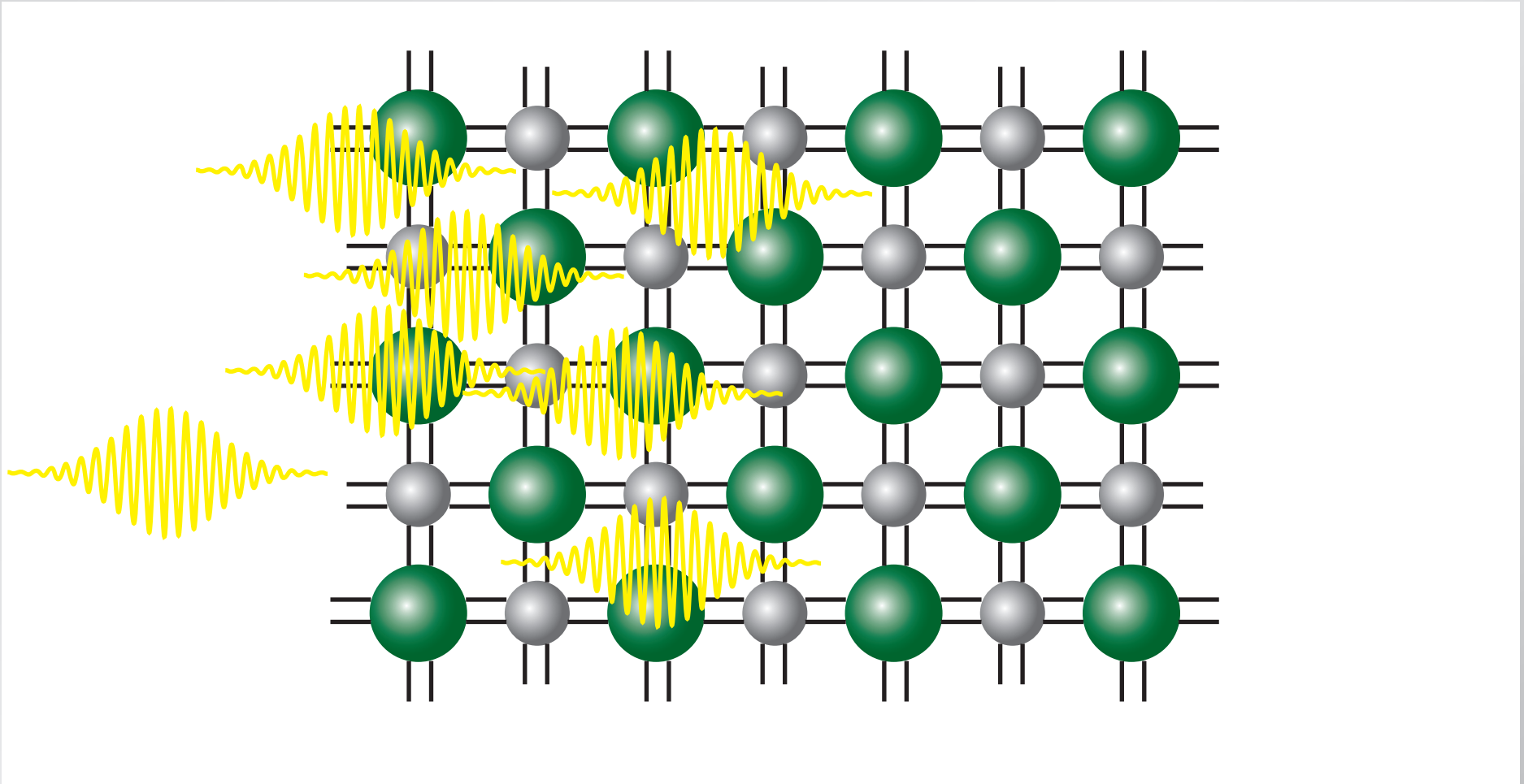
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can we optically control the state of a solid?



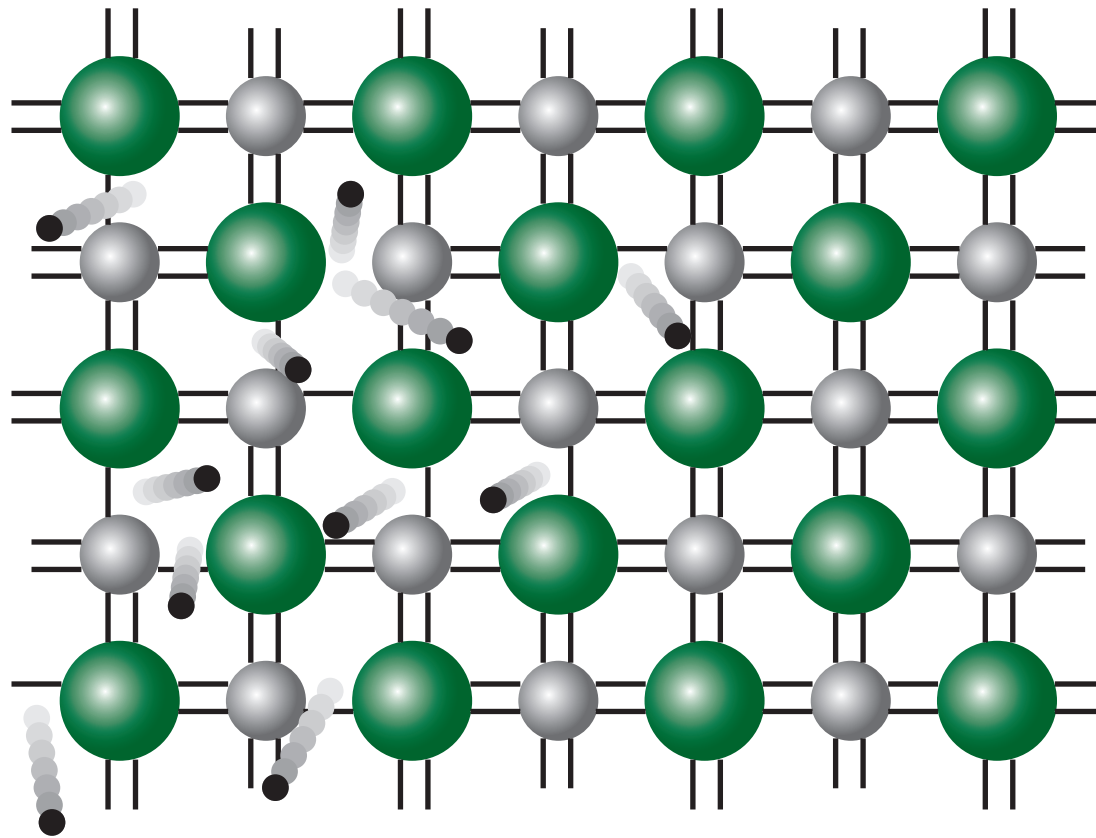
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photons excite valence electrons...



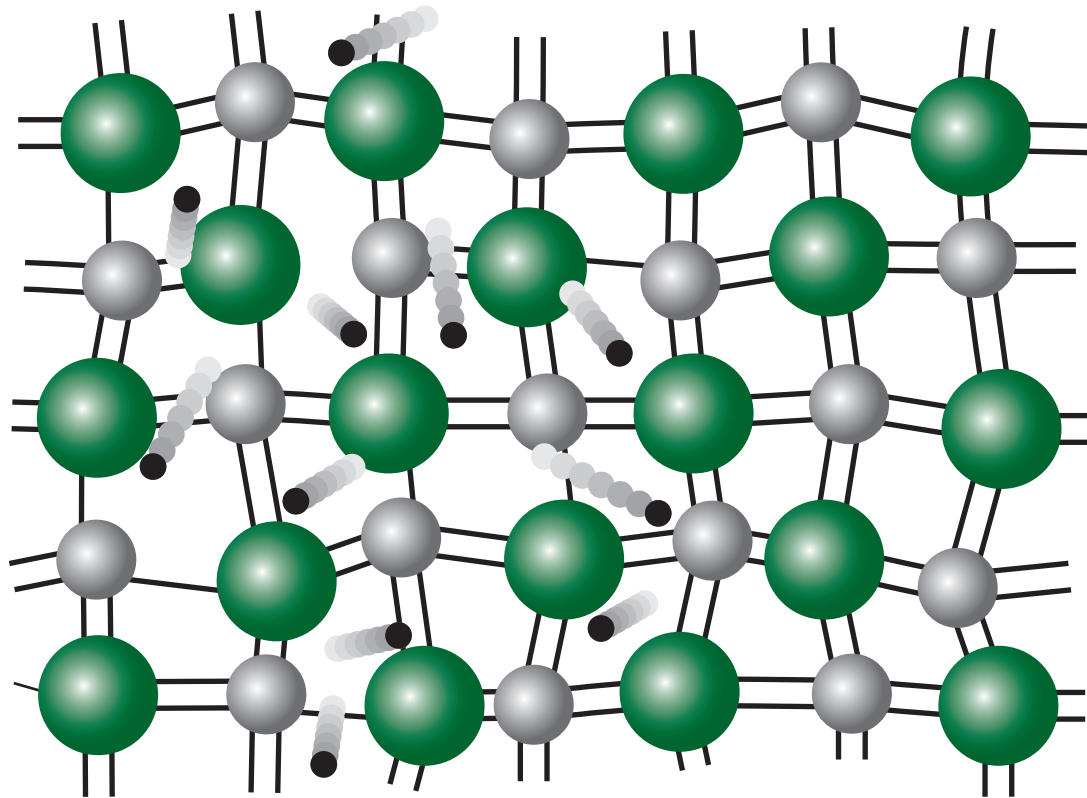
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...and create free carriers...



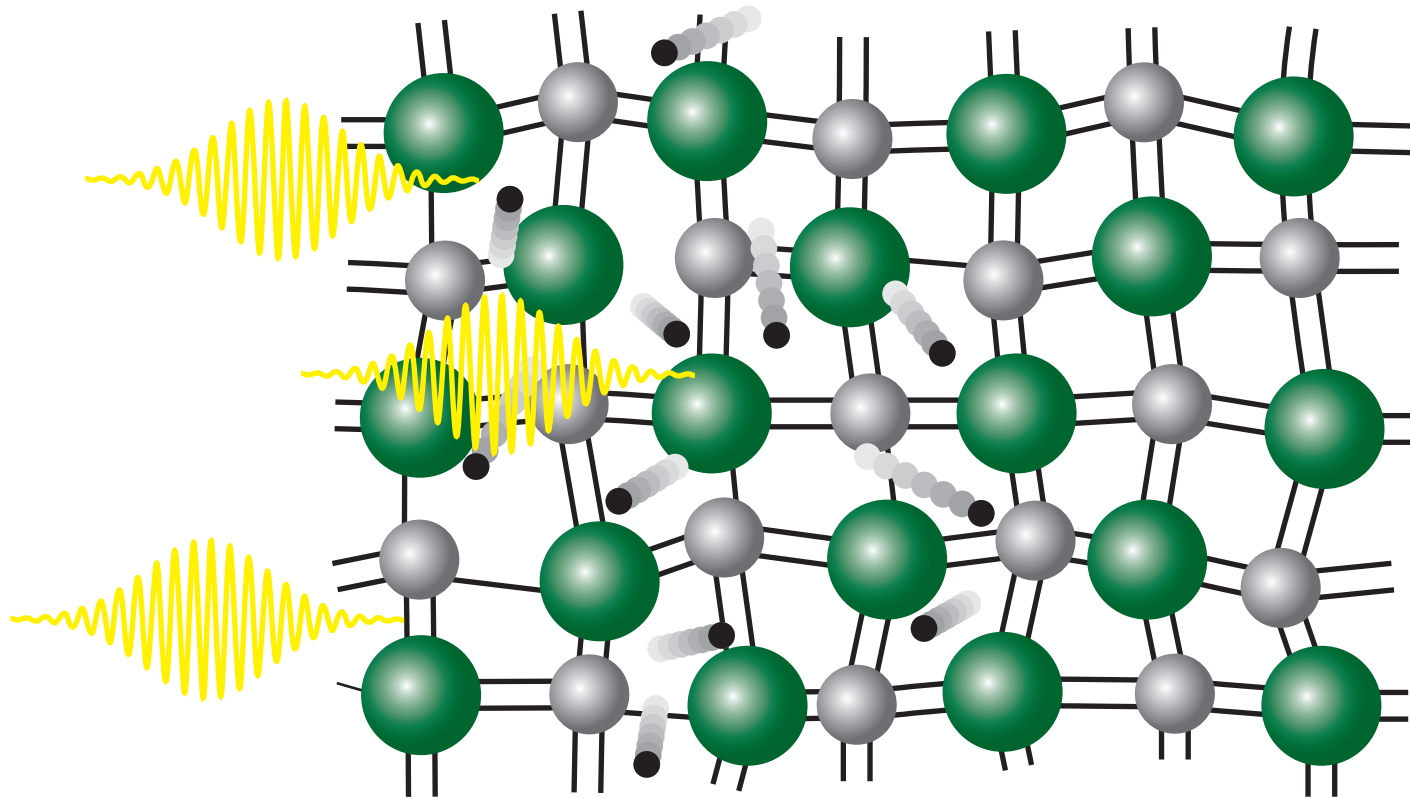
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...causing electronic and structural changes...



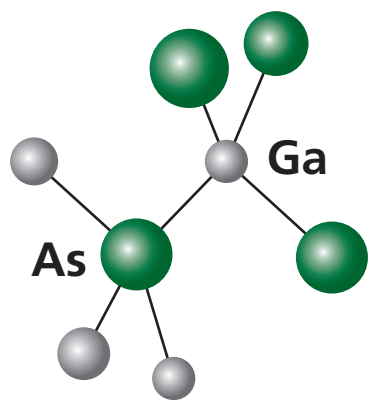
Introduction

...which we detect with a second laser pulse.



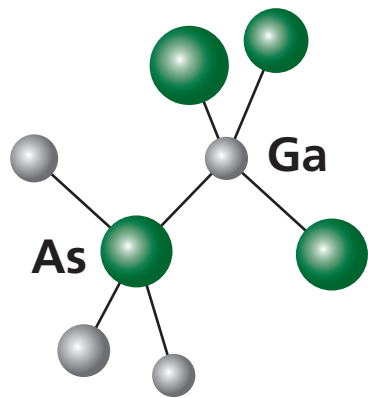
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structure

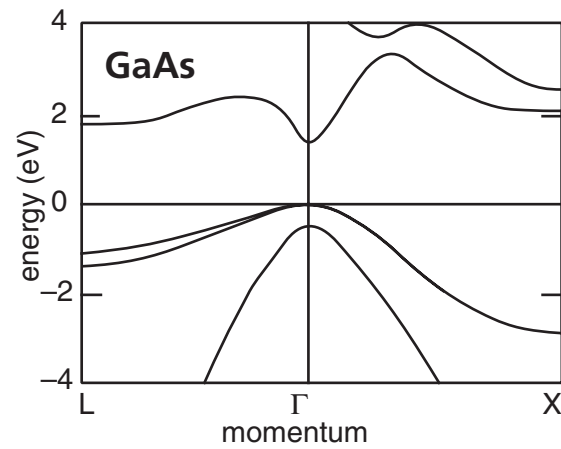


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structure

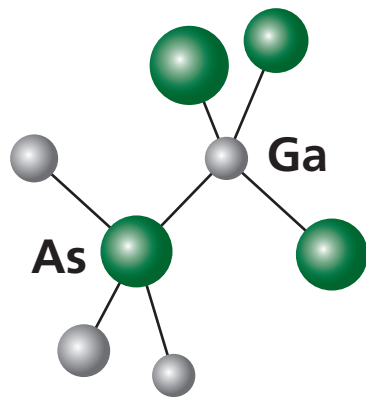


band structure

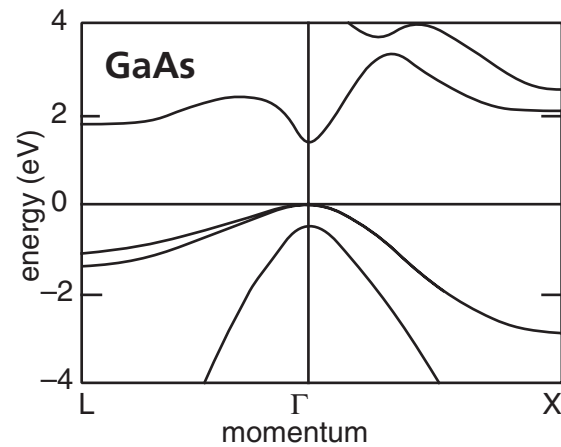


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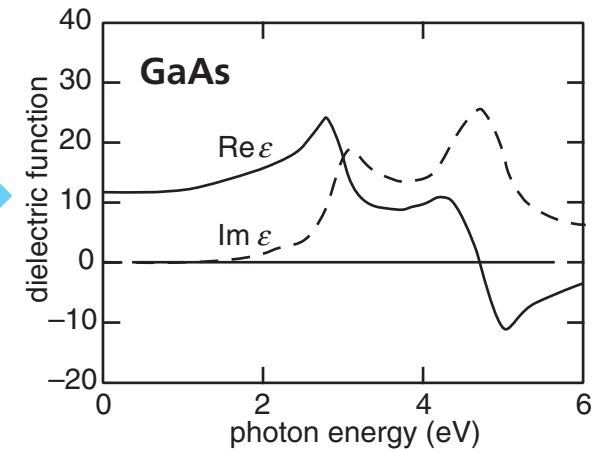
structure



band structure

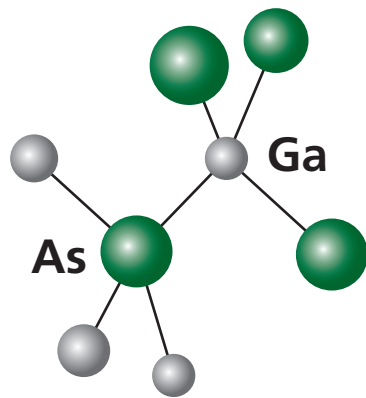


dielectric function

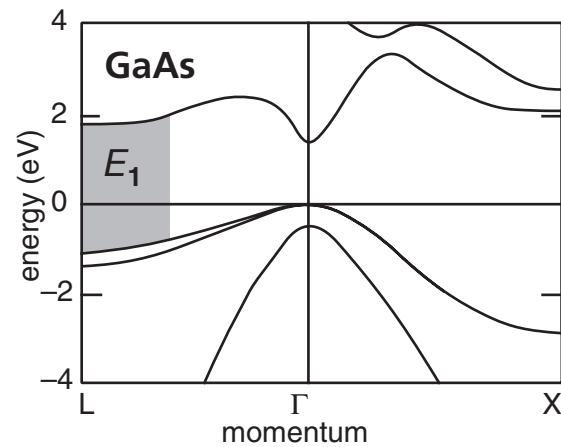


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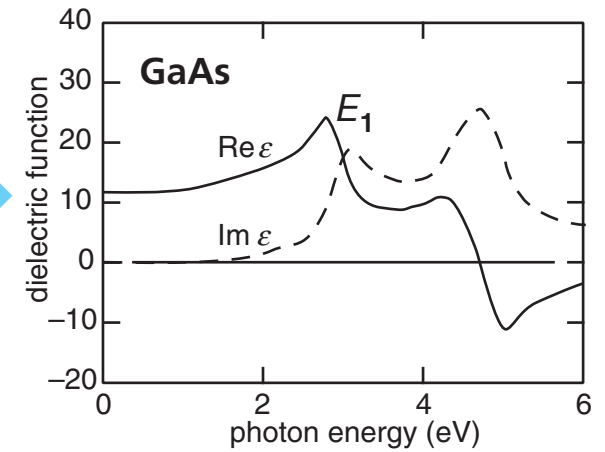
structure



band structure

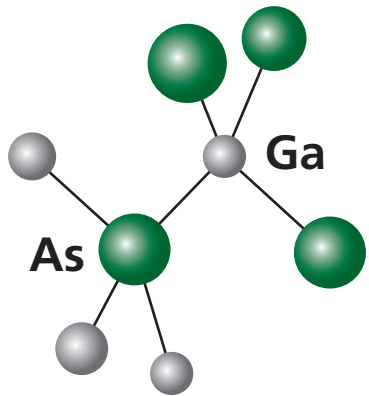


dielectric function

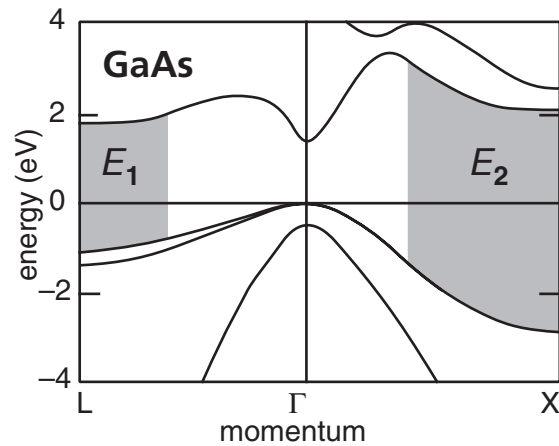


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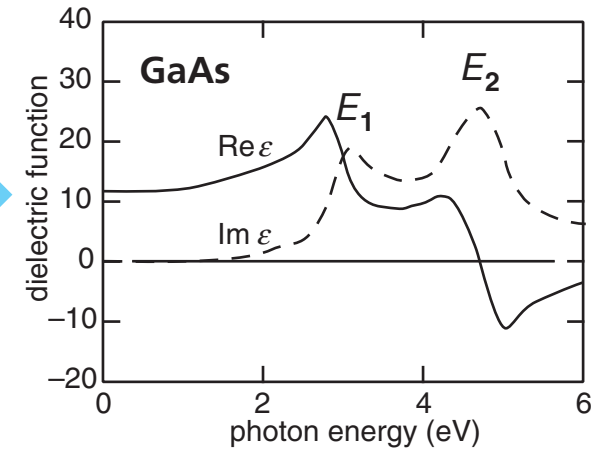
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band structure

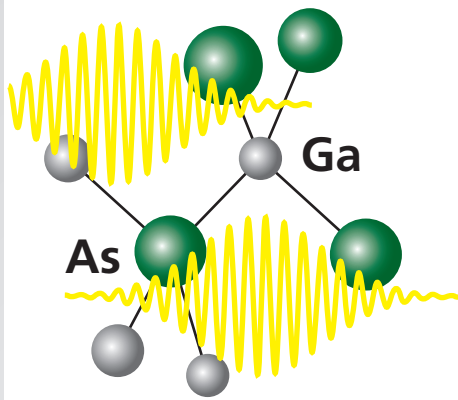


dielectric function

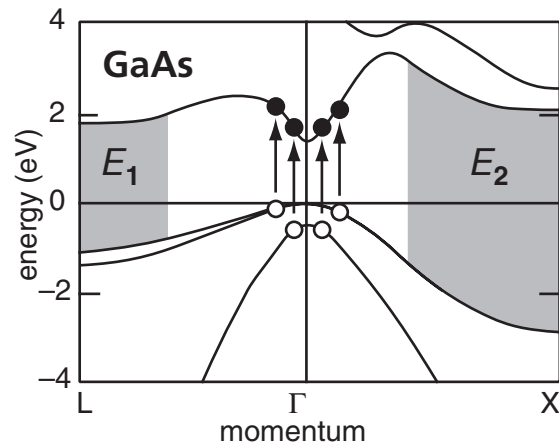


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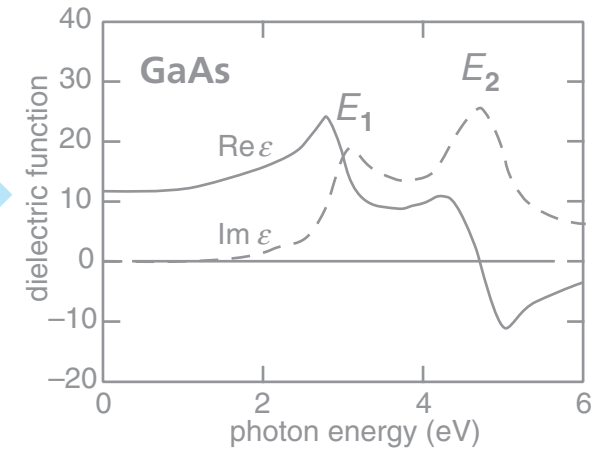
structure



band structure

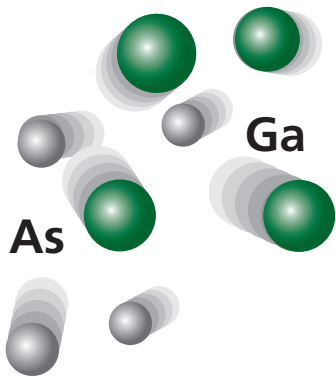


dielectric function

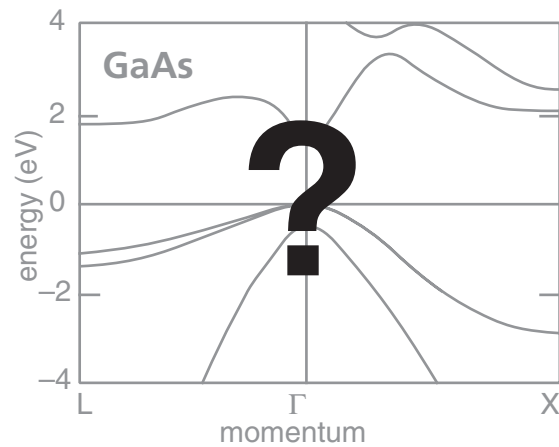


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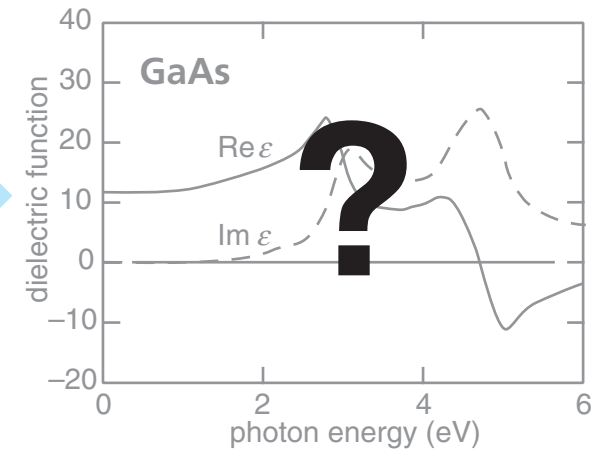
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band structure

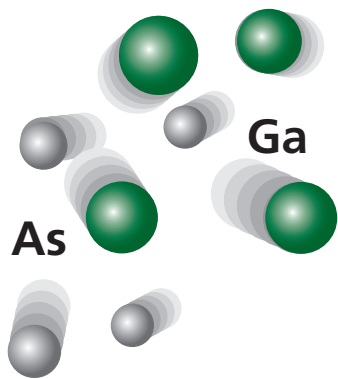


dielectric function

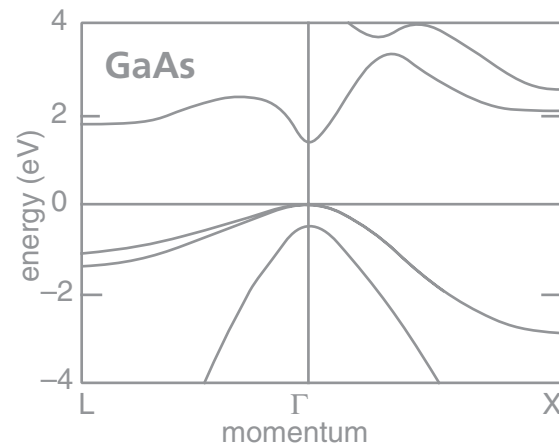


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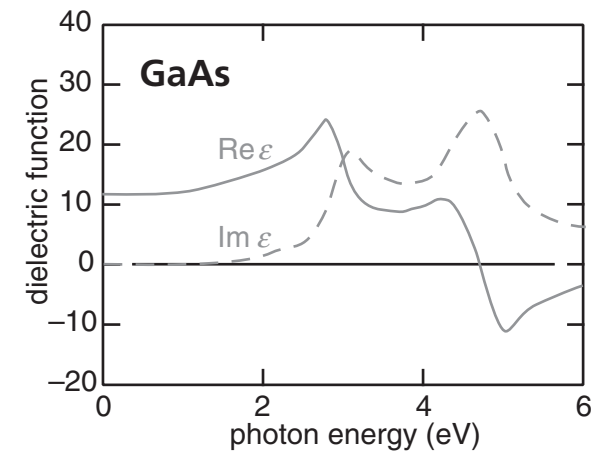
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band structure

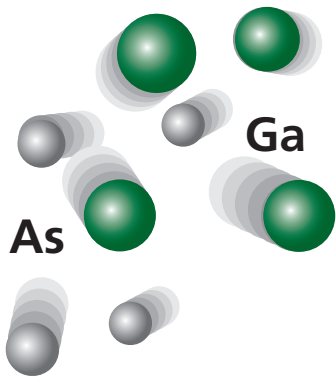


dielectric function

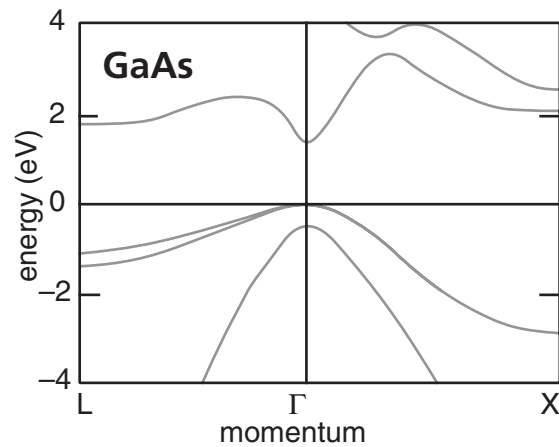


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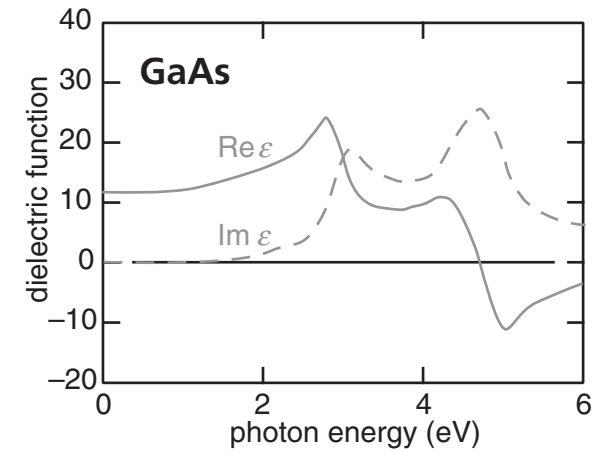
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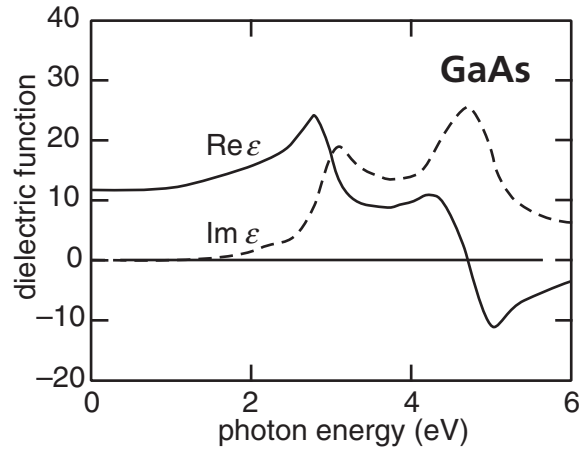
band structure



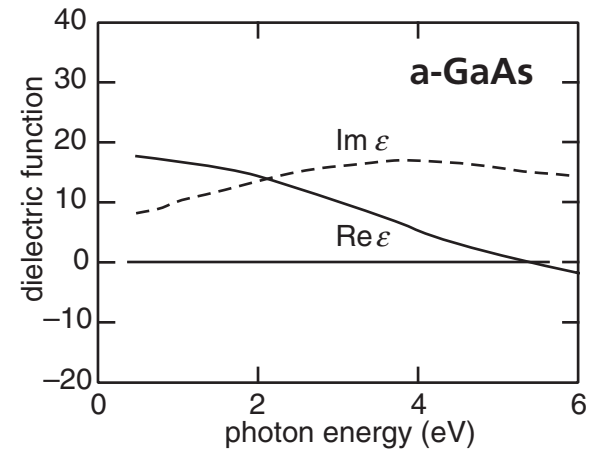
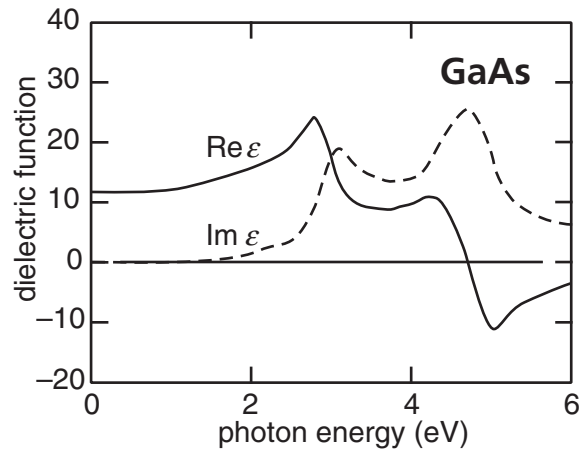
dielectric function



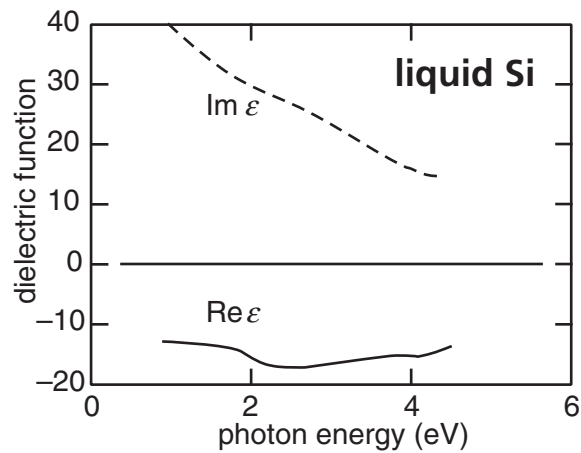
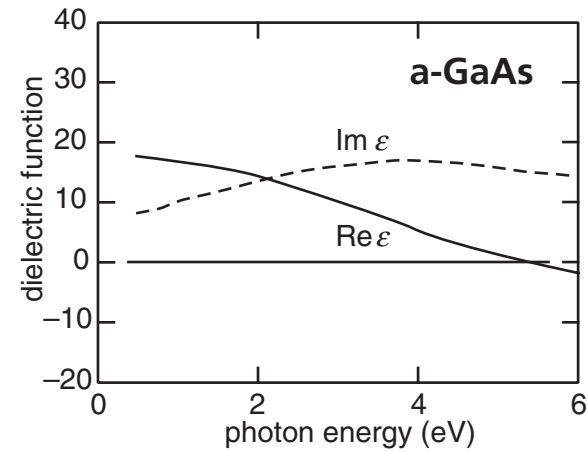
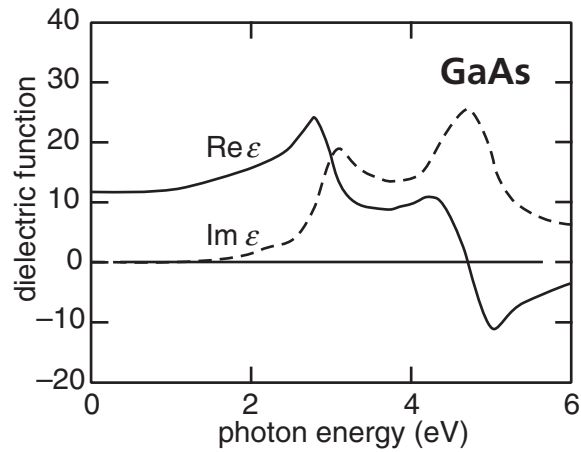
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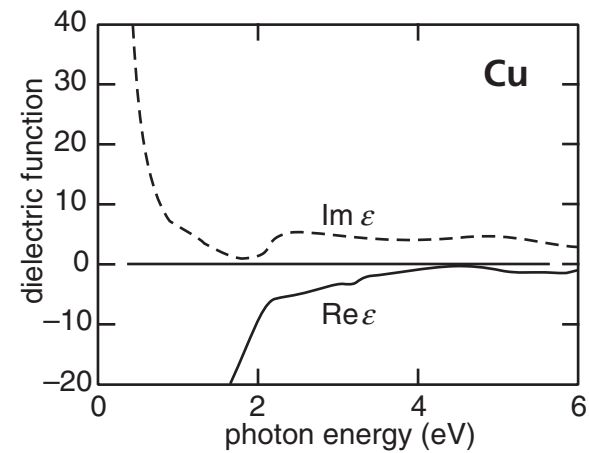
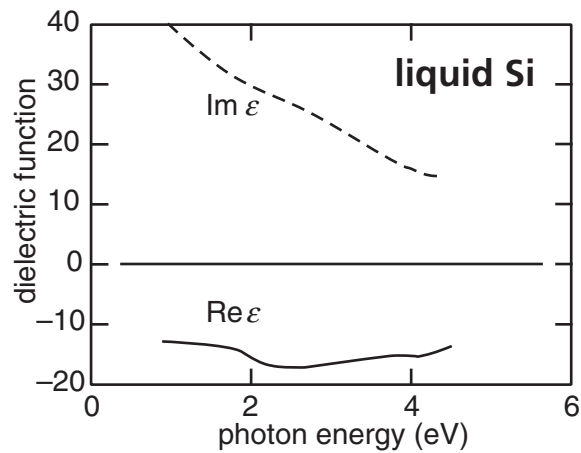
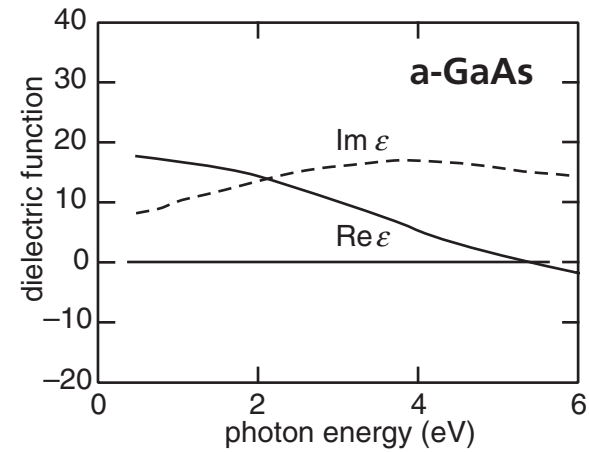
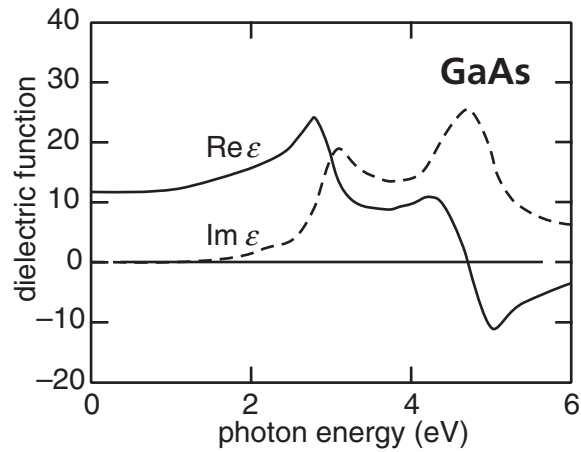
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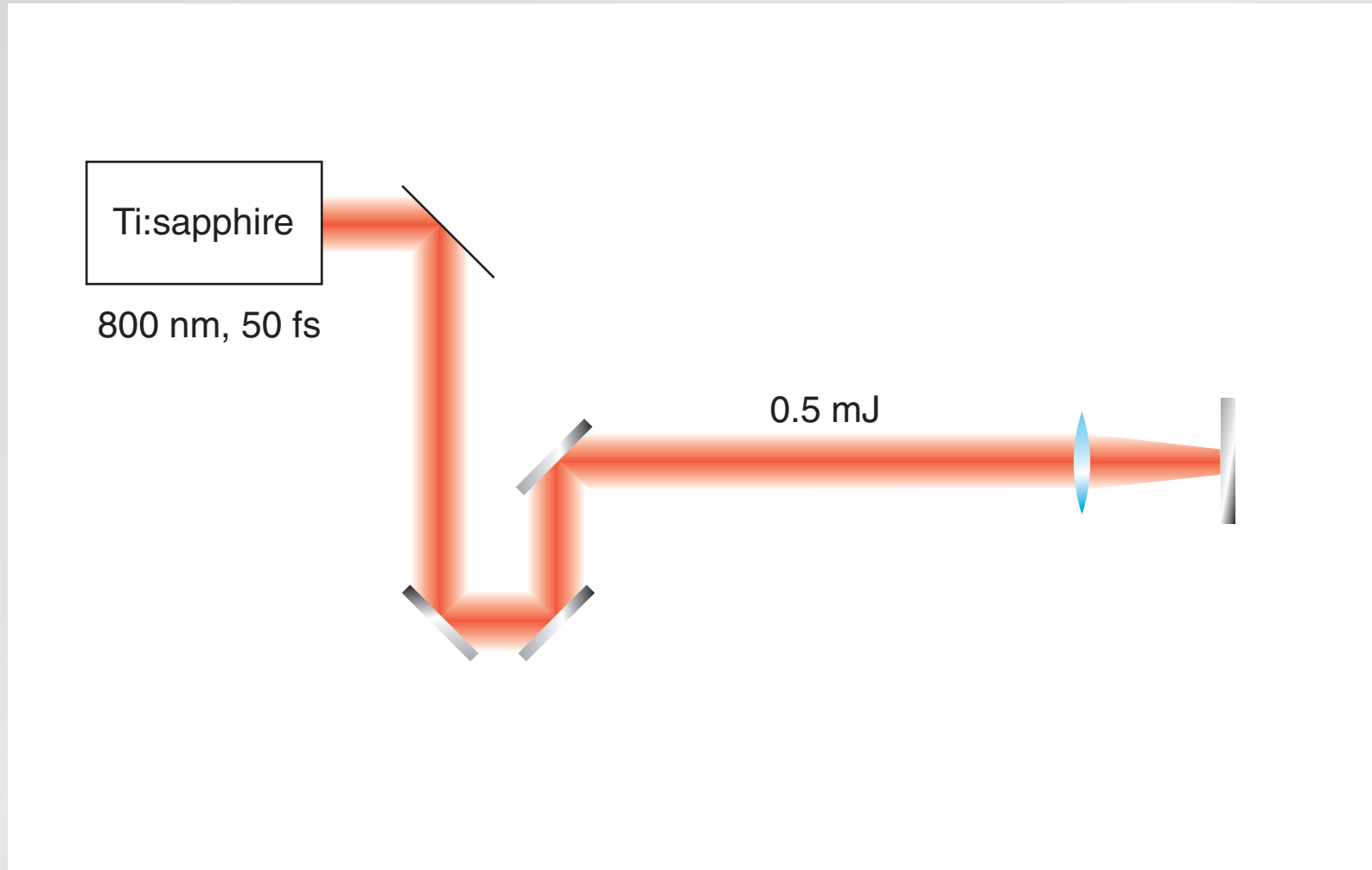


Outline

- **experimental**
- **coherent phonons**
- **optical control**

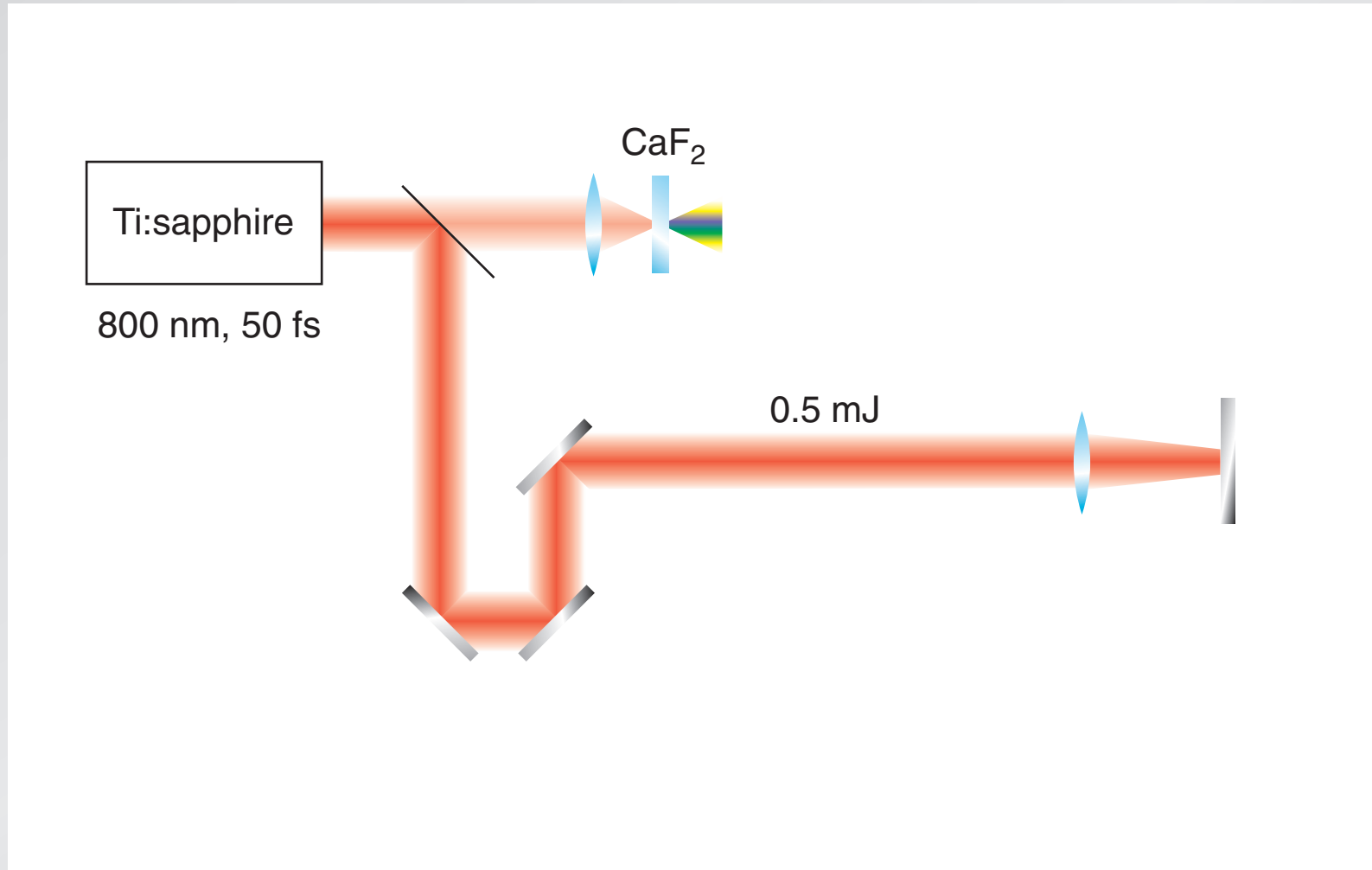
Experimental

time-resolved dual-angle reflectometry



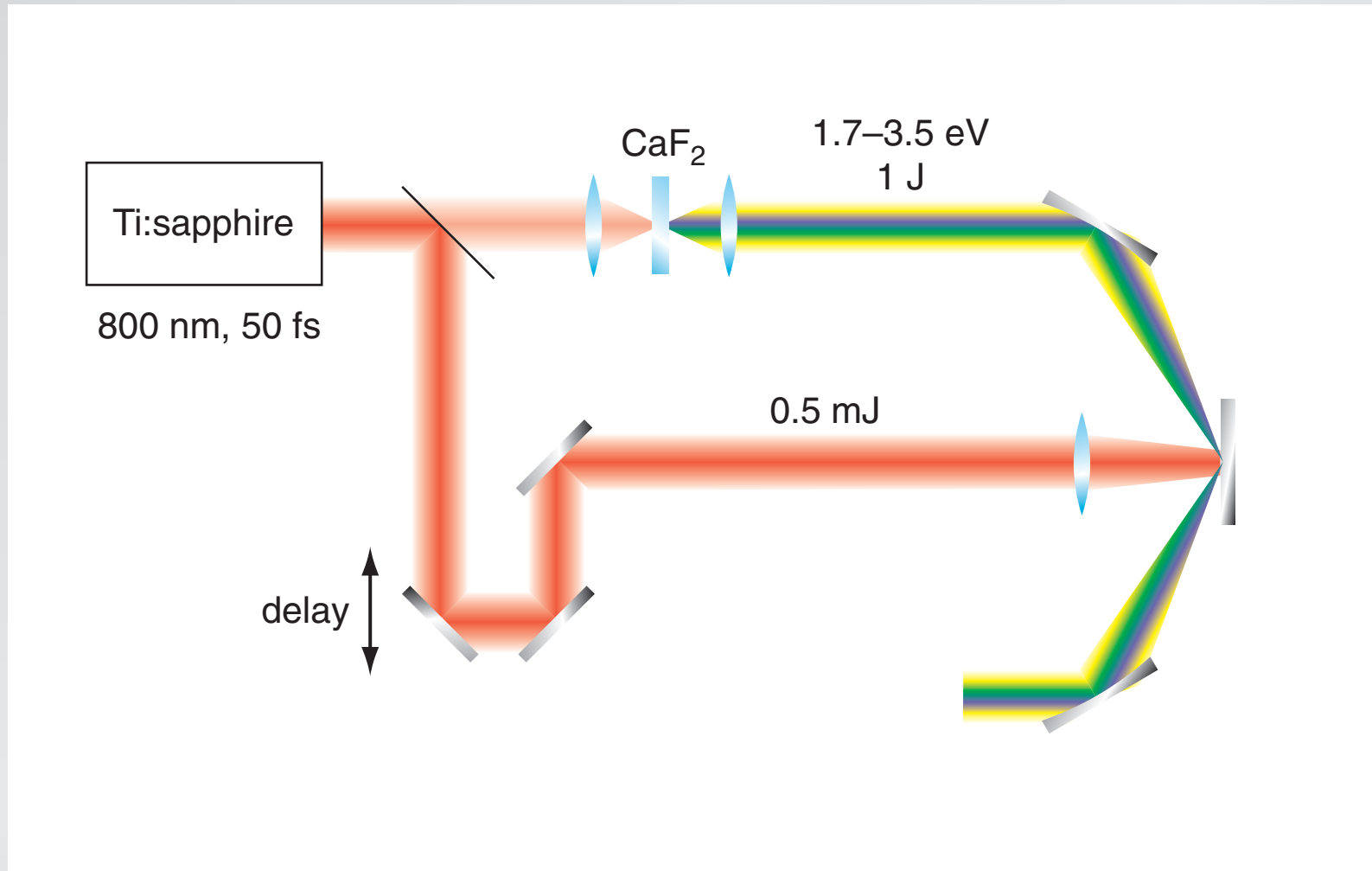
Experimental

time-resolved dual-angle reflectometry



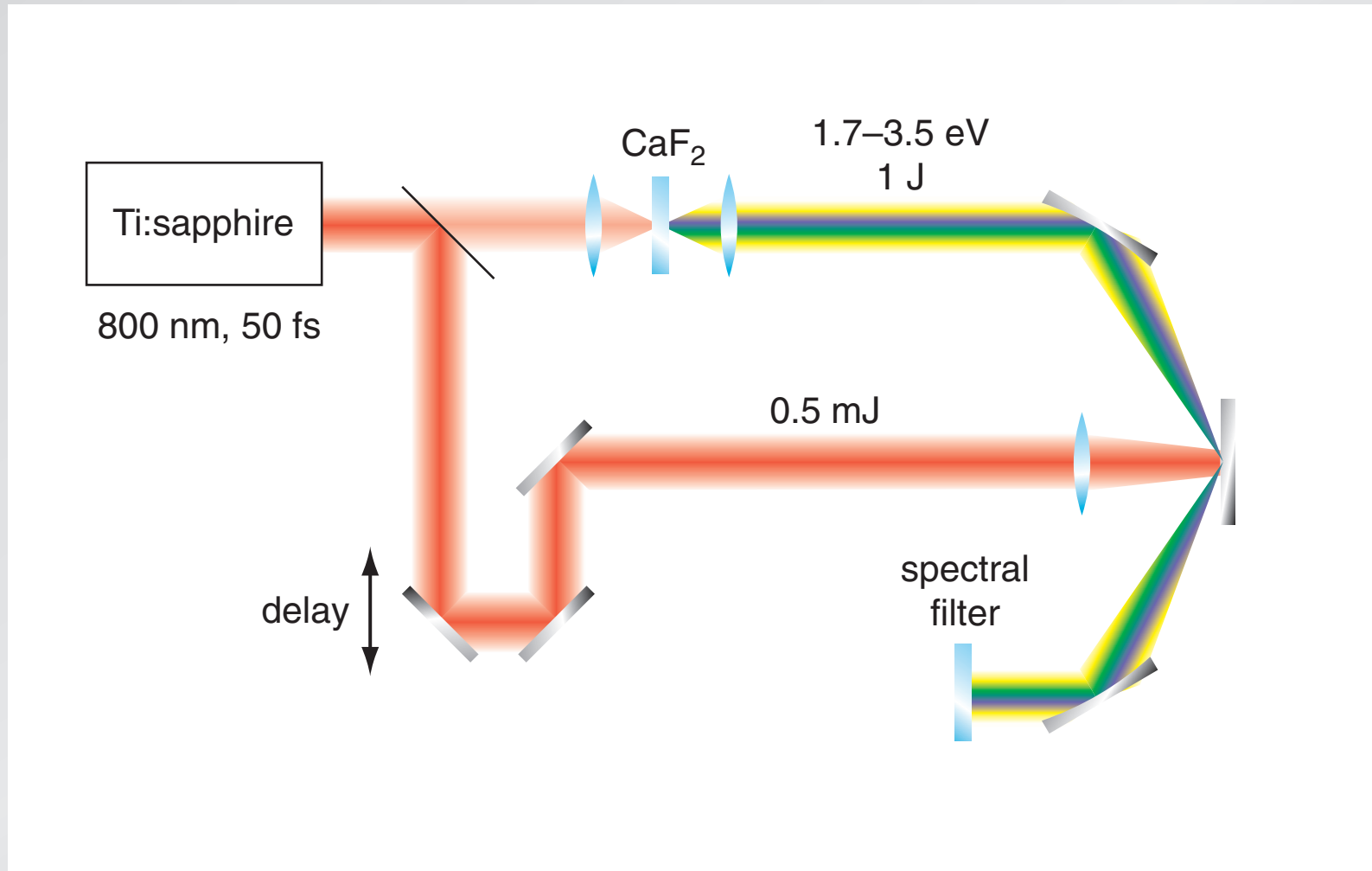
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time-resolved dual-angle reflectometry



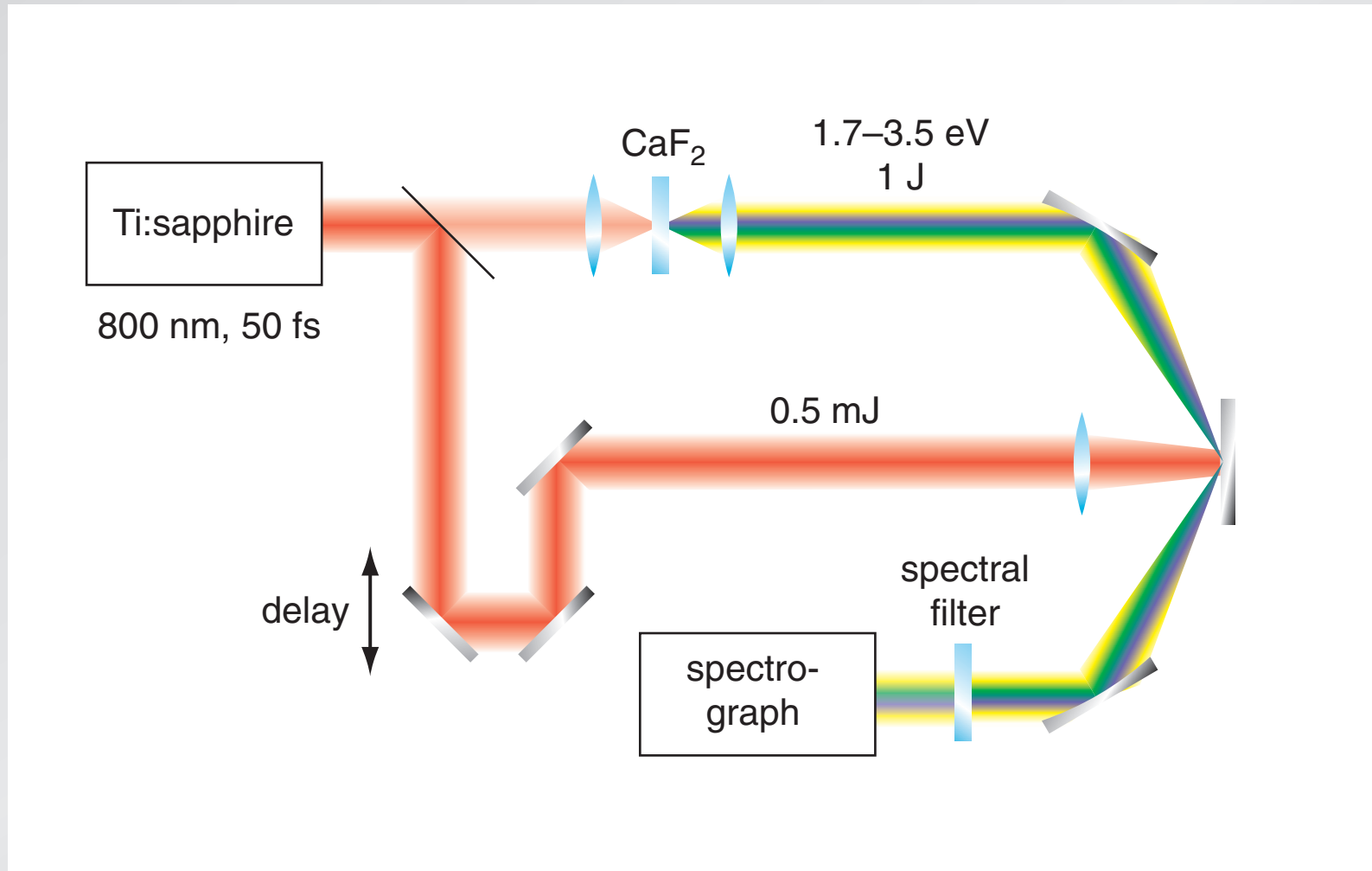
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time-resolved dual-angle reflectometry



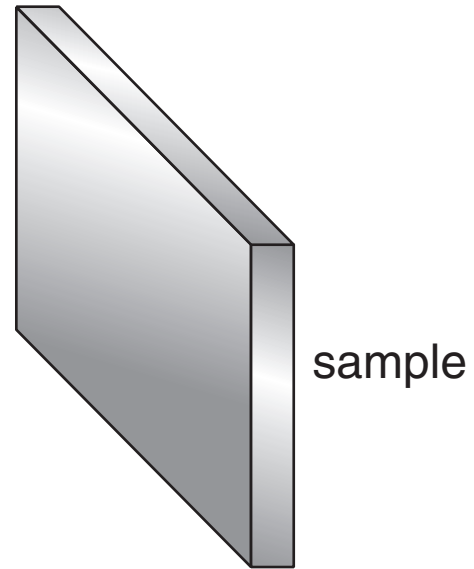
Experimental

time-resolved dual-angle reflectometry



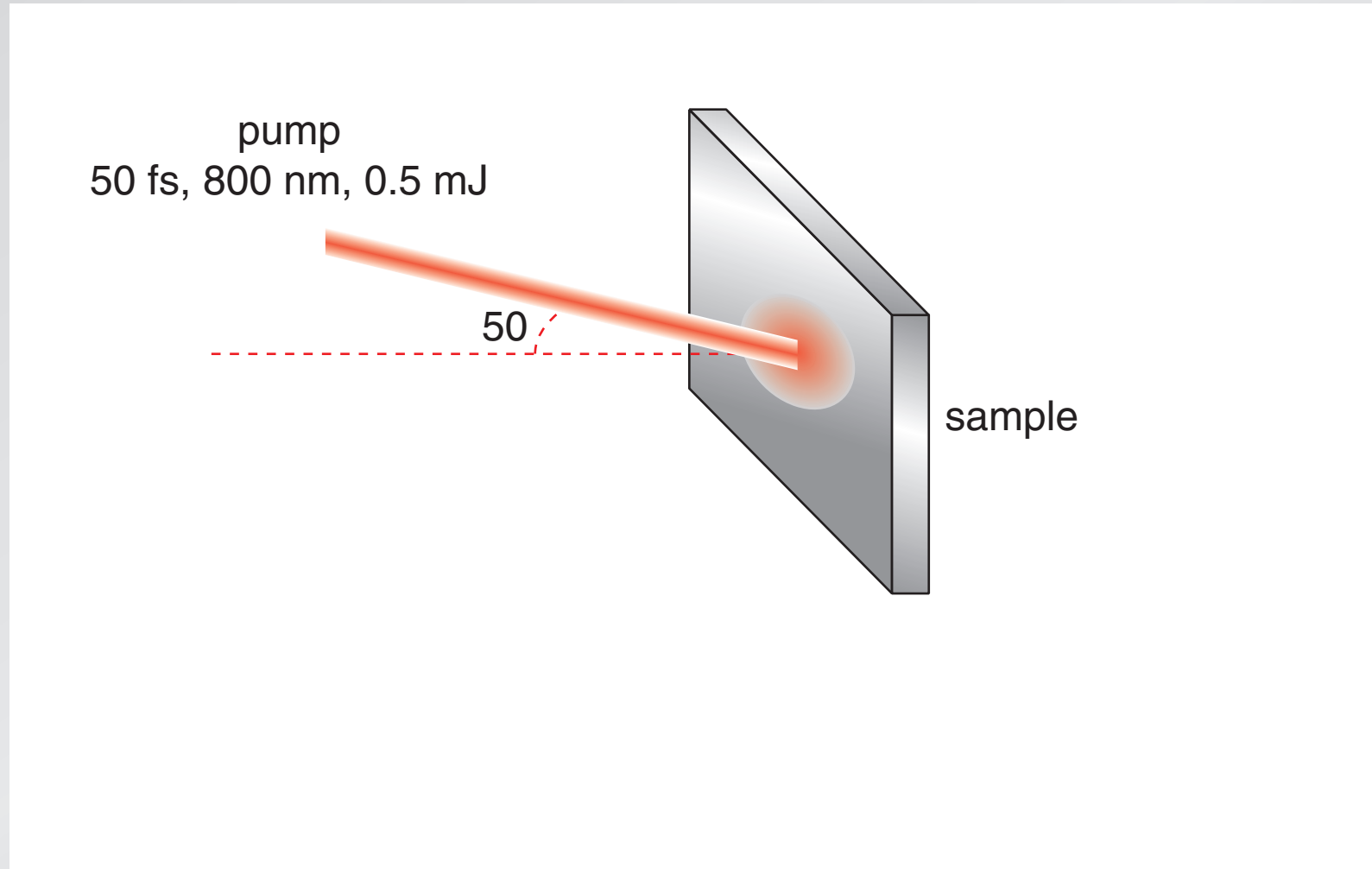
Experimental

time-resolved dual-angle reflectometry



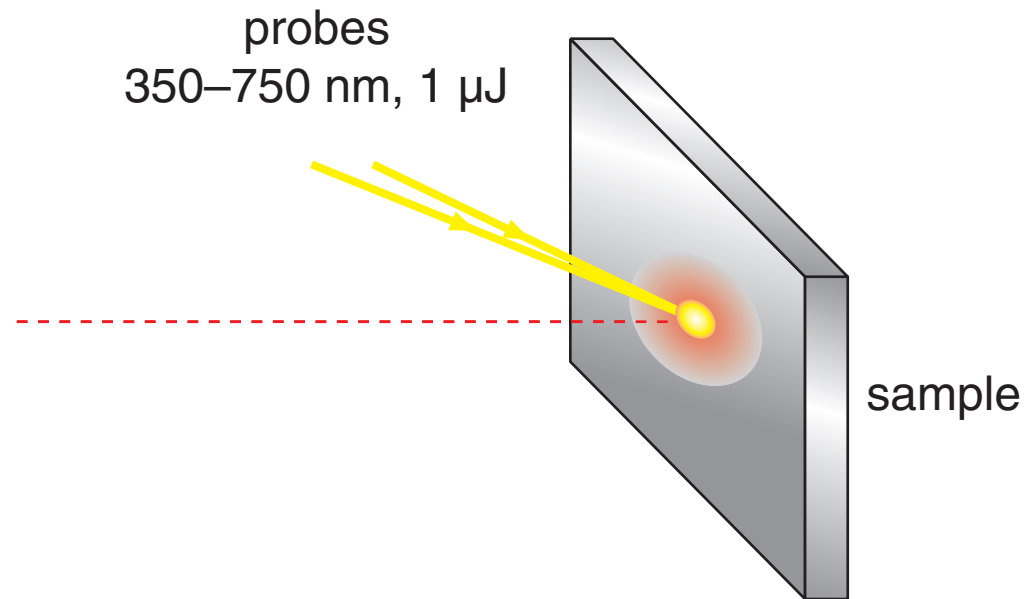
Experimental

time-resolved dual-angle reflectometry



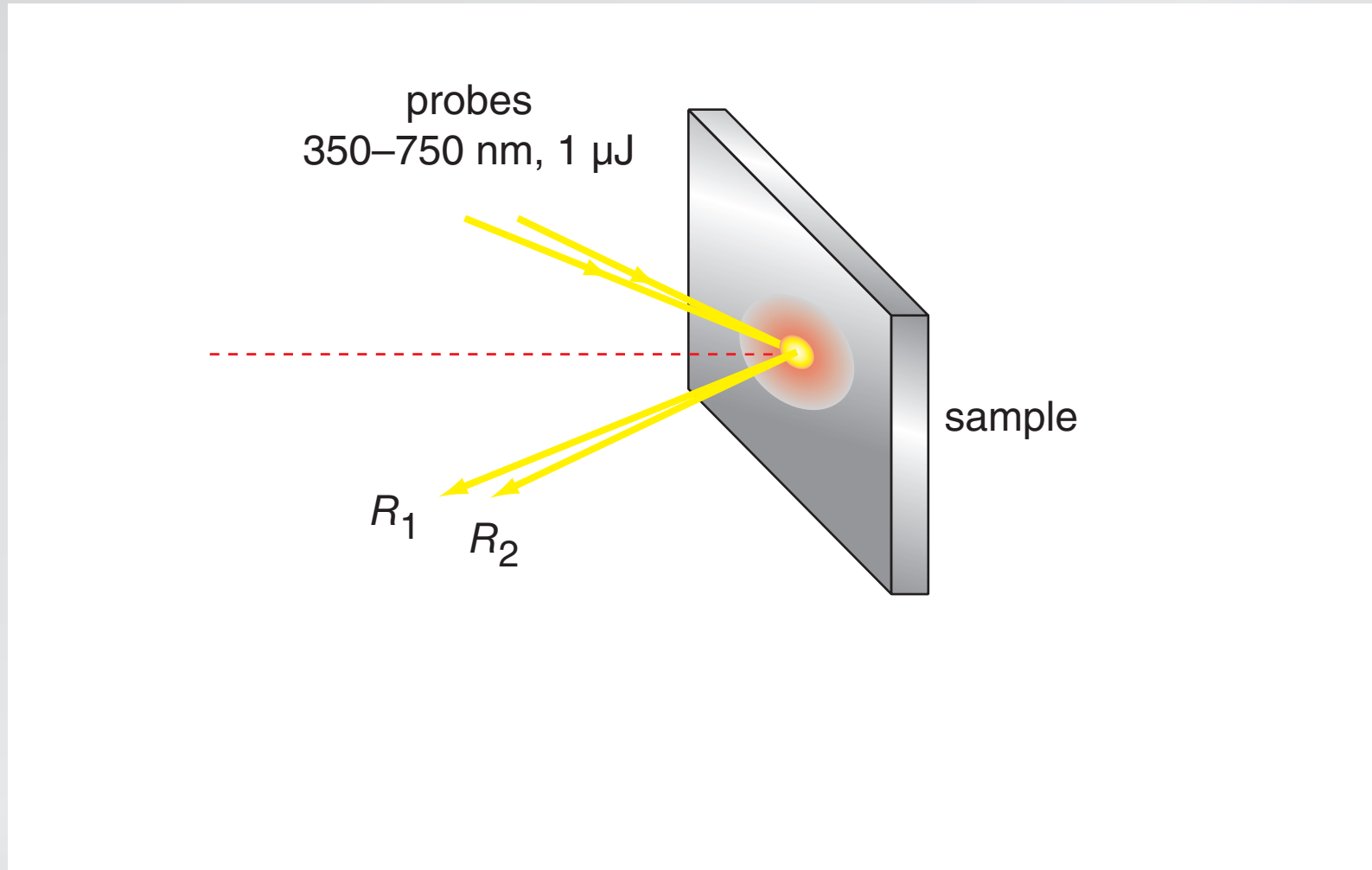
Experimental

time-resolved dual-angle reflectometry



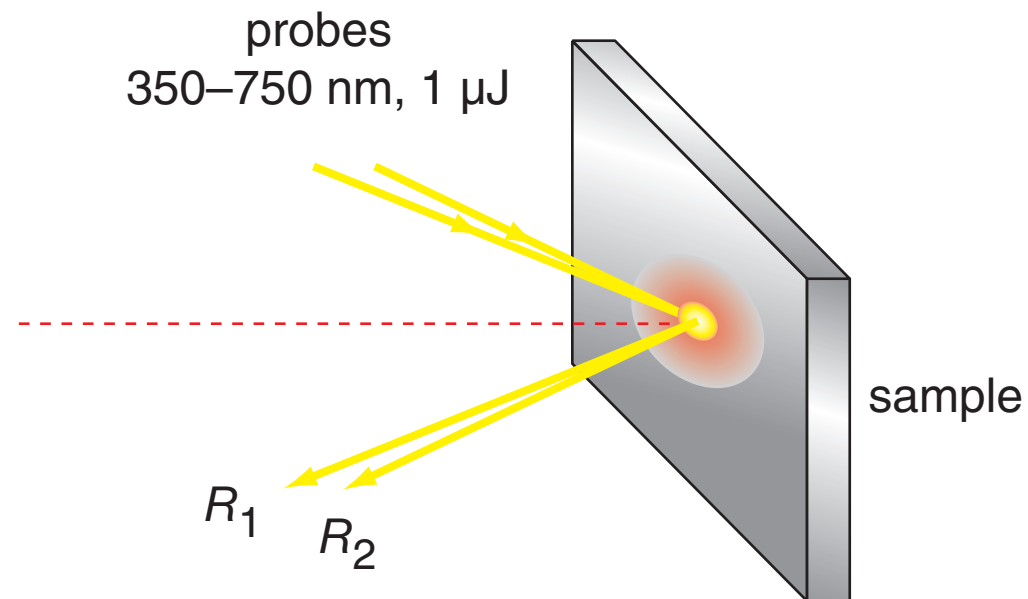
Experimental

time-resolved dual-angle reflectometry



Experimental

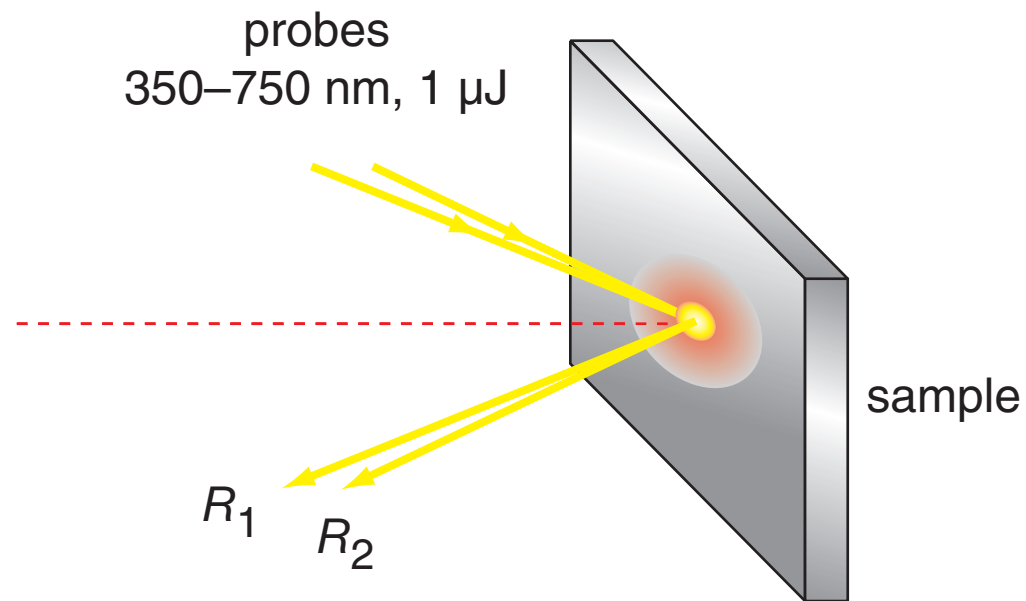
time-resolved dual-angle reflectometry



Fresnel
equations

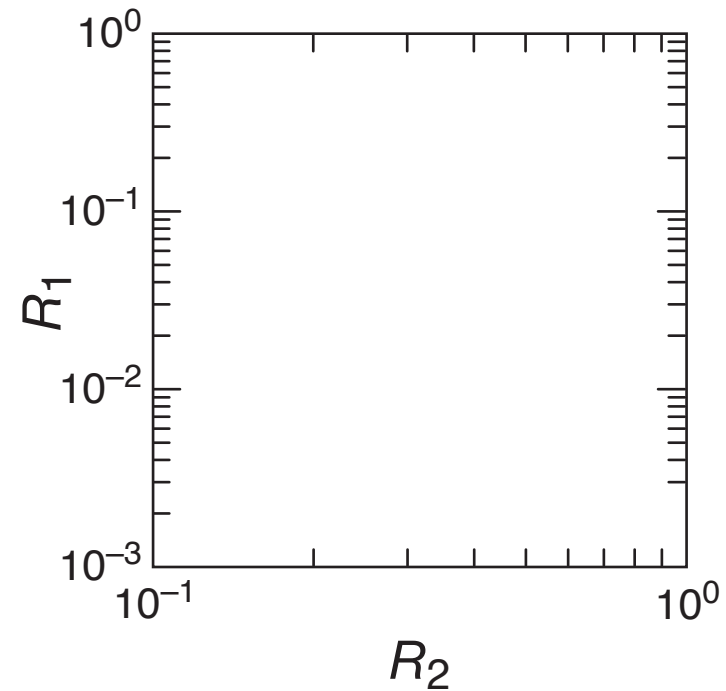
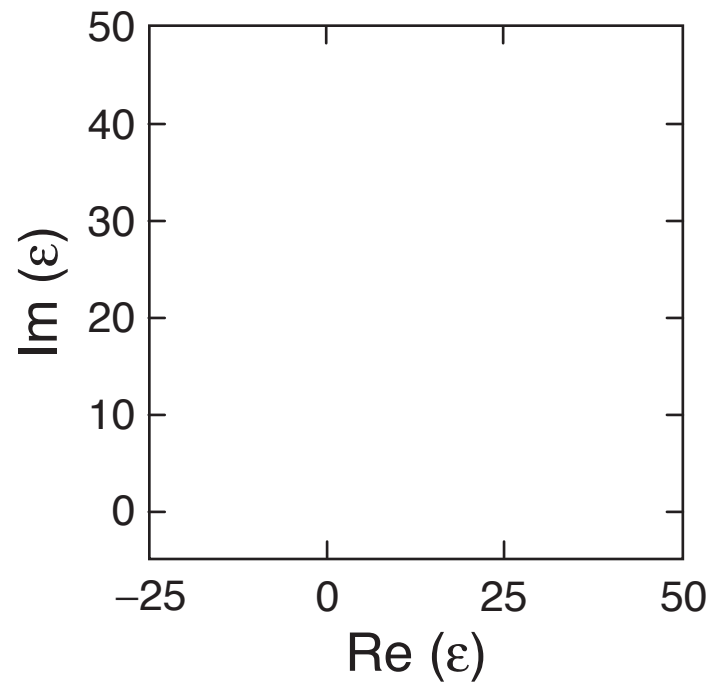
Experimental

time-resolved dual-angle reflectometry



Experimental

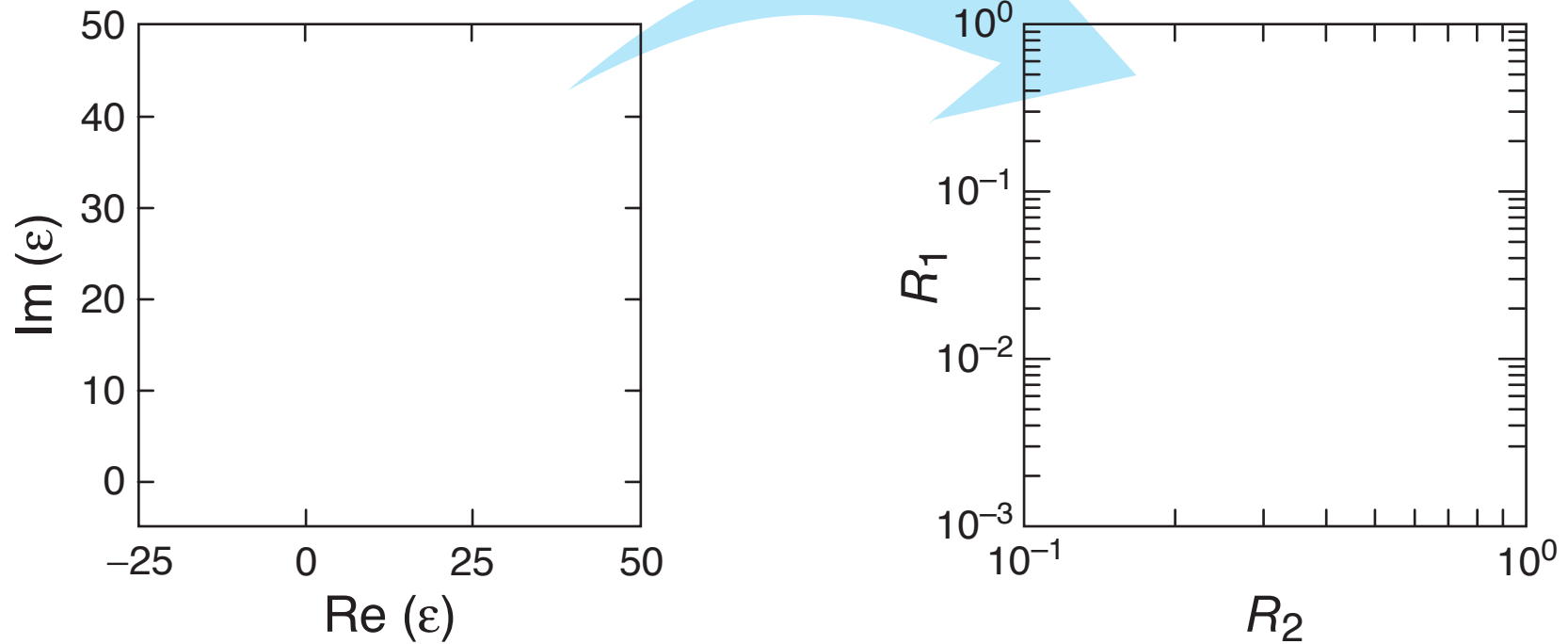
choice of angles



Fresnel equations cannot be inverted analytically

Experimental

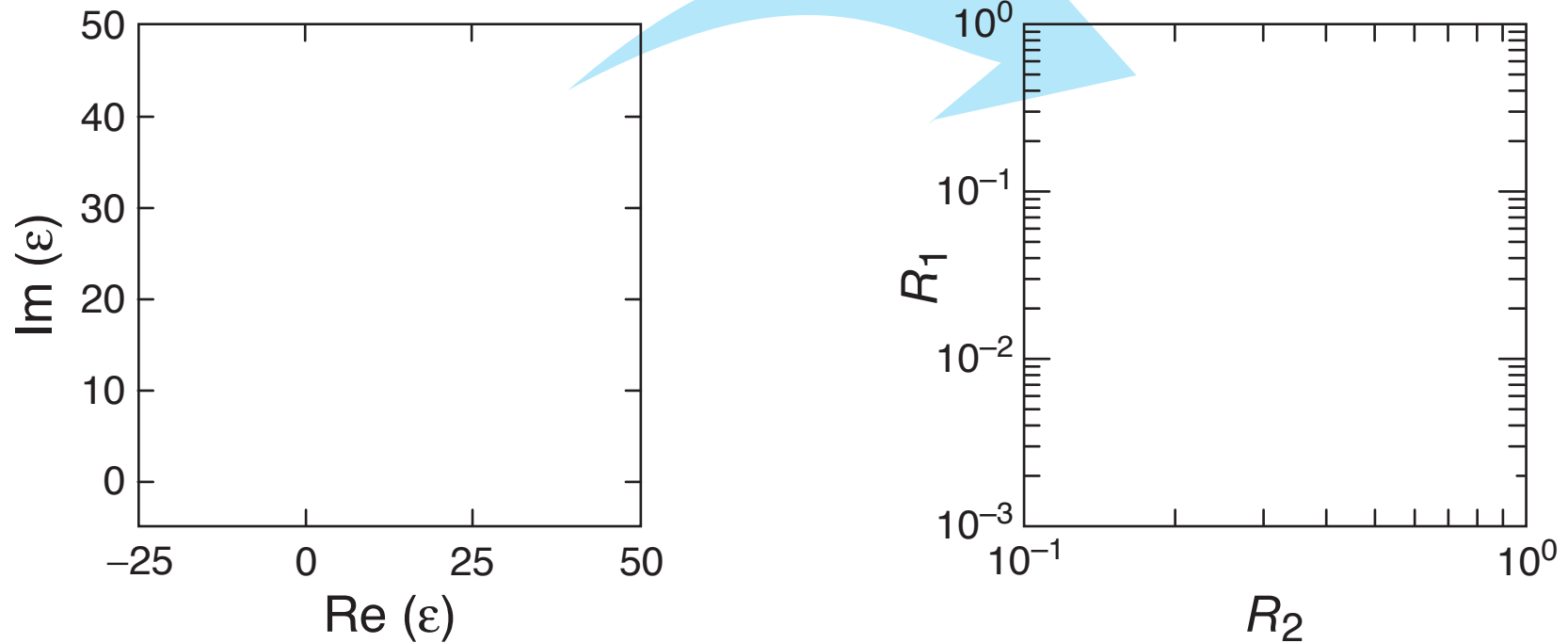
choice of angles



need numerical inversion

Experimental

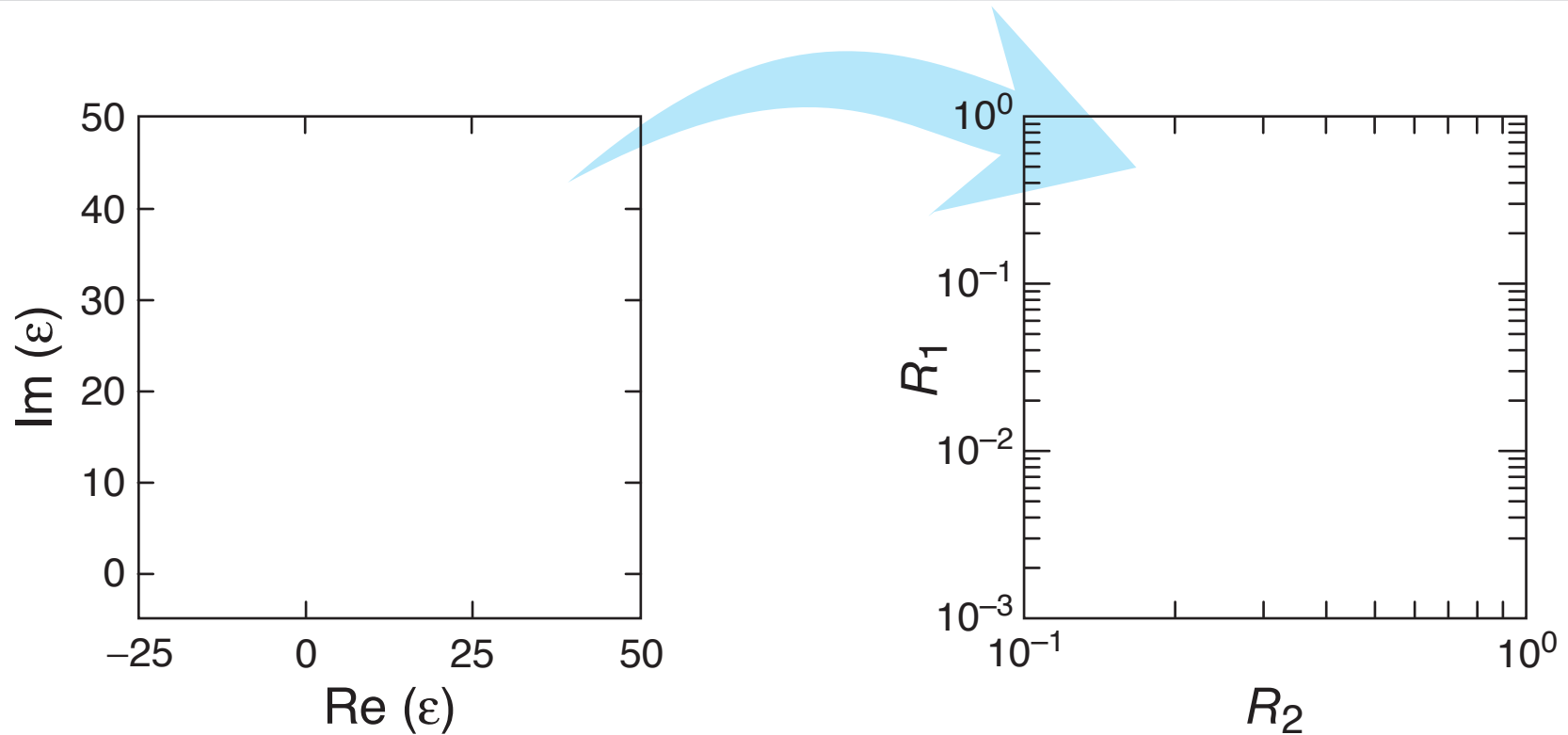
choice of angles



$R_1 = 45^\circ$ *p*-pol, $R_2 = 45^\circ$ *s*-pol

Experimental

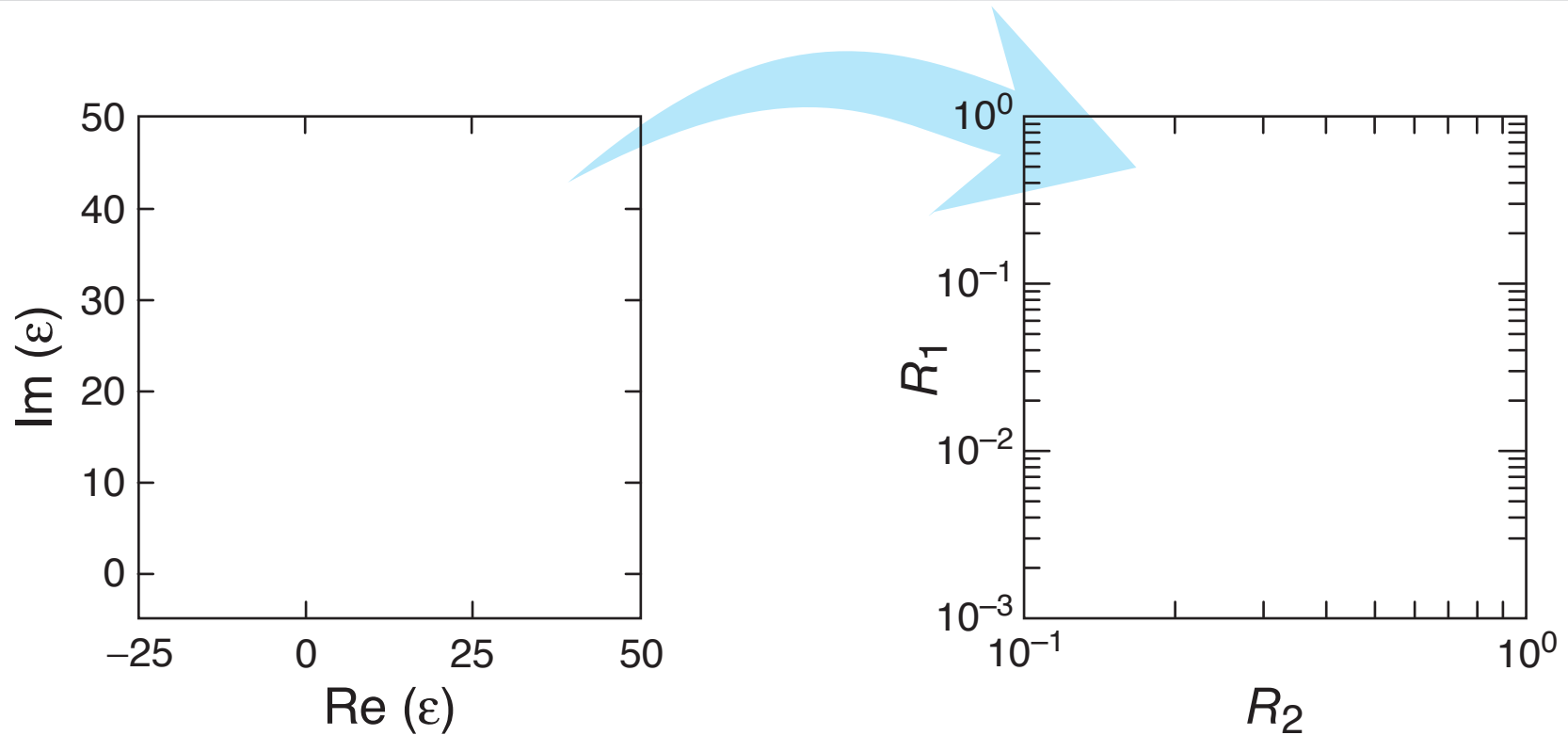
choice of angles



$R_1 = 60^\circ$ *p*-pol, $R_2 = 45^\circ$ *p*-pol

Experimental

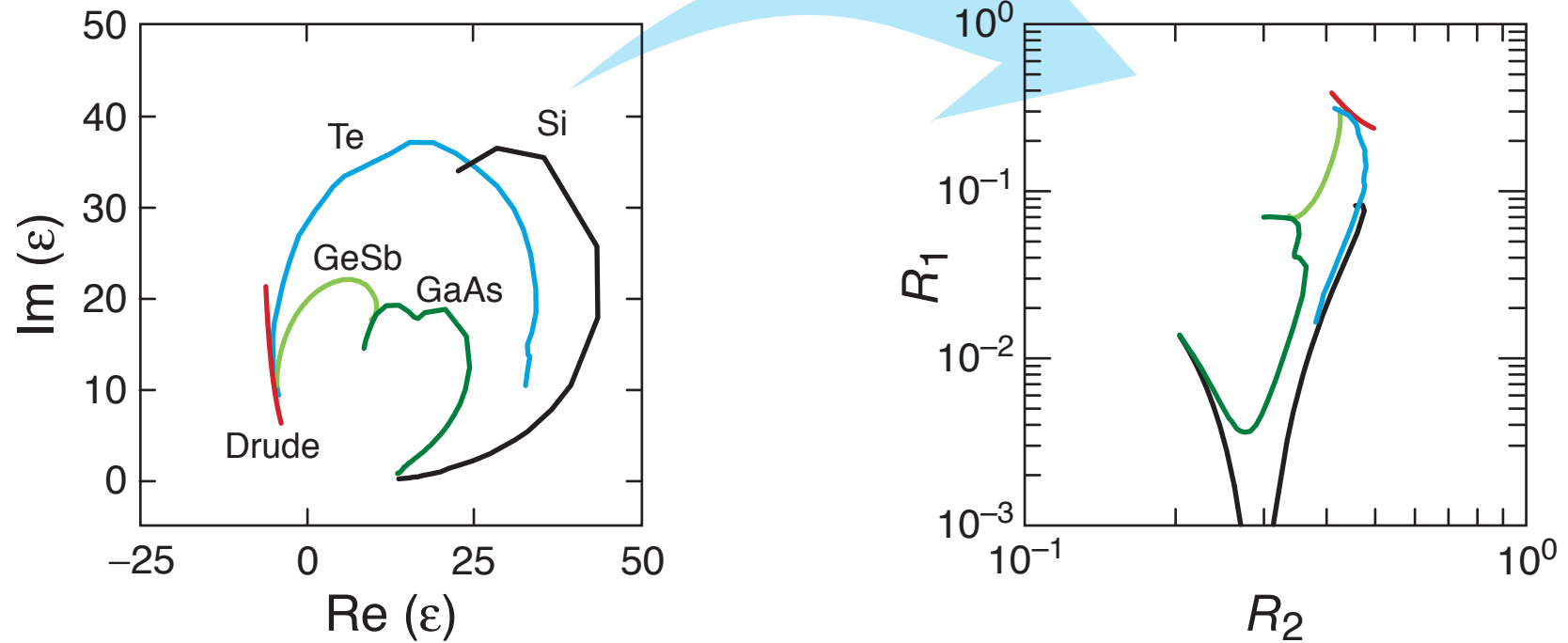
choice of angles



$R_1 = 78^\circ$ *p*-pol, $R_2 = 45^\circ$ *p*-pol

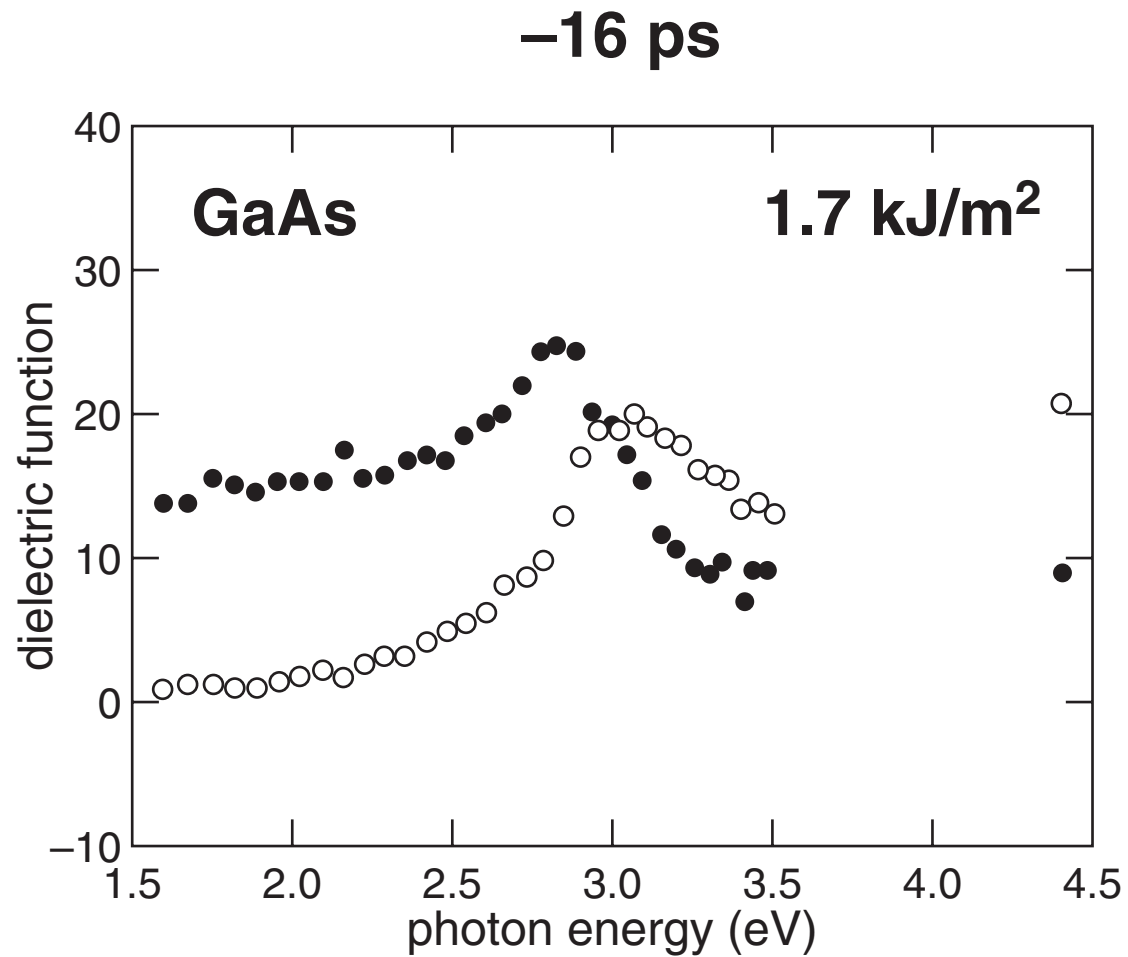
Experimental

choice of angles

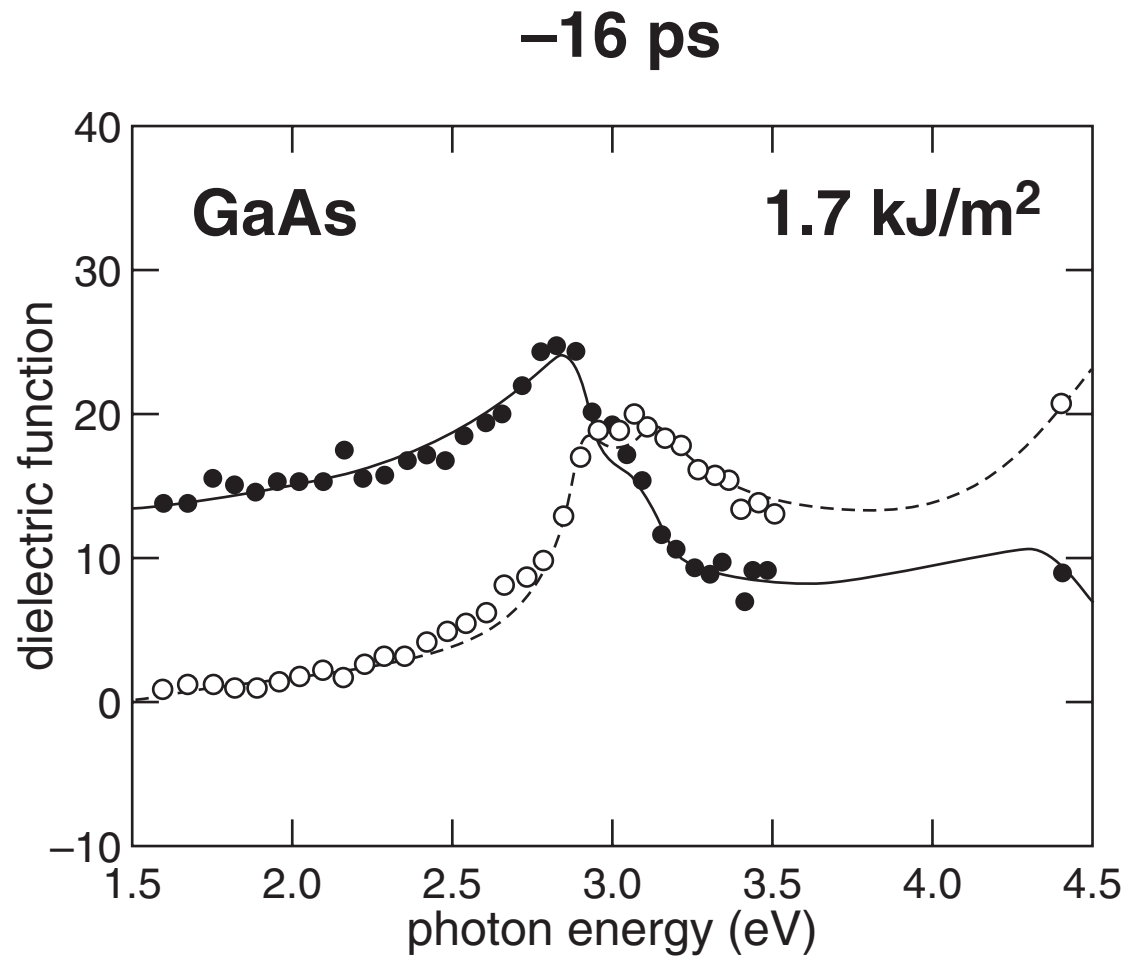


$R_1 = 78^\circ$ p-pol, $R_2 = 45^\circ$ p-pol

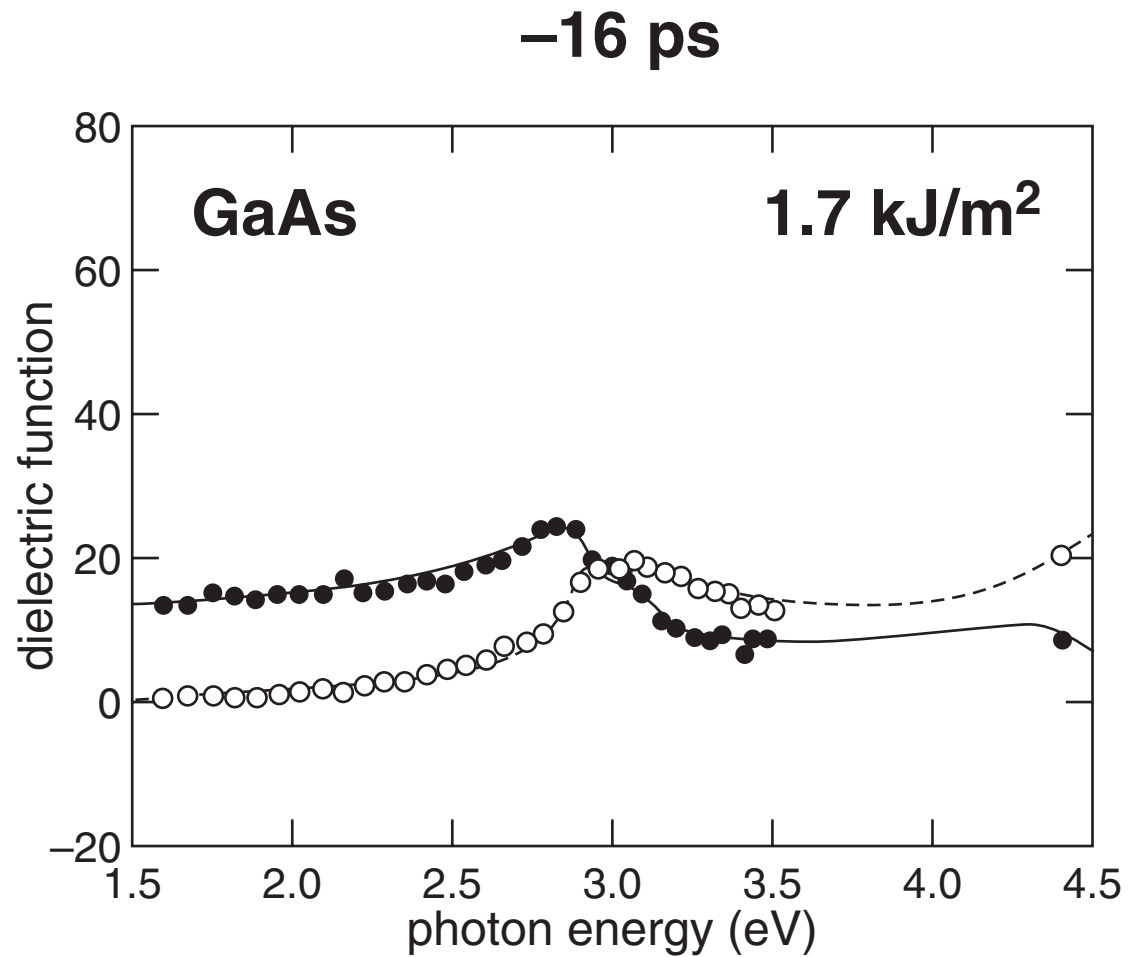
Experimental



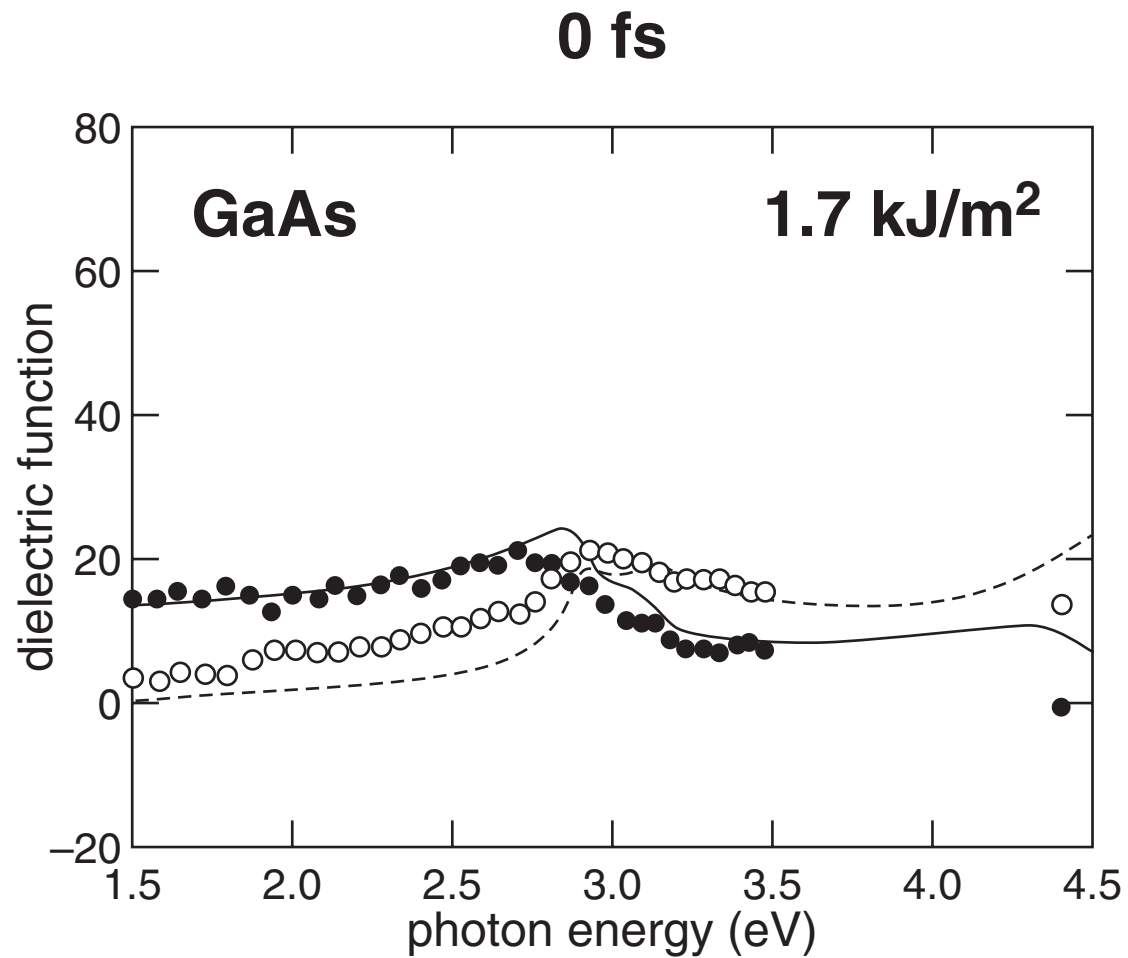
Experimental



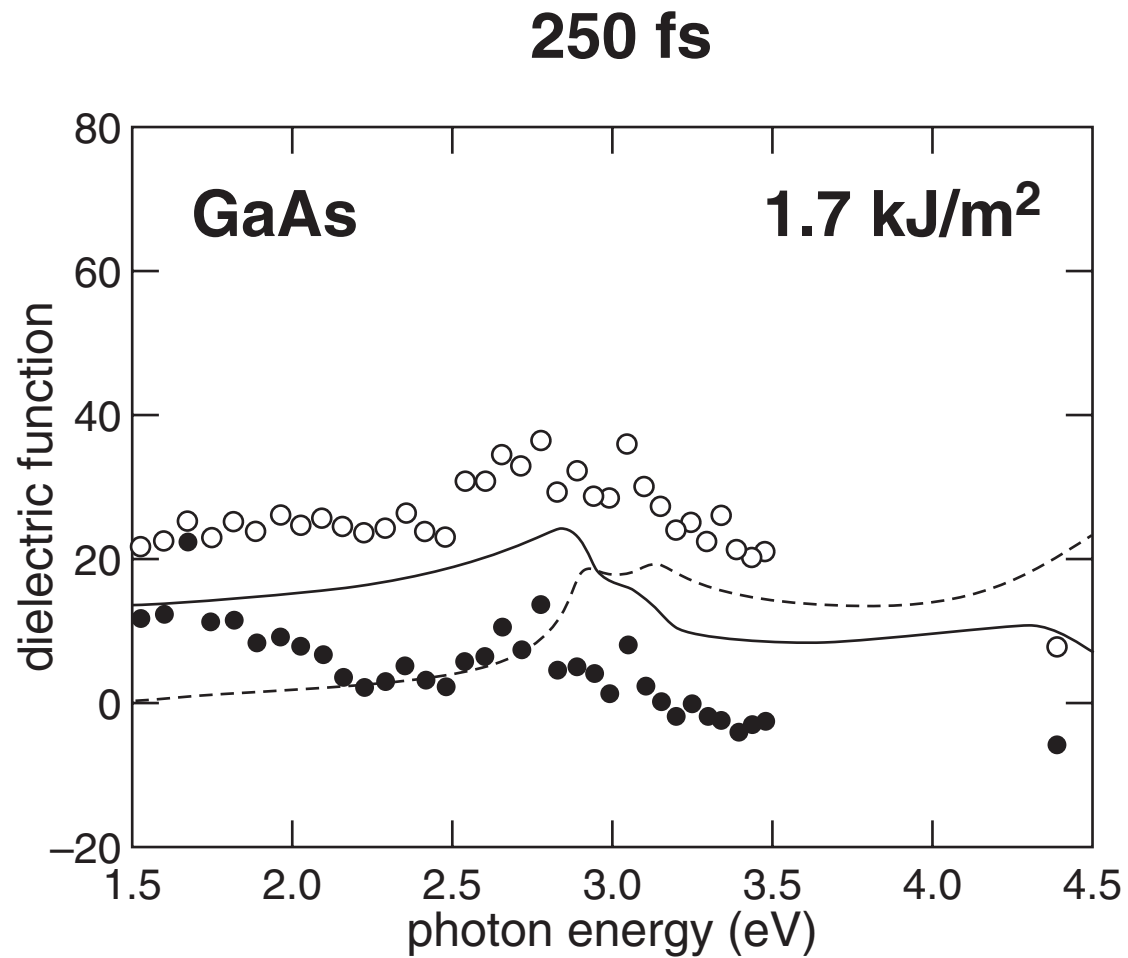
Experimental



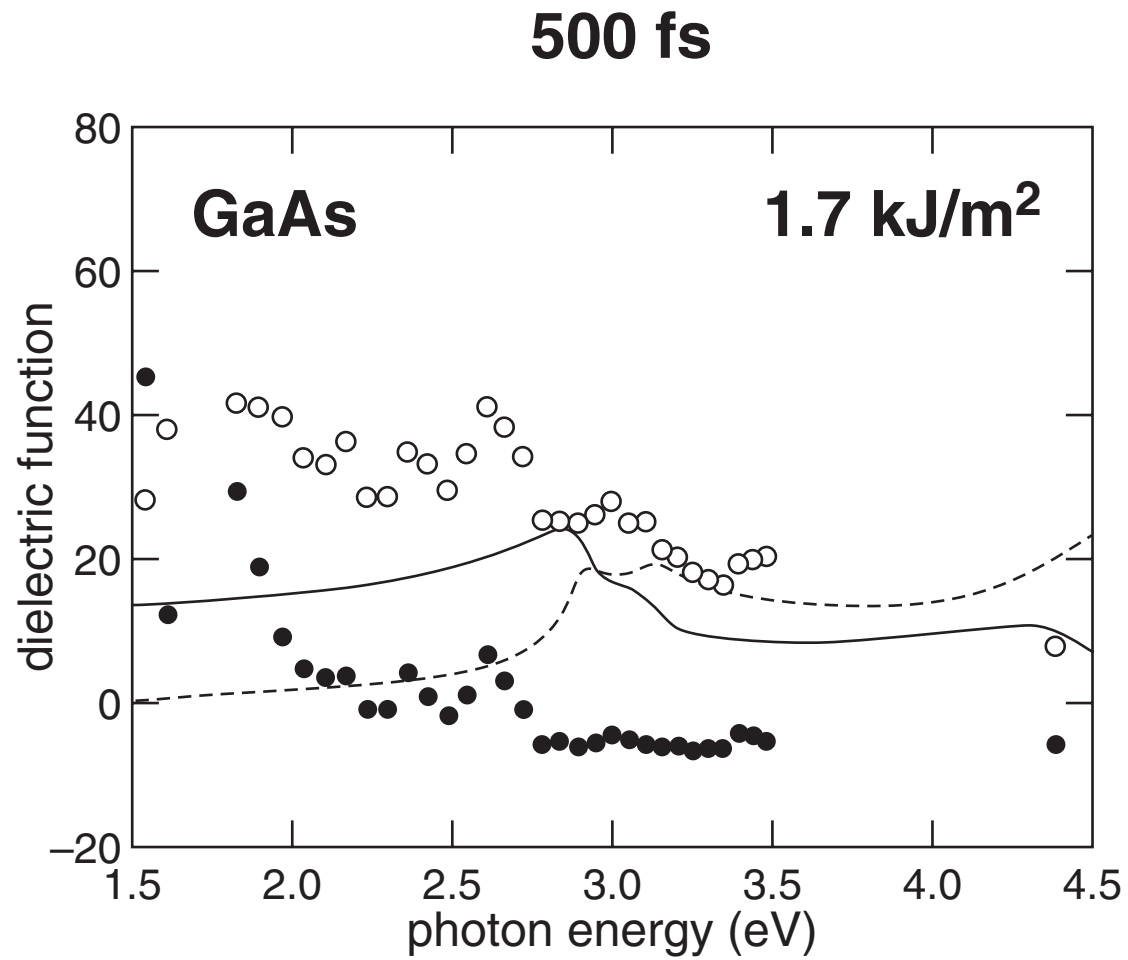
Experimental



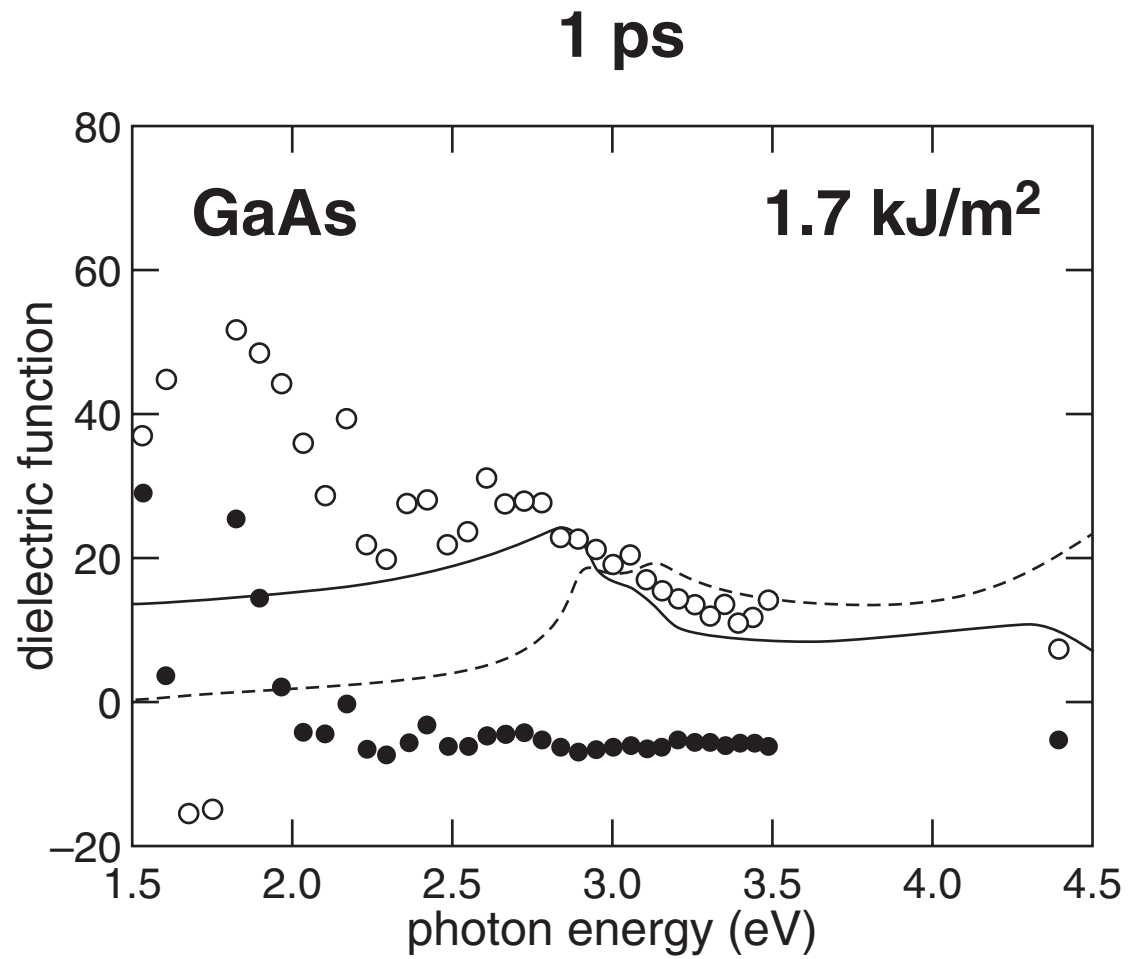
Experimental



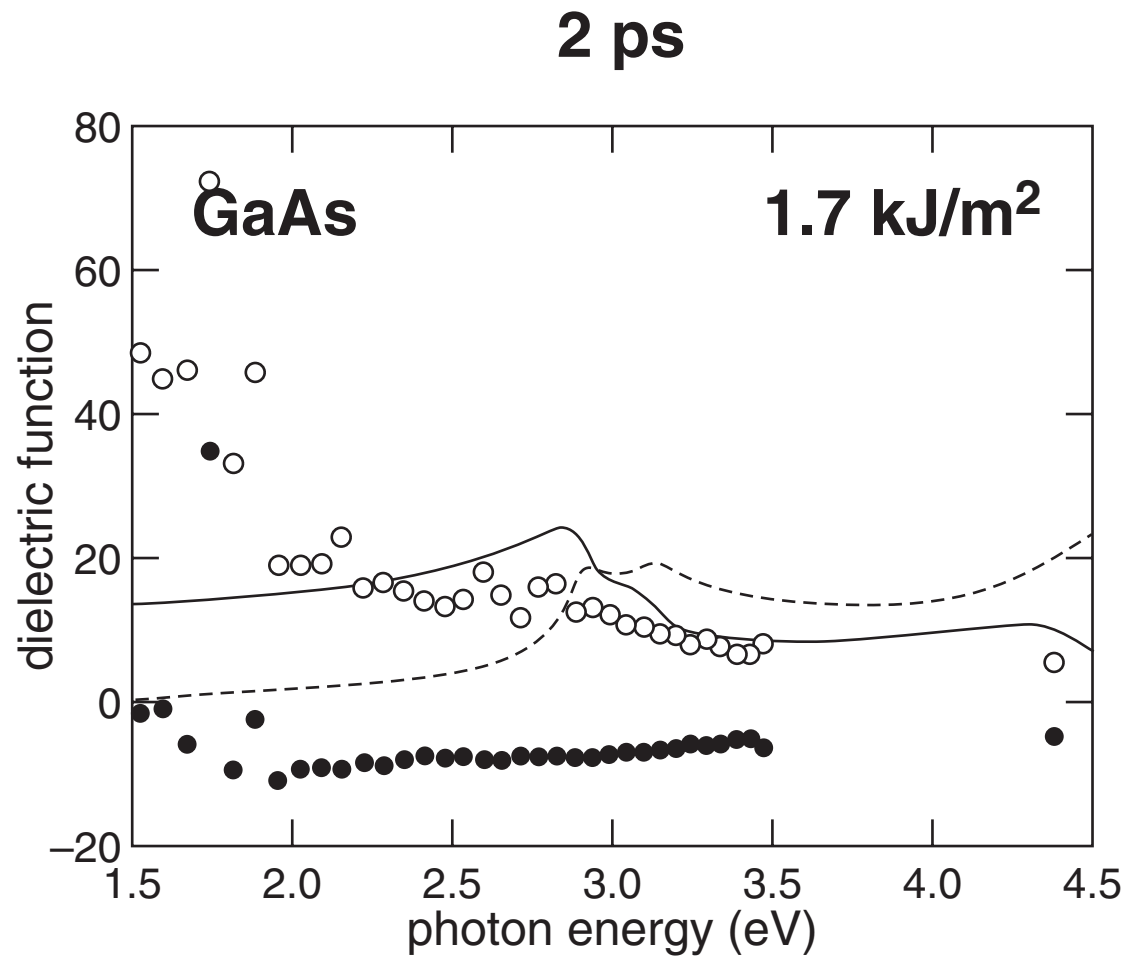
Experimental



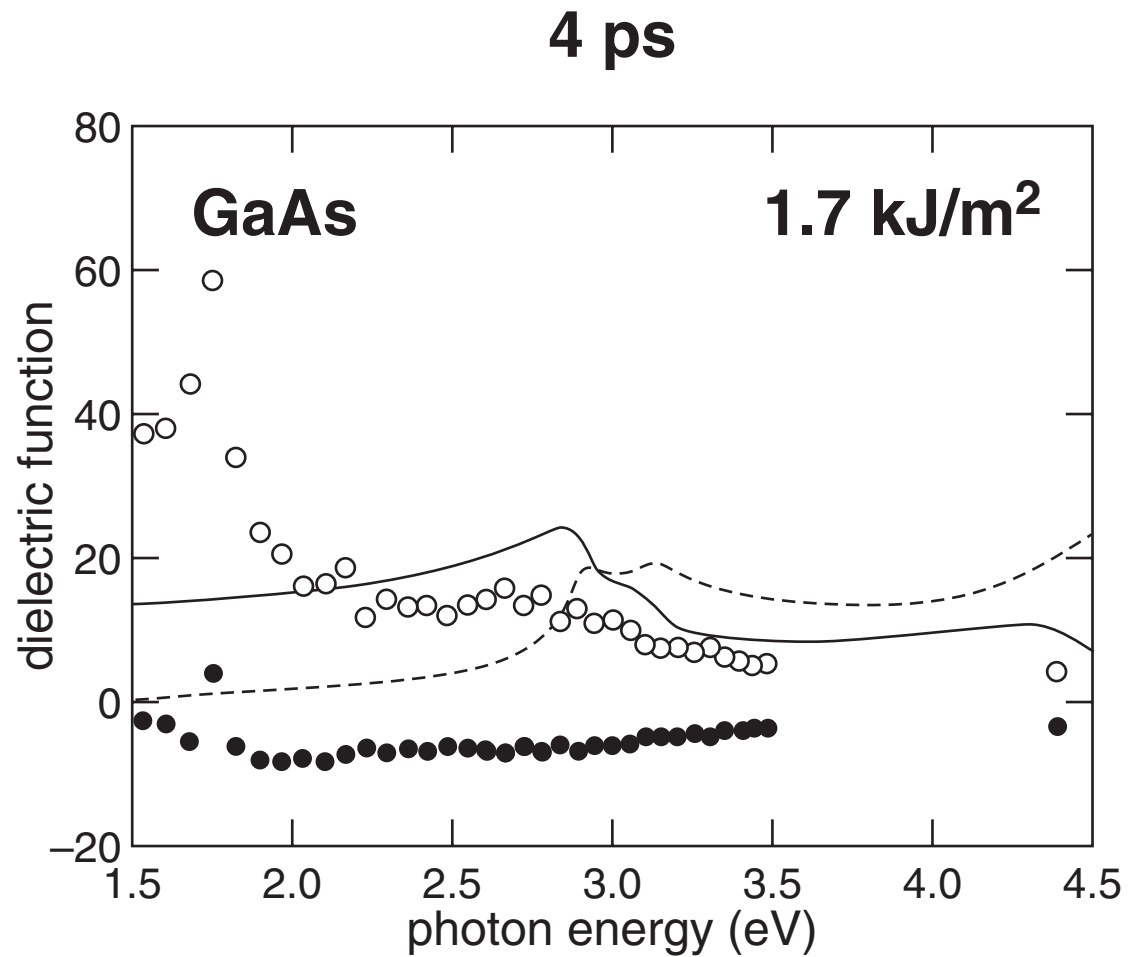
Experimental



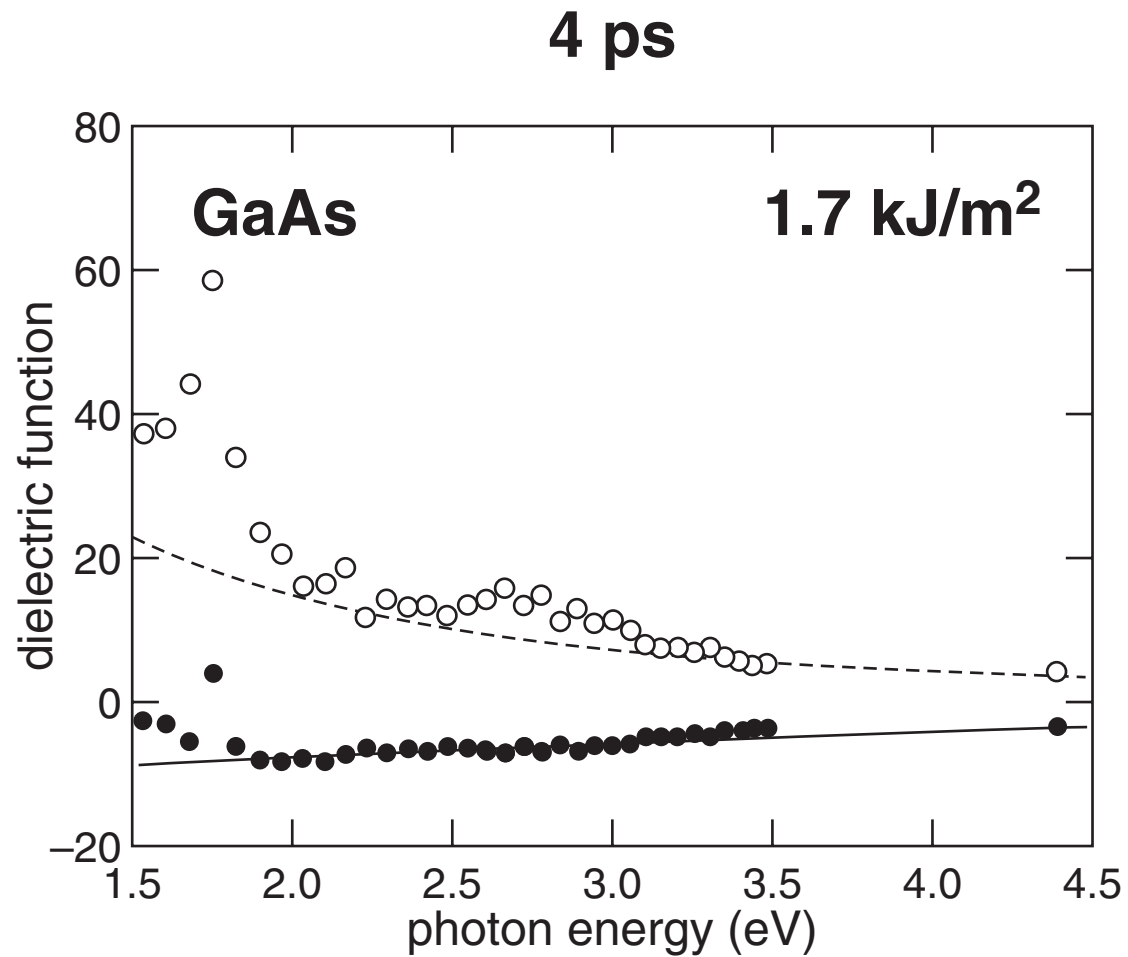
Experimental



Experimental



Experimental



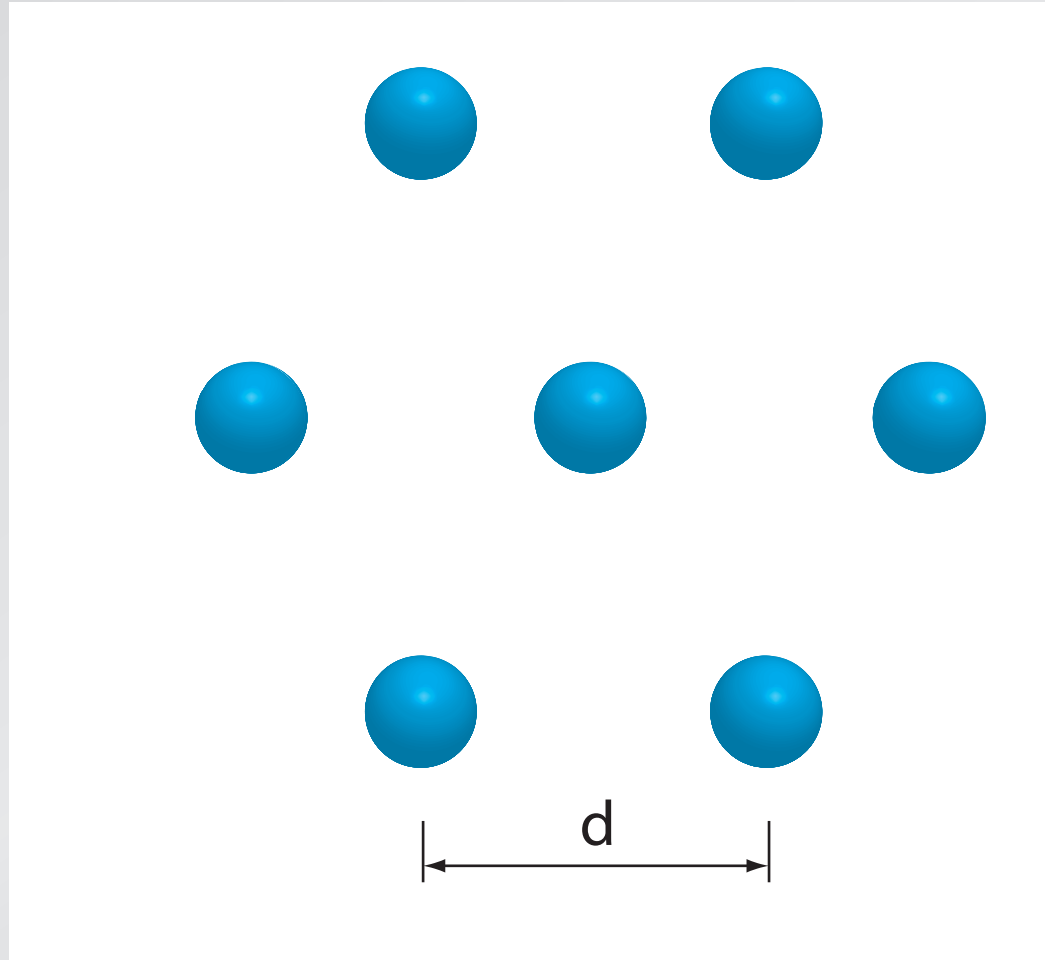
Experimental

can observe dielectric to metallic transition

Outline

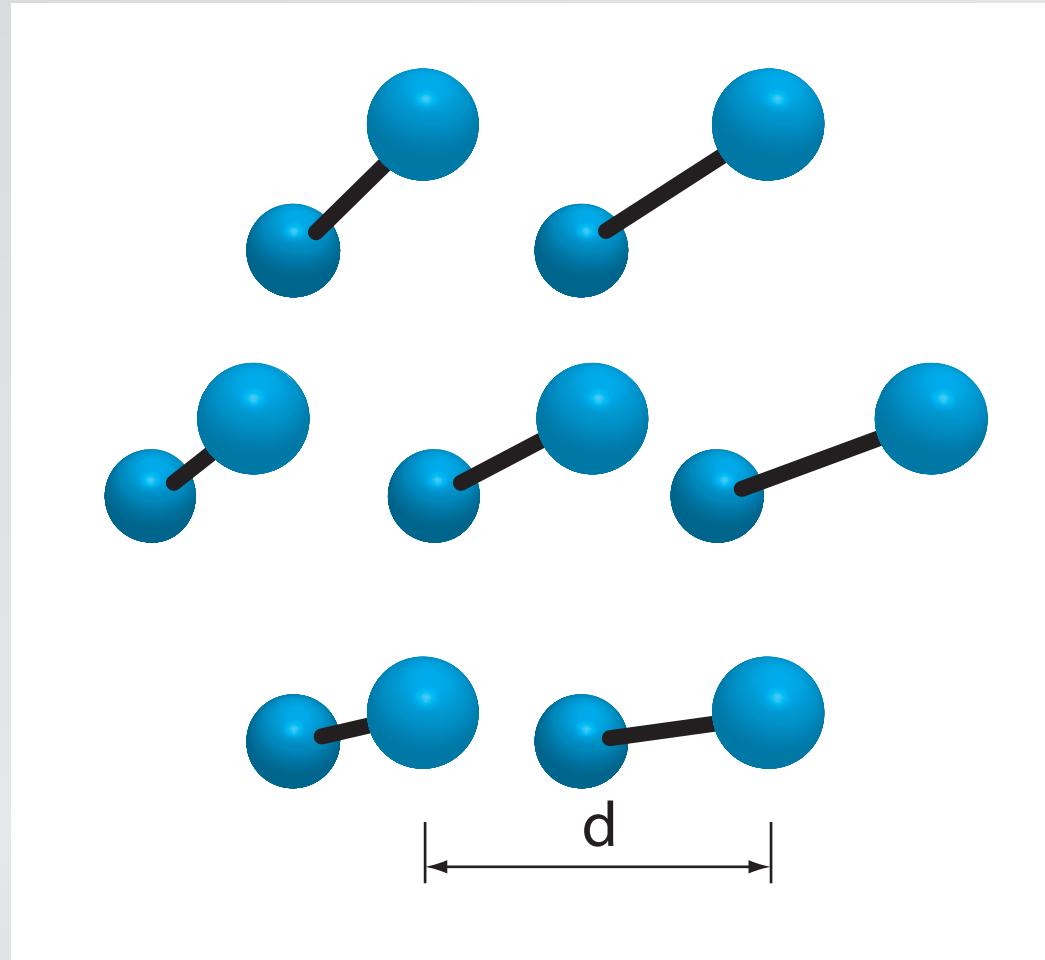
- experimental
- **coherent phonons**
- optical control

Coherent phonons



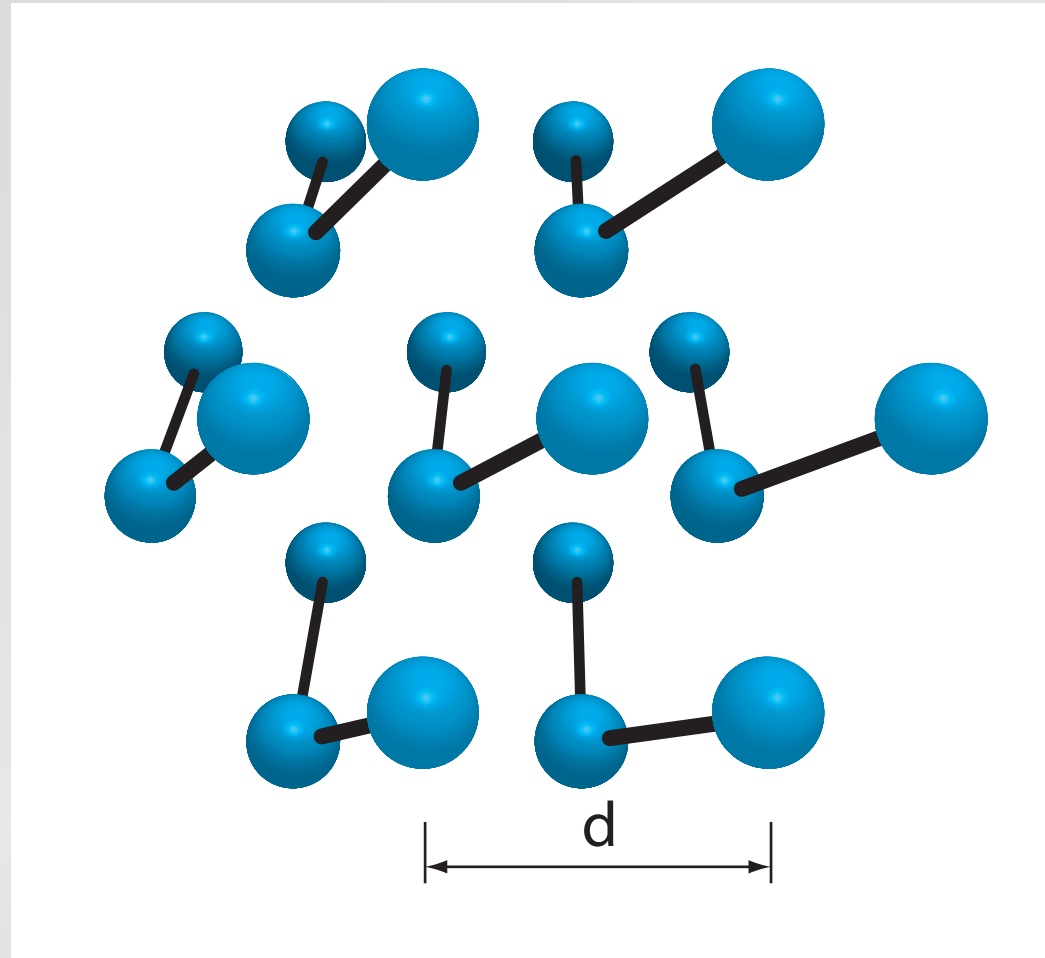
tellurium has hexagonal arrangement

Coherent phonons



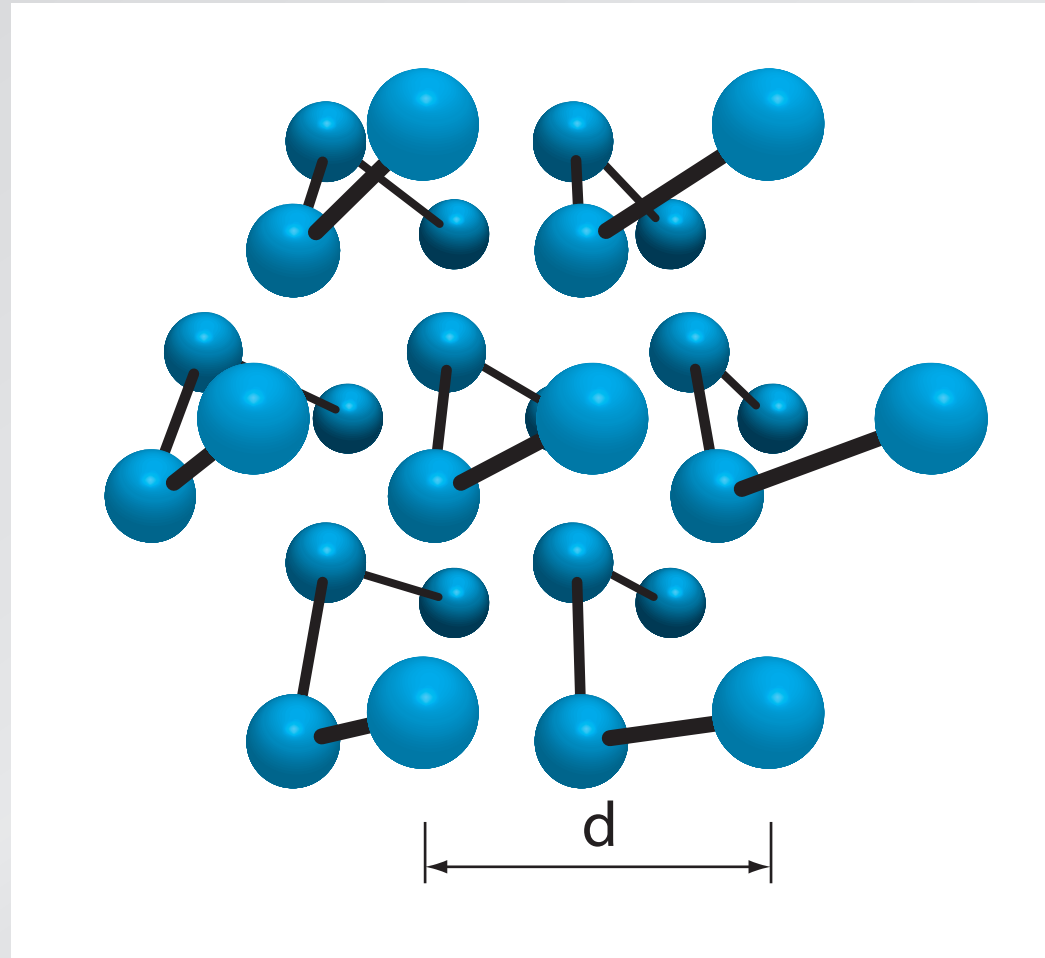
with lattice planes offset in spiral fashion

Coherent phonons



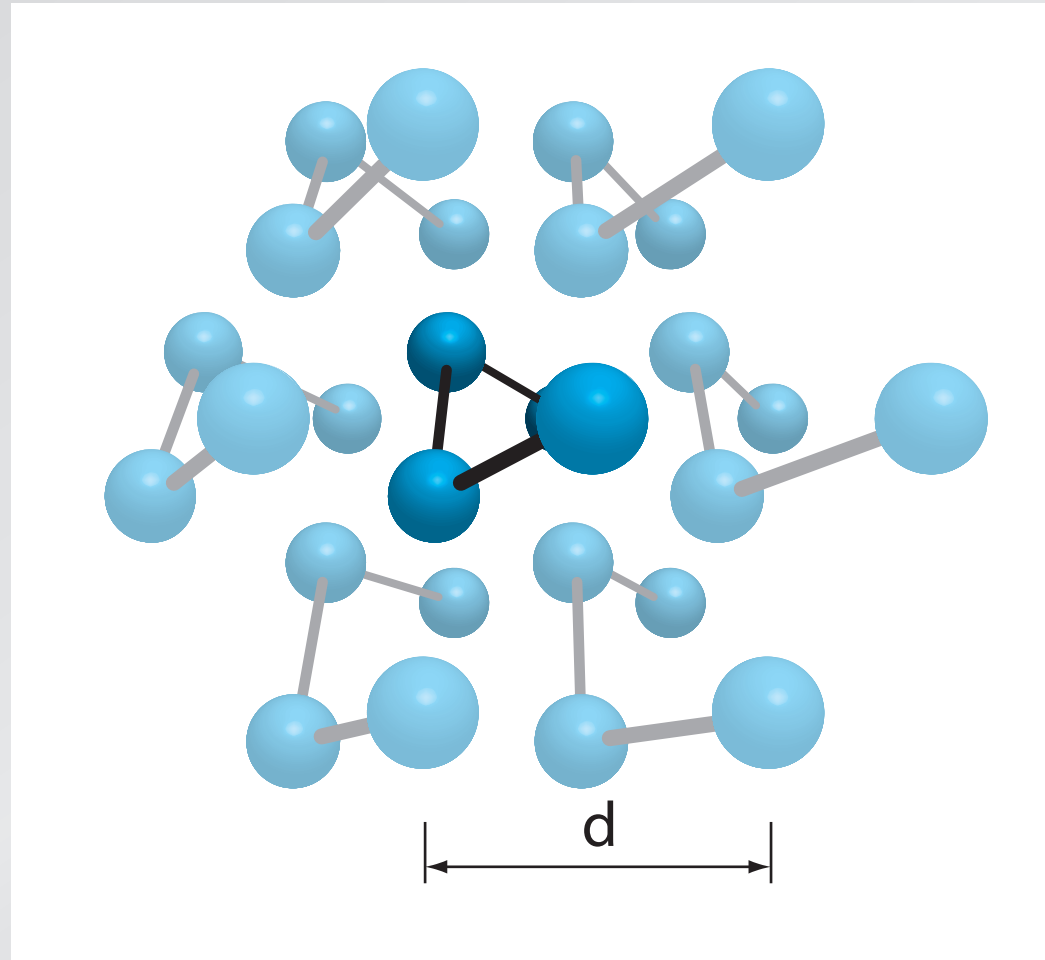
with lattice planes offset in spiral fashion

Coherent phonons



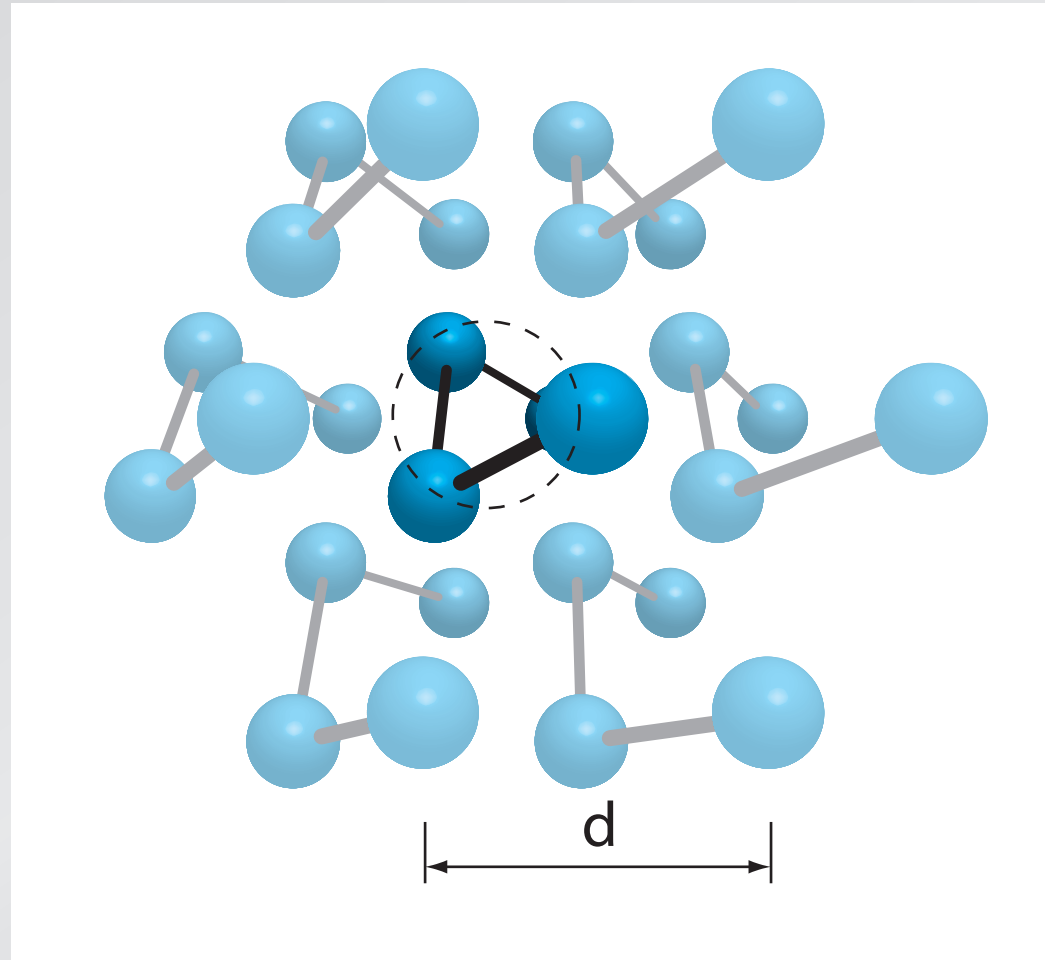
with lattice planes offset in spiral fashion

Coherent phonons



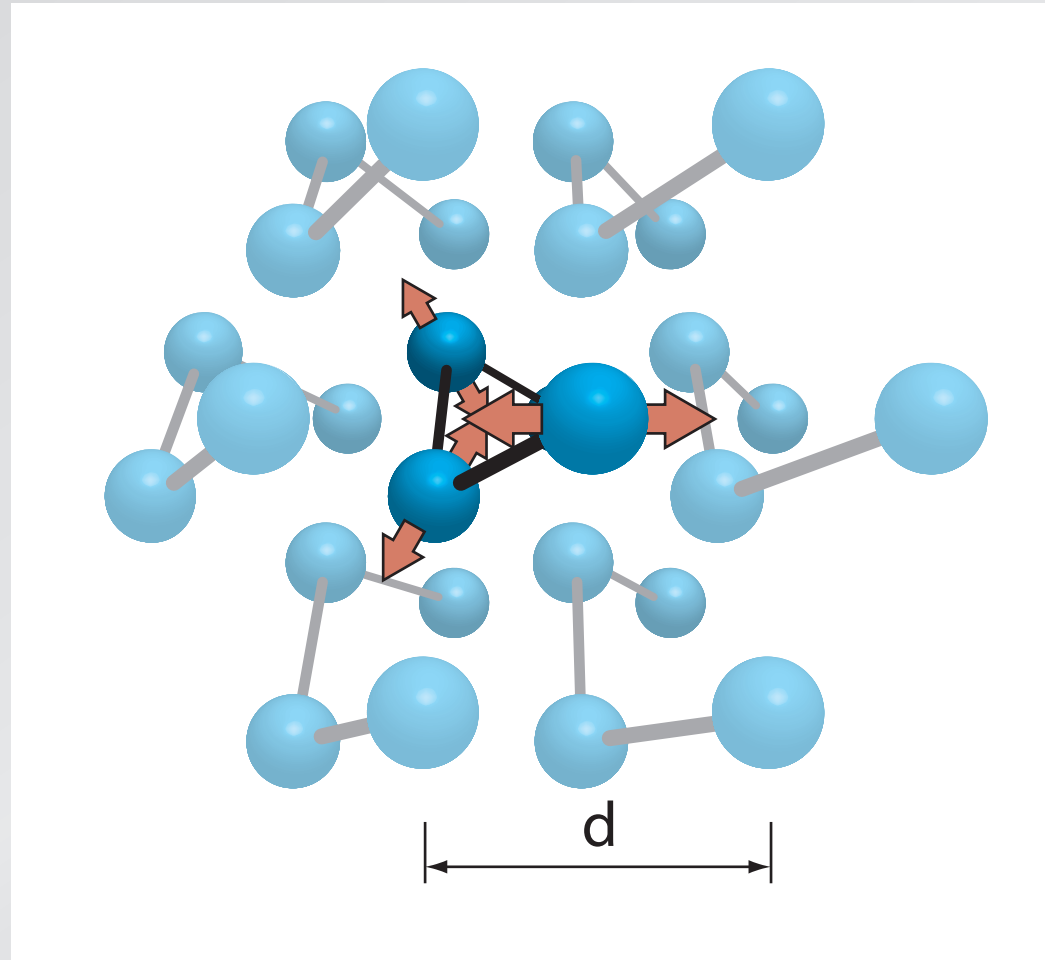
with lattice planes offset in spiral fashion

Coherent phonons



helical radius $x = 0.27d$

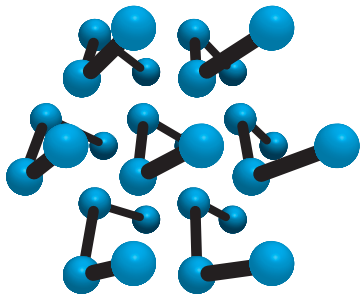
Coherent phonons



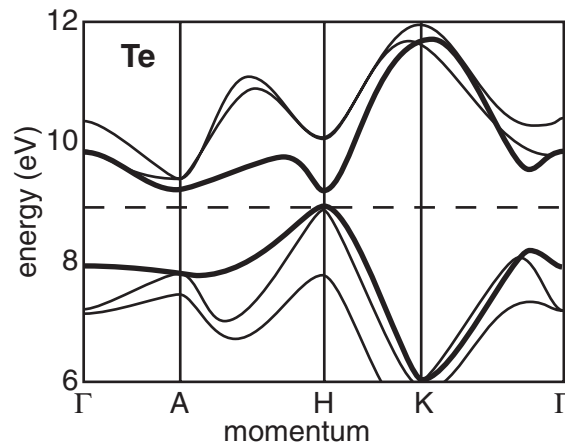
A_1 mode modulates x

Coherent phonons

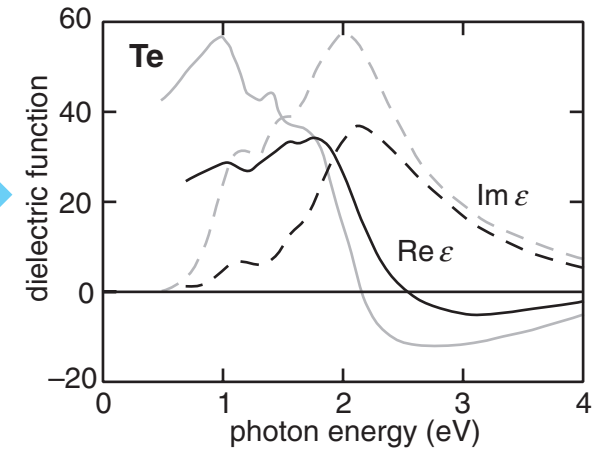
structure



band structure

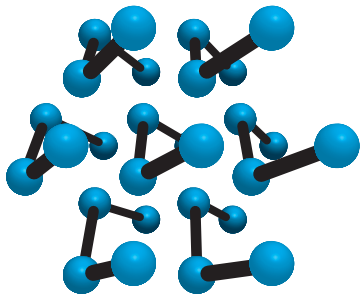


dielectric function

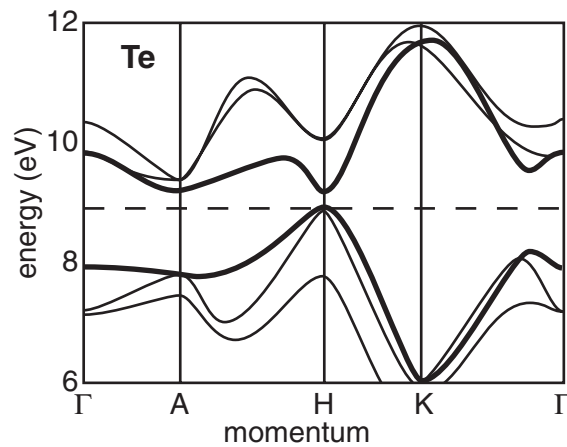


Coherent phonons

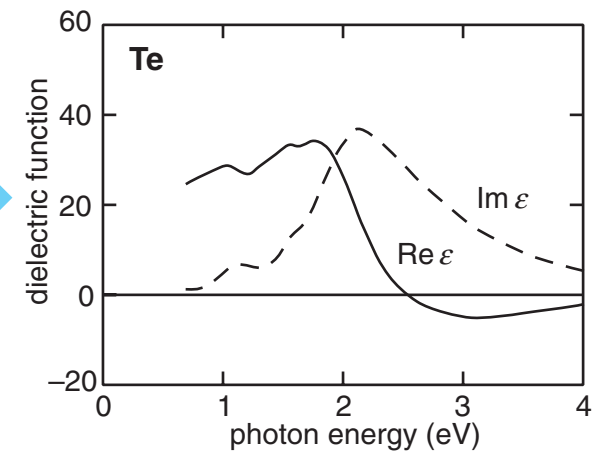
structure



band structure

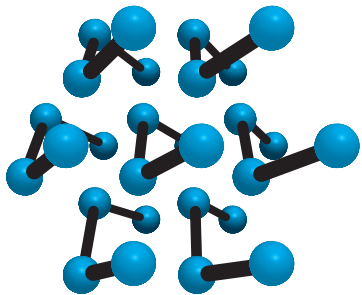


dielectric function

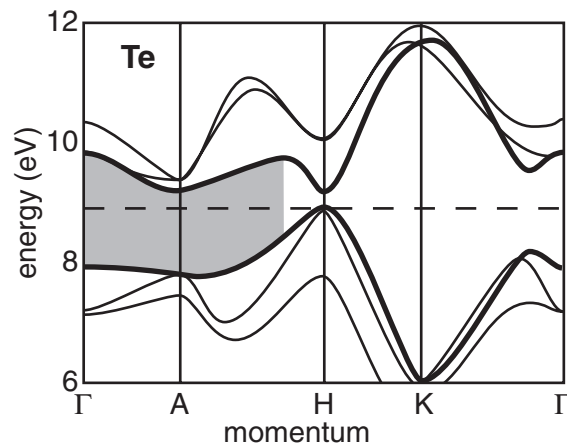


Coherent phonons

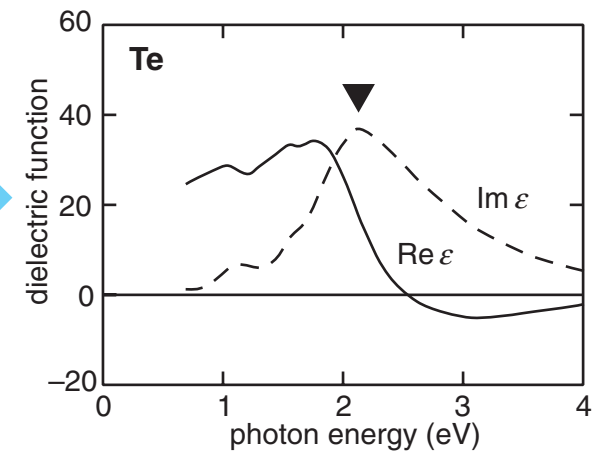
structure



band structure



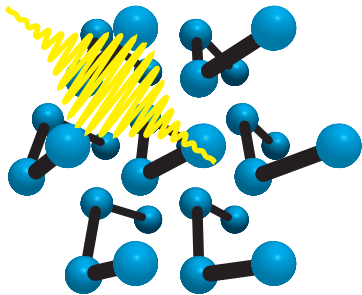
dielectric function



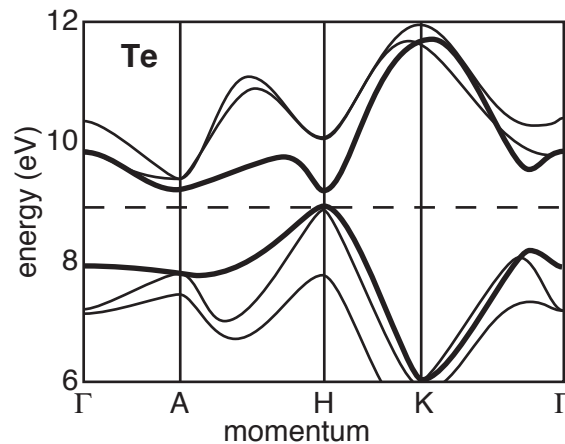
Coherent phonons

photoexcitation causes modulation of helical radius

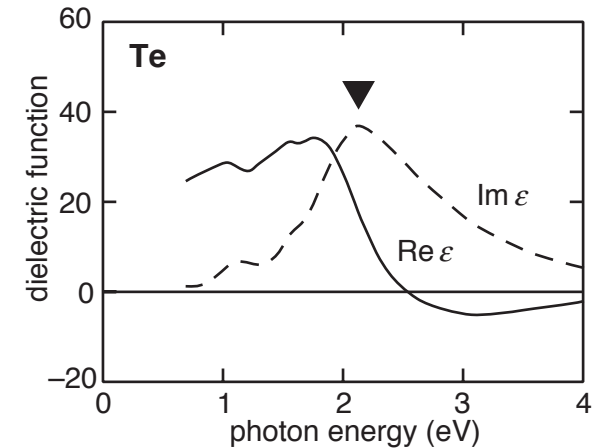
structure



band structure



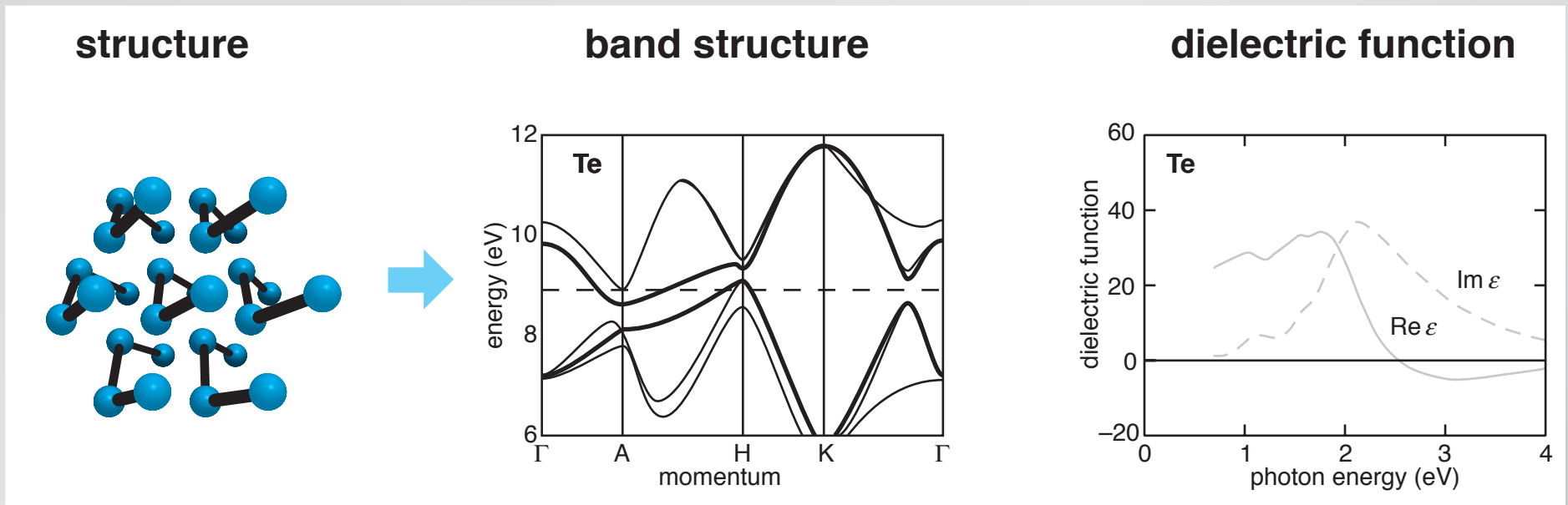
dielectric function



band structure very sensitive to helical radius

Coherent phonons

15% change drastically alters band structure

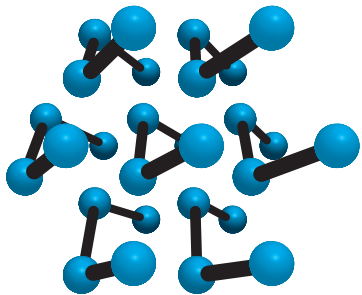


Tangney and Fahy, *Phys. Rev. B* 65, 054302 (2002)

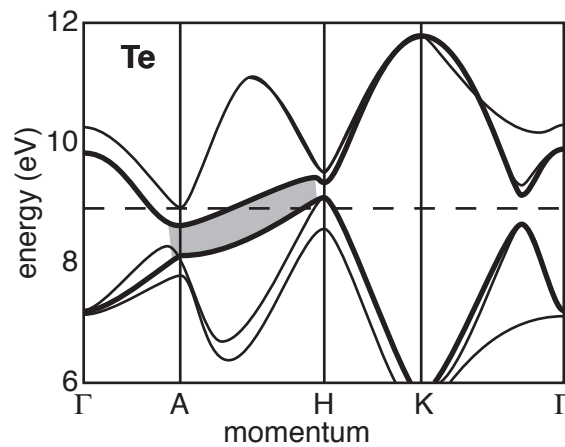
Coherent phonons

should cause a red-shift of dielectric function

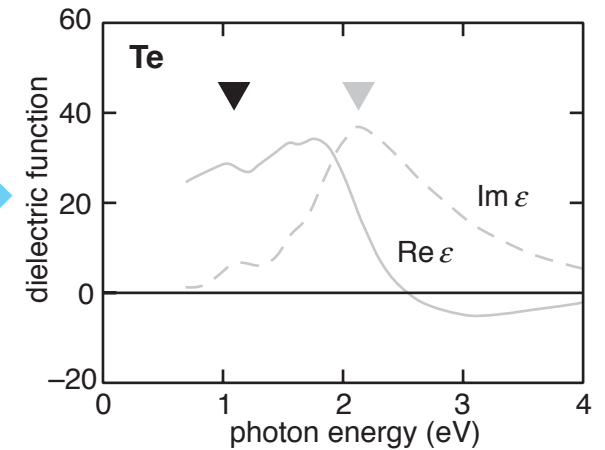
structure



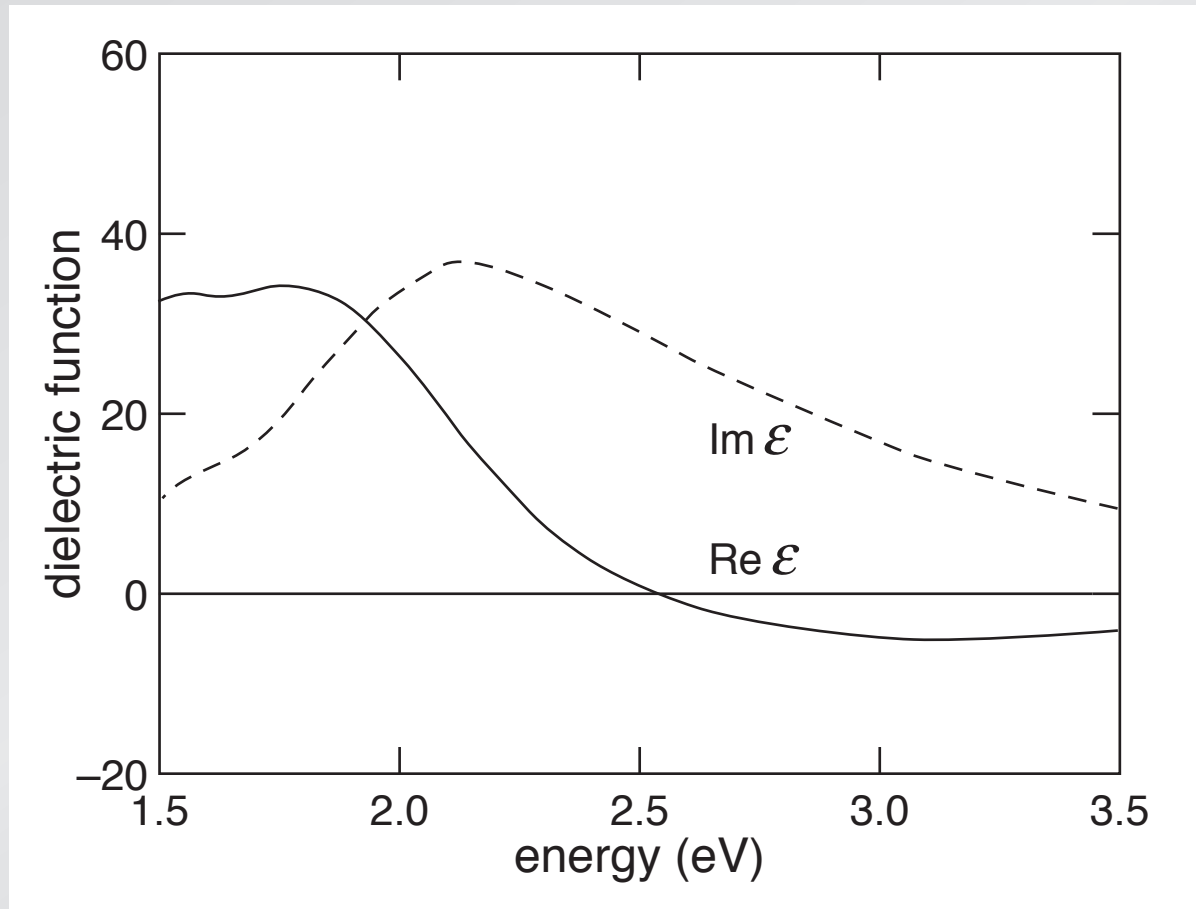
band structure



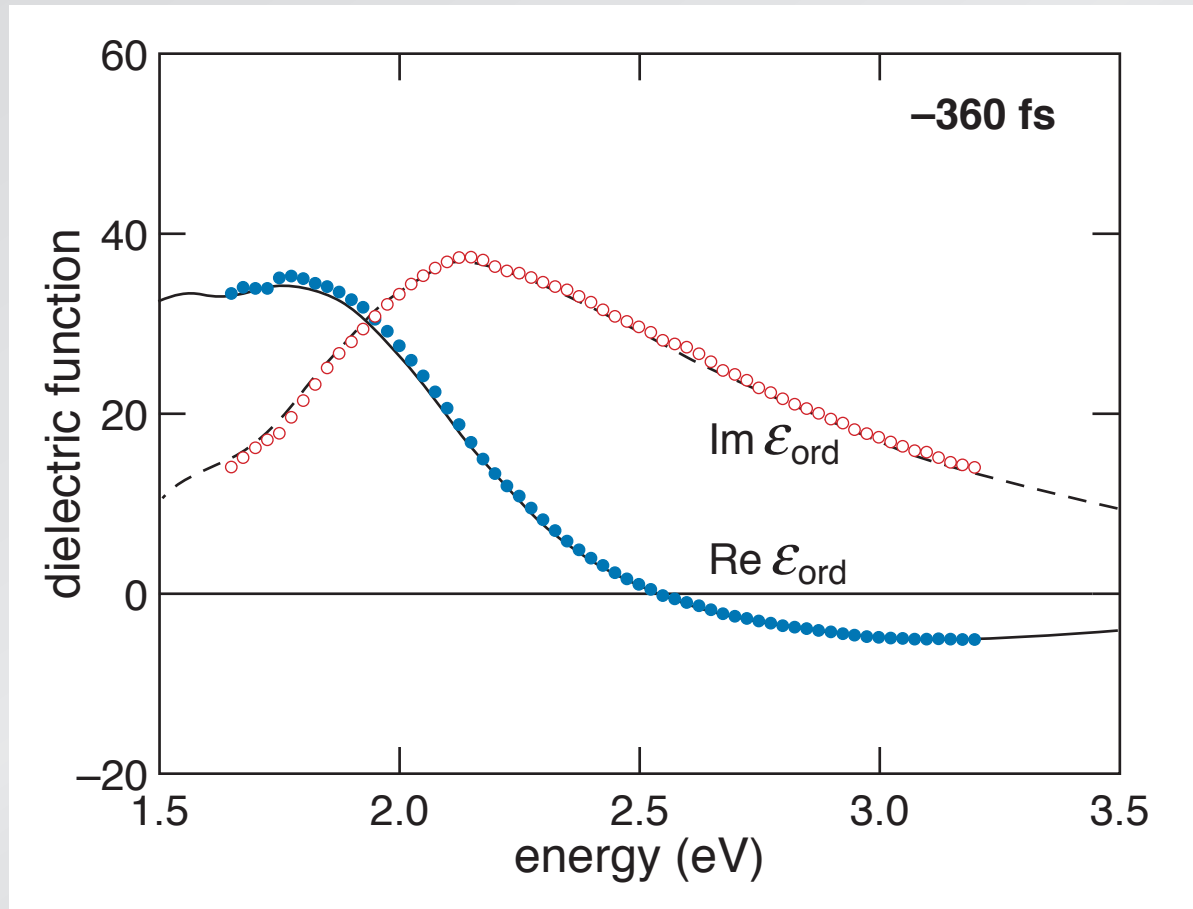
dielectric function



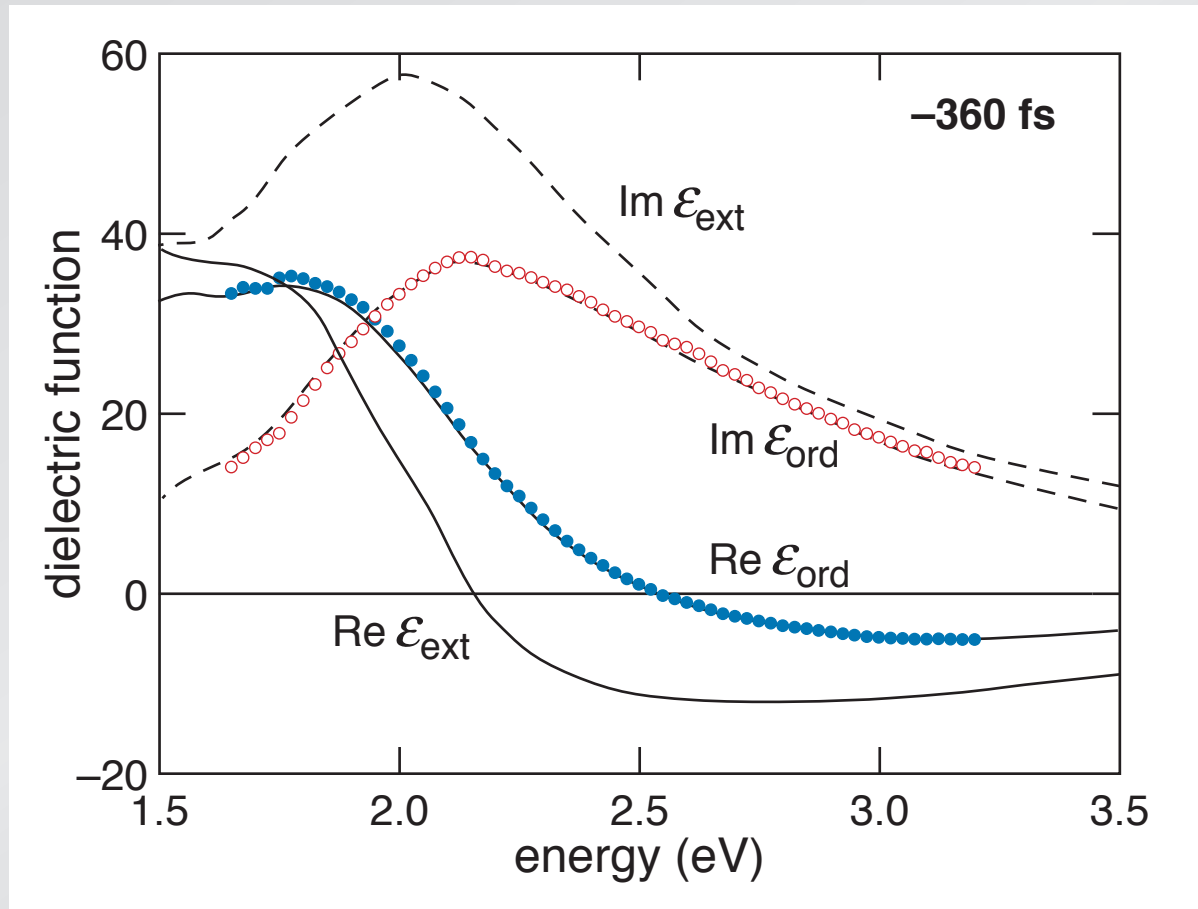
Coherent phonons



Coherent phonons

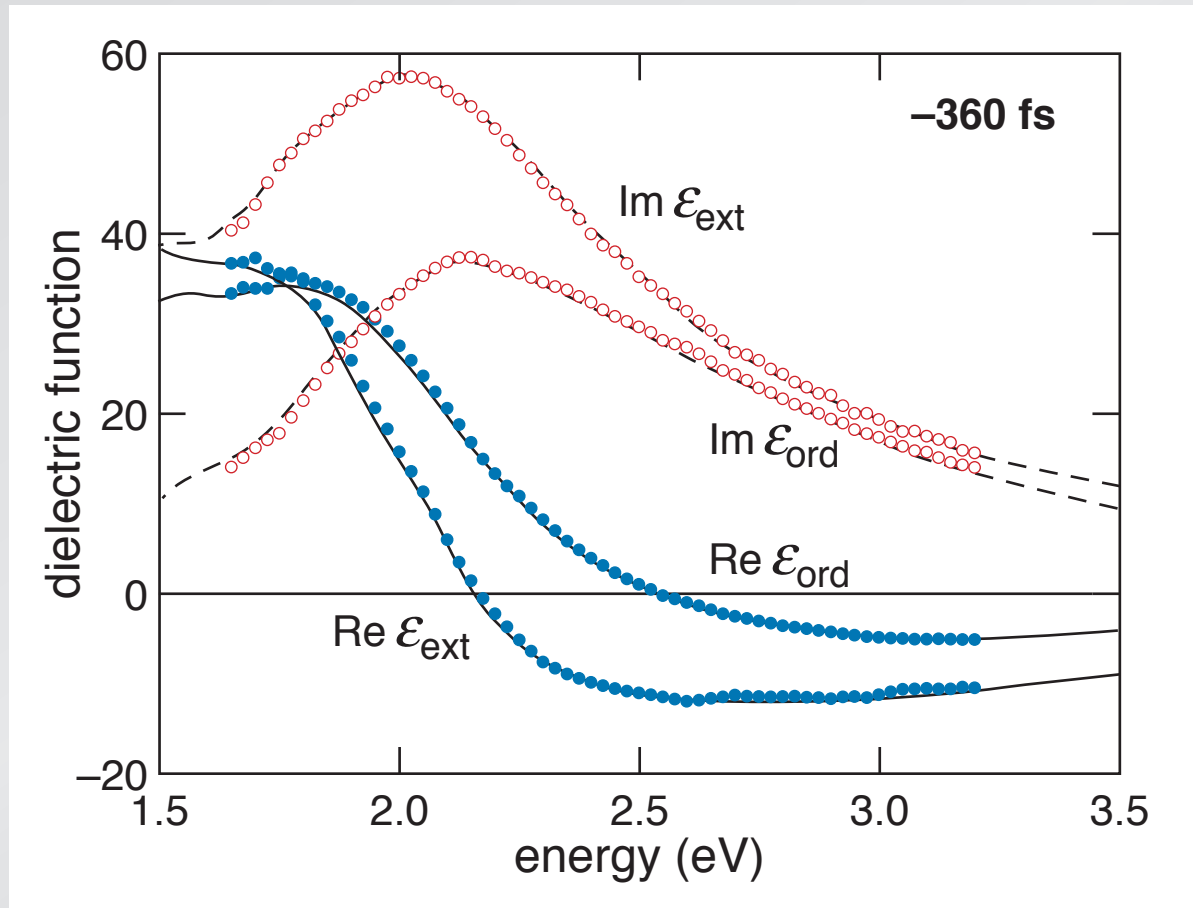


Coherent phonons



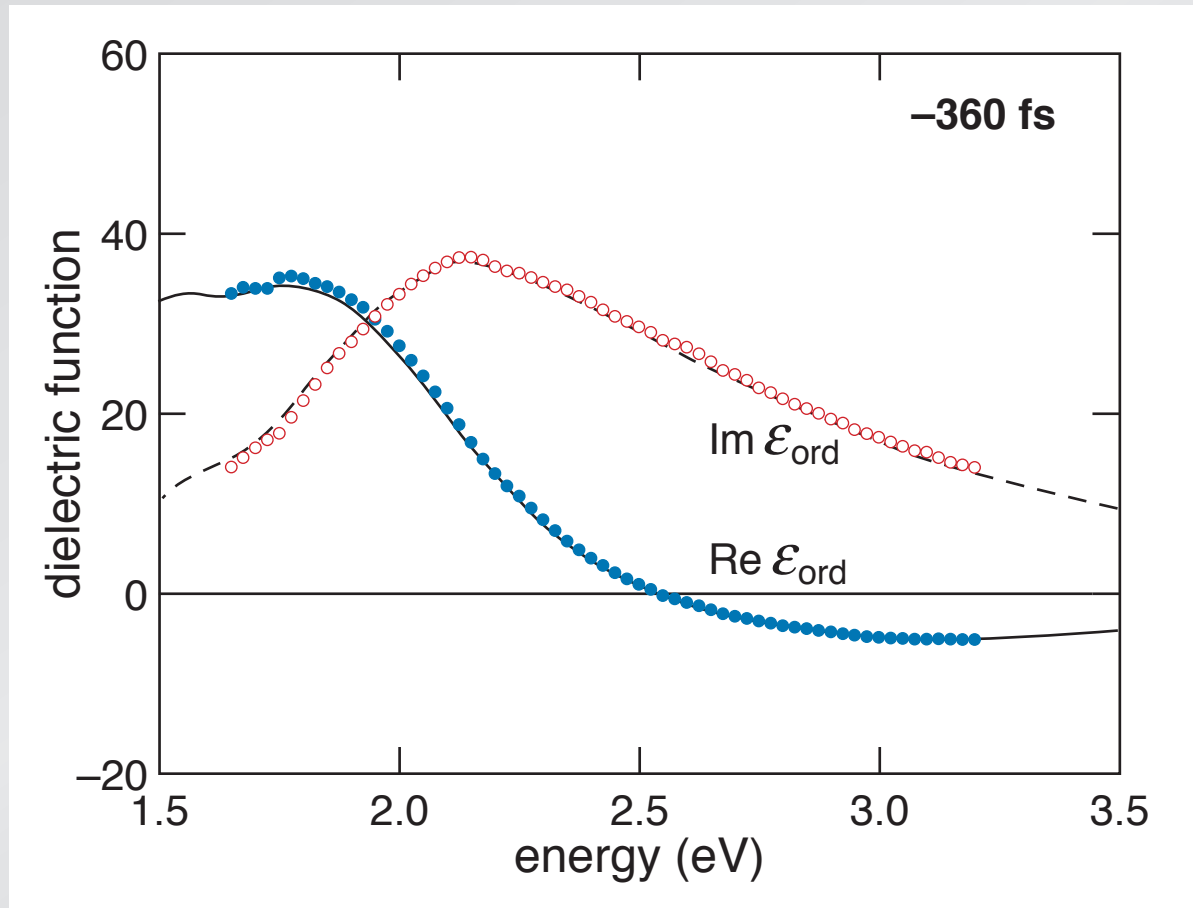
Coherent phonons

data agree well with literature values

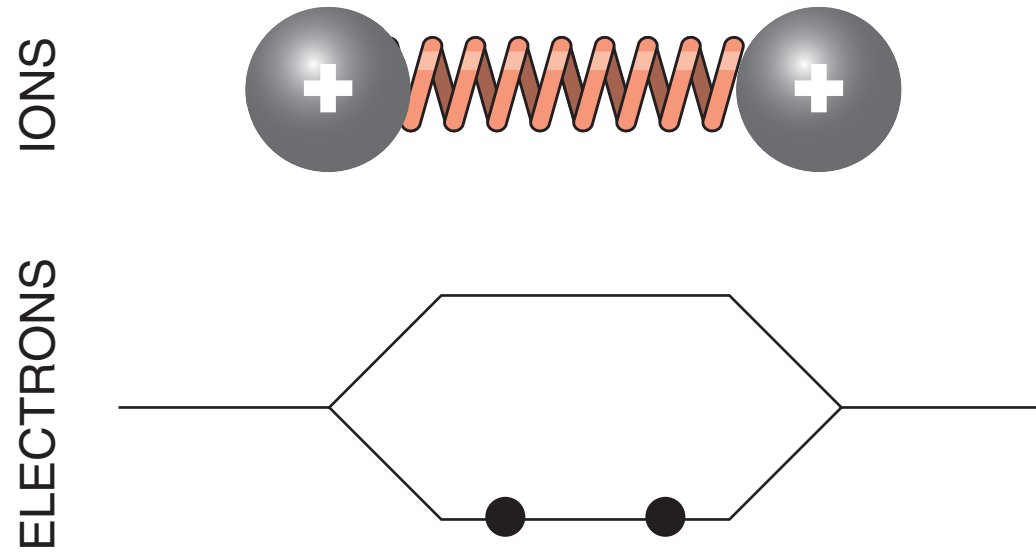


Coherent phonons

now vary pump probe delay

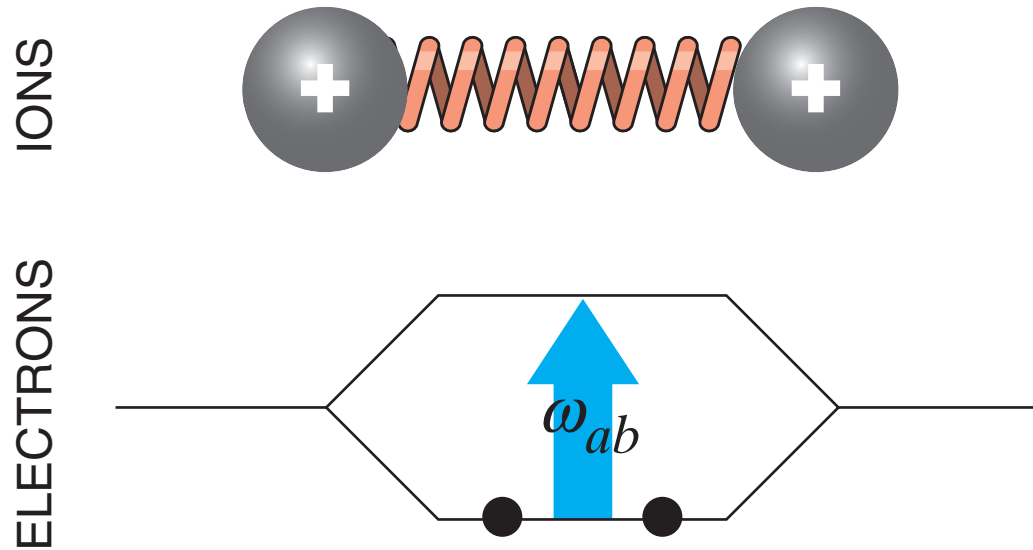


Coherent phonons



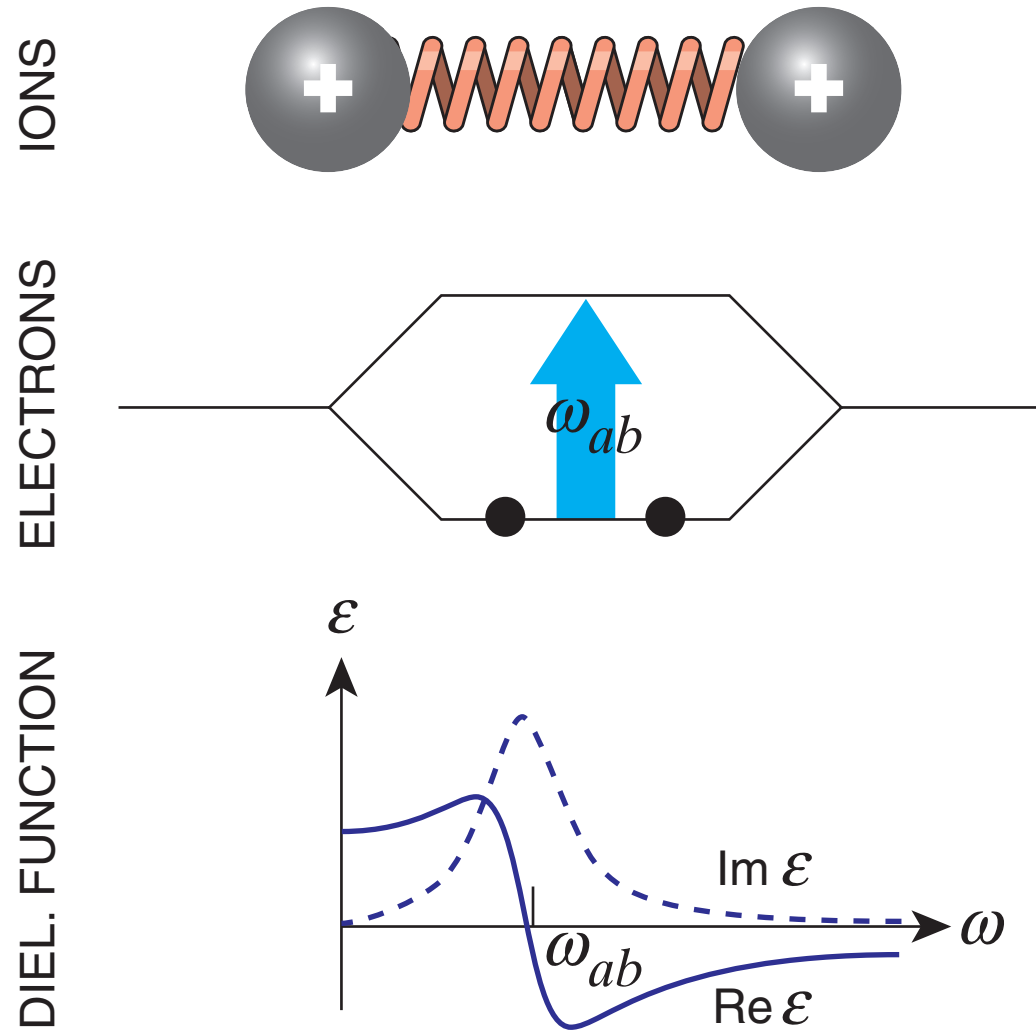
“two-atom model”

Coherent phonons



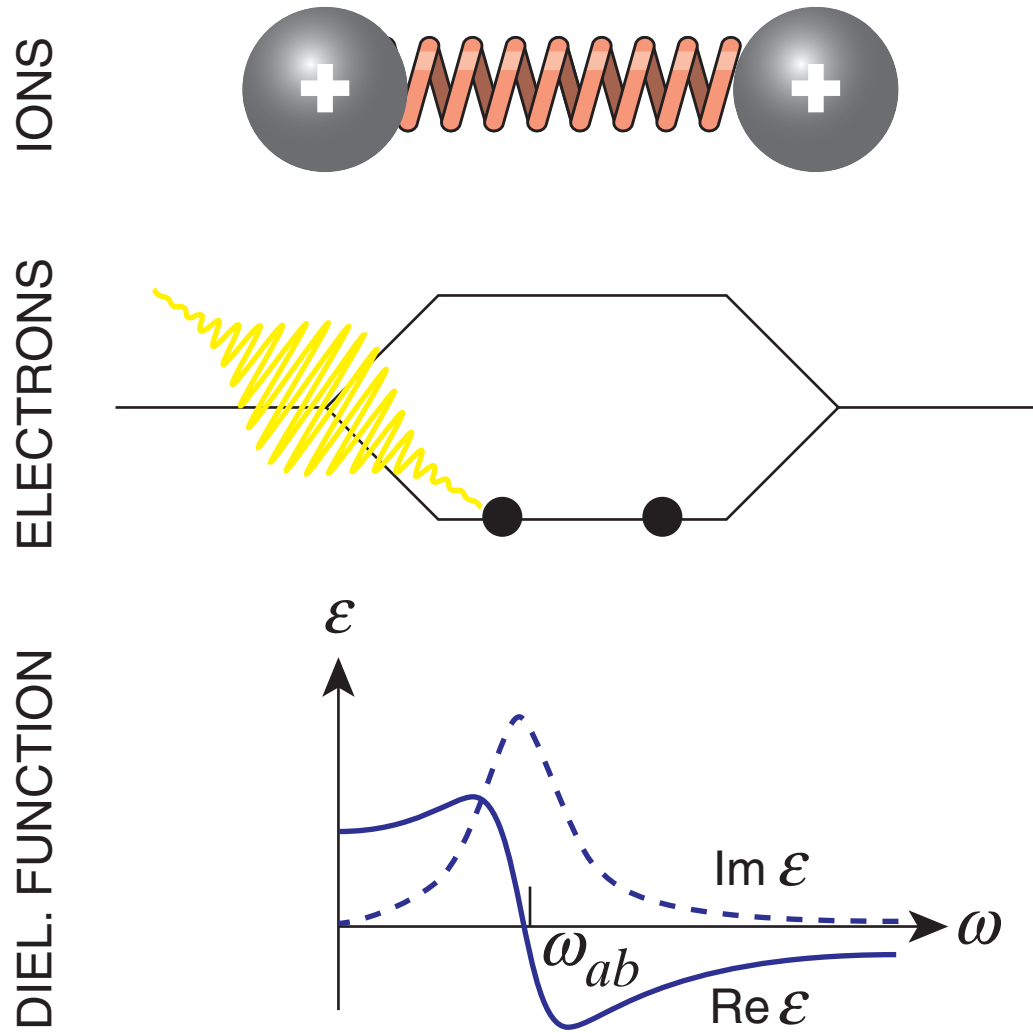
bonding-antibonding splitting

Coherent phonons



Lorentz model

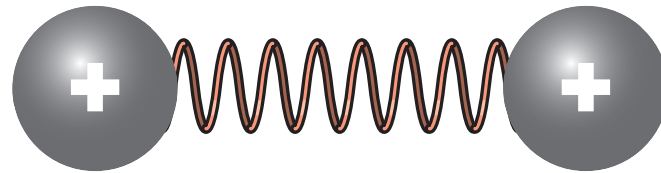
Coherent phonons



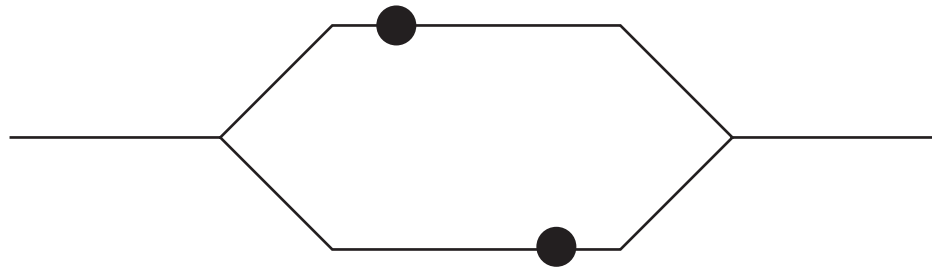
photon promotes electron...

Coherent phonons

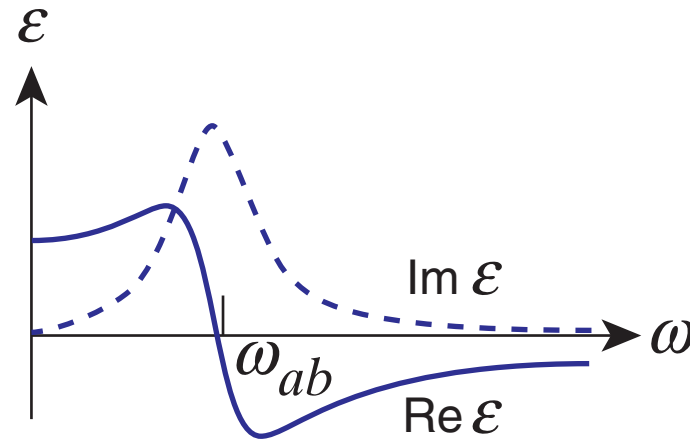
IONS



ELECTRONS

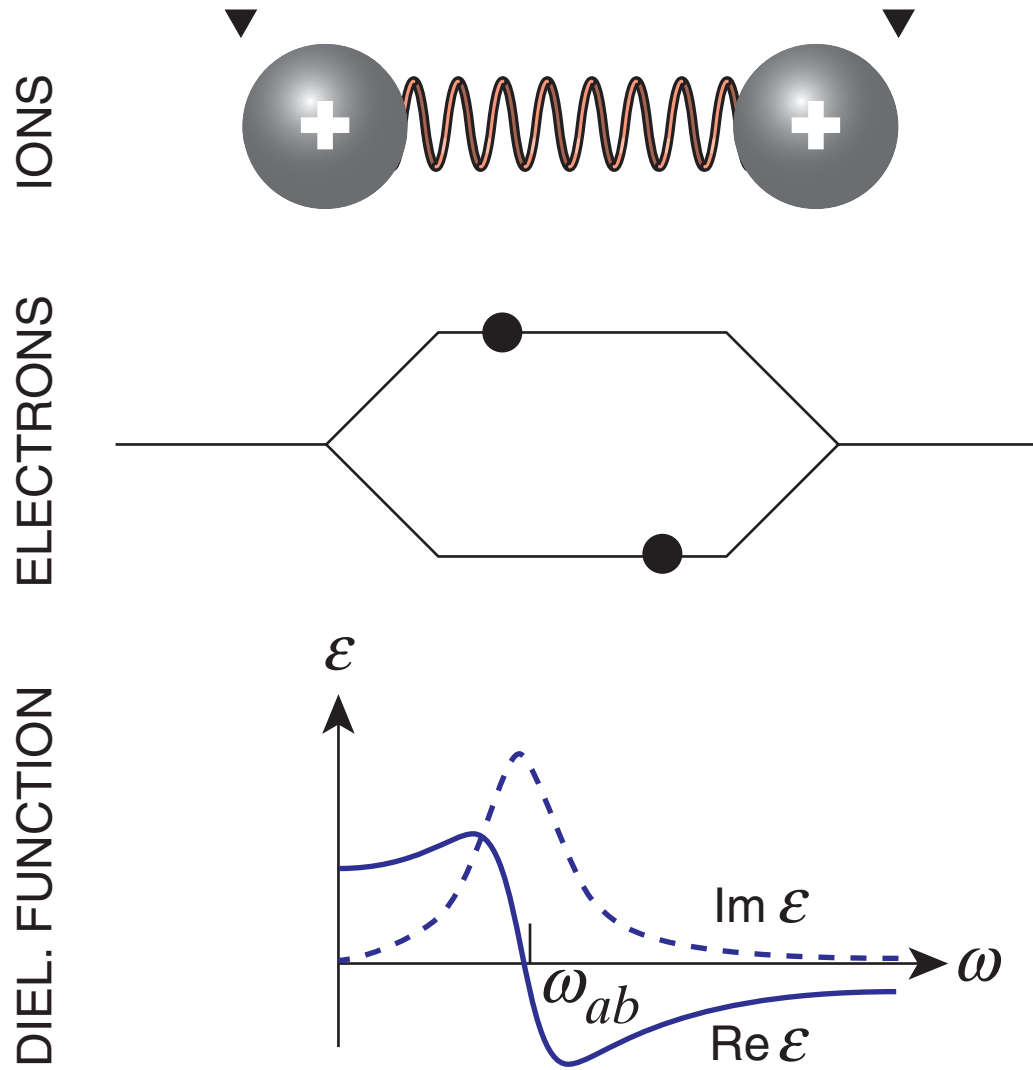


DIEL. FUNCTION



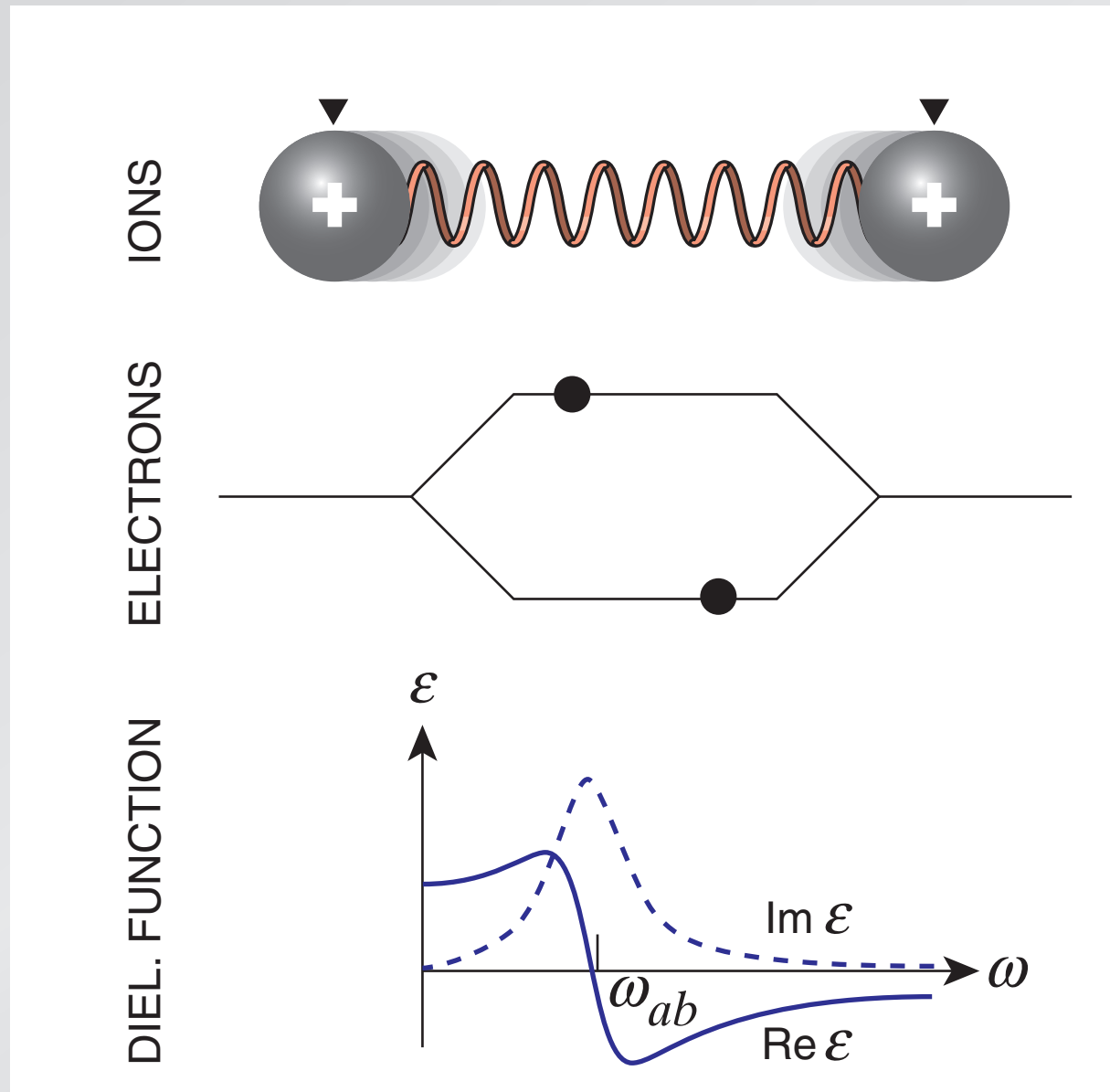
...weakening binding force...

Coherent phonons



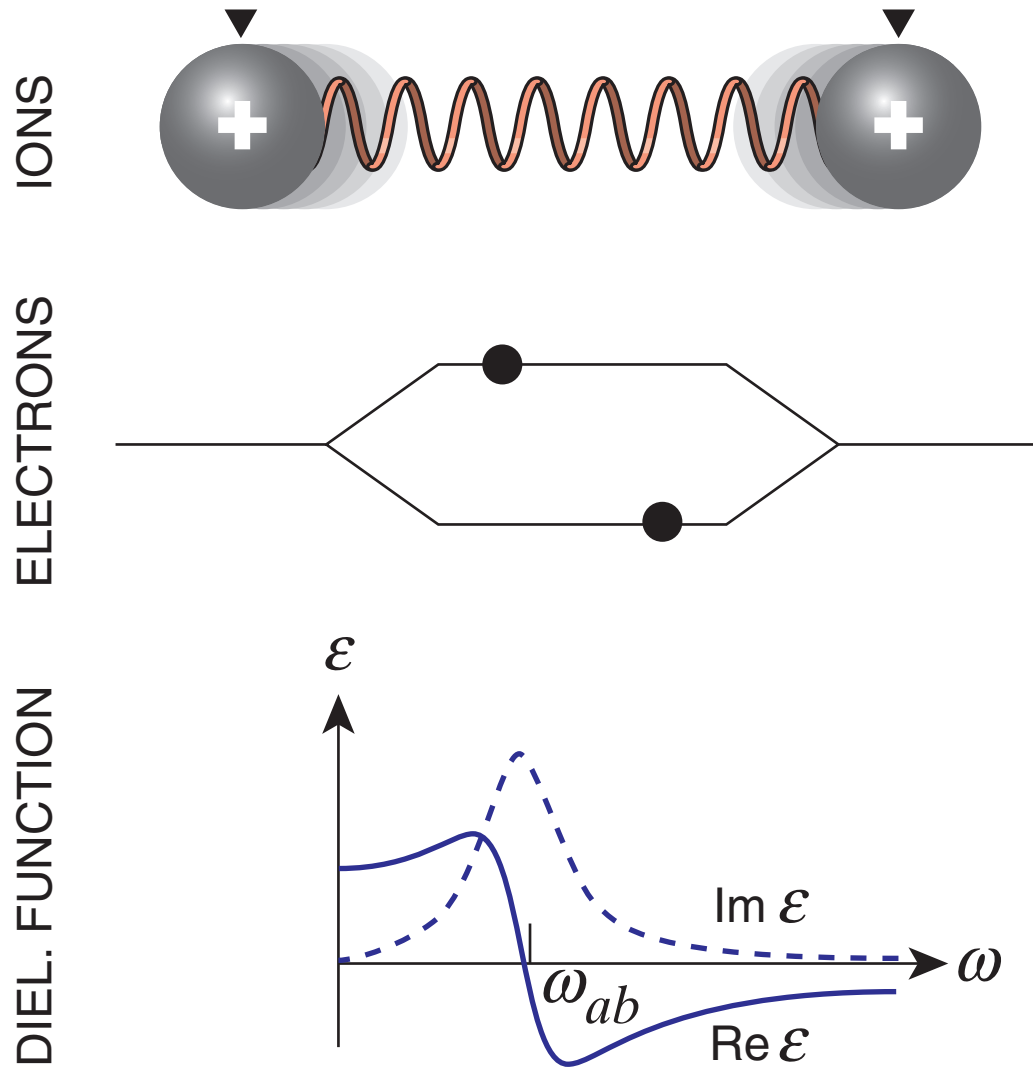
...establishing new equilibrium positions

Coherent phonons



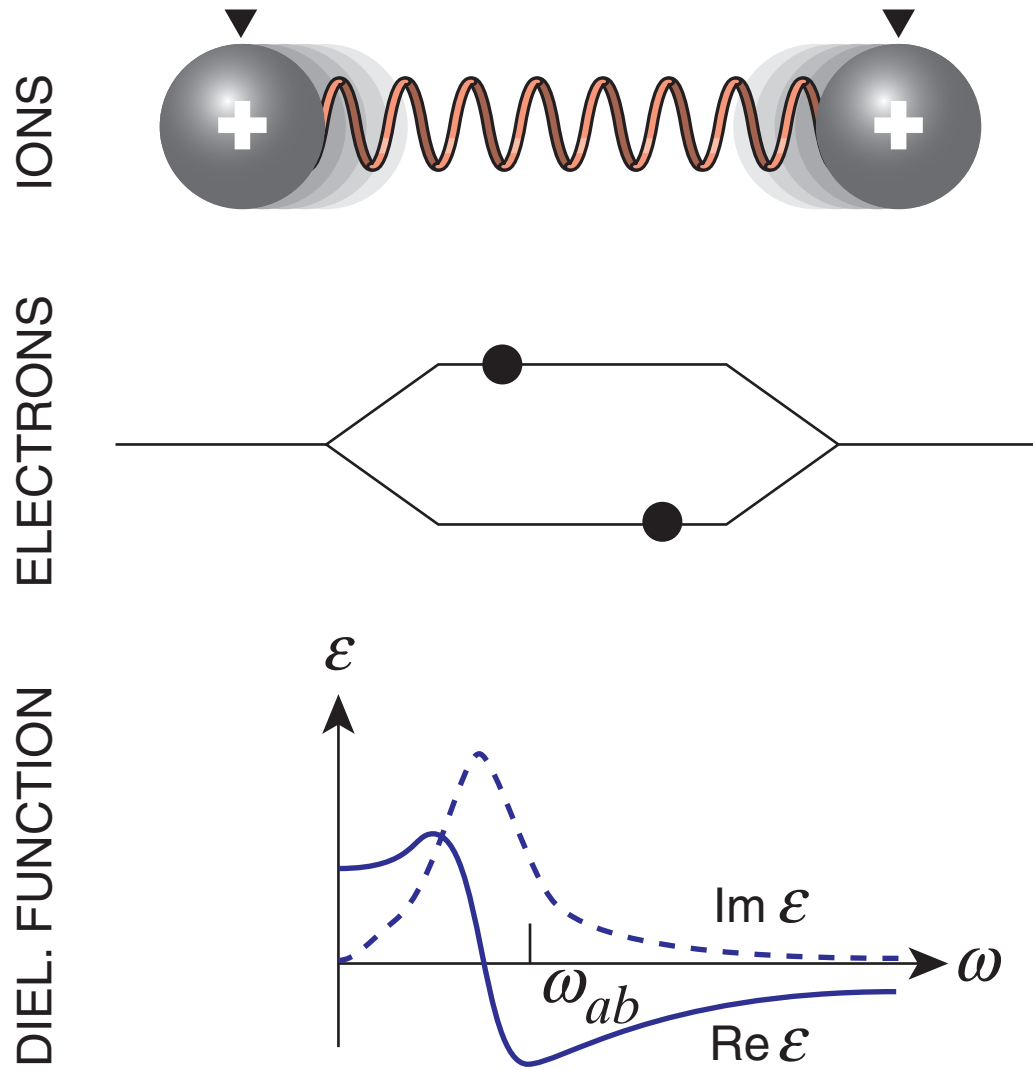
ions move to new equilibrium positions...

Coherent phonons



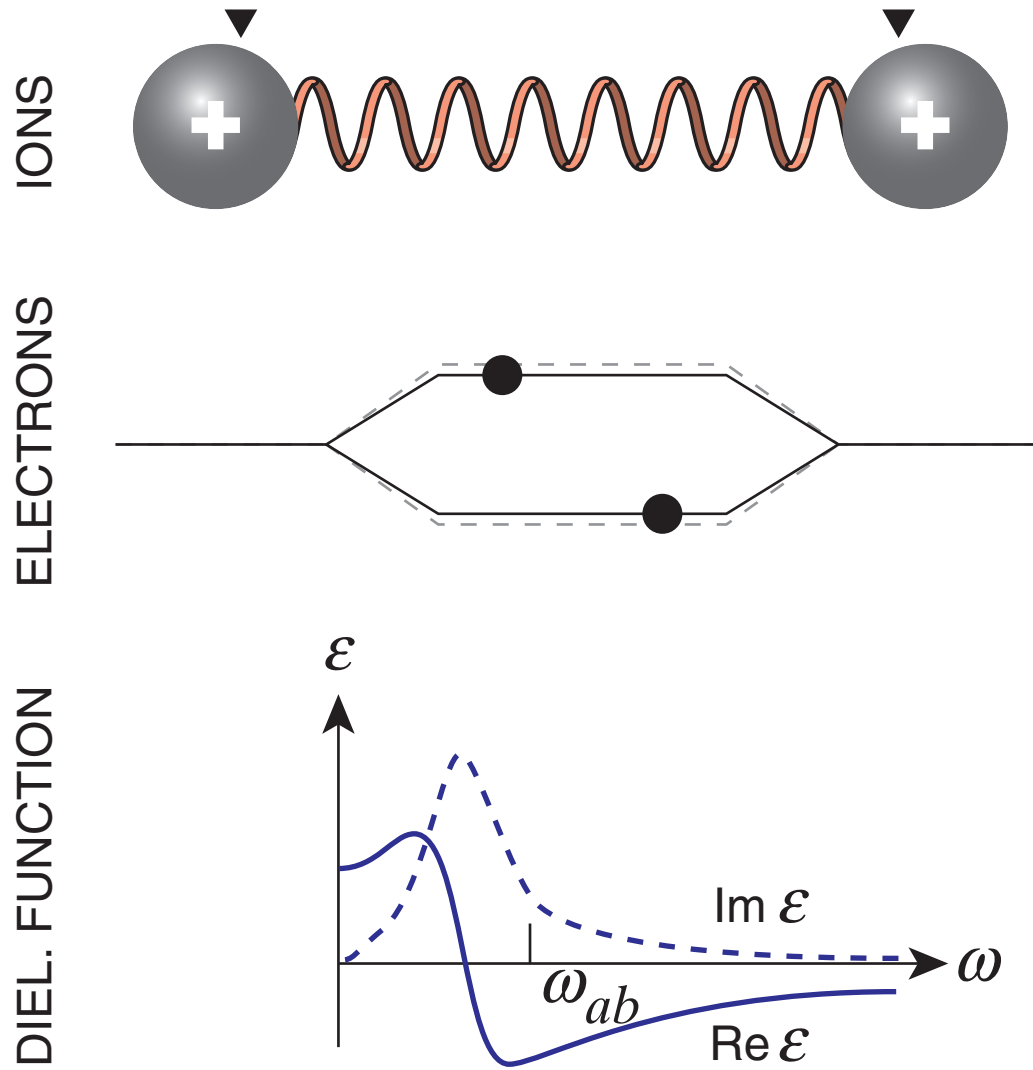
...diminishing splitting...

Coherent phonons



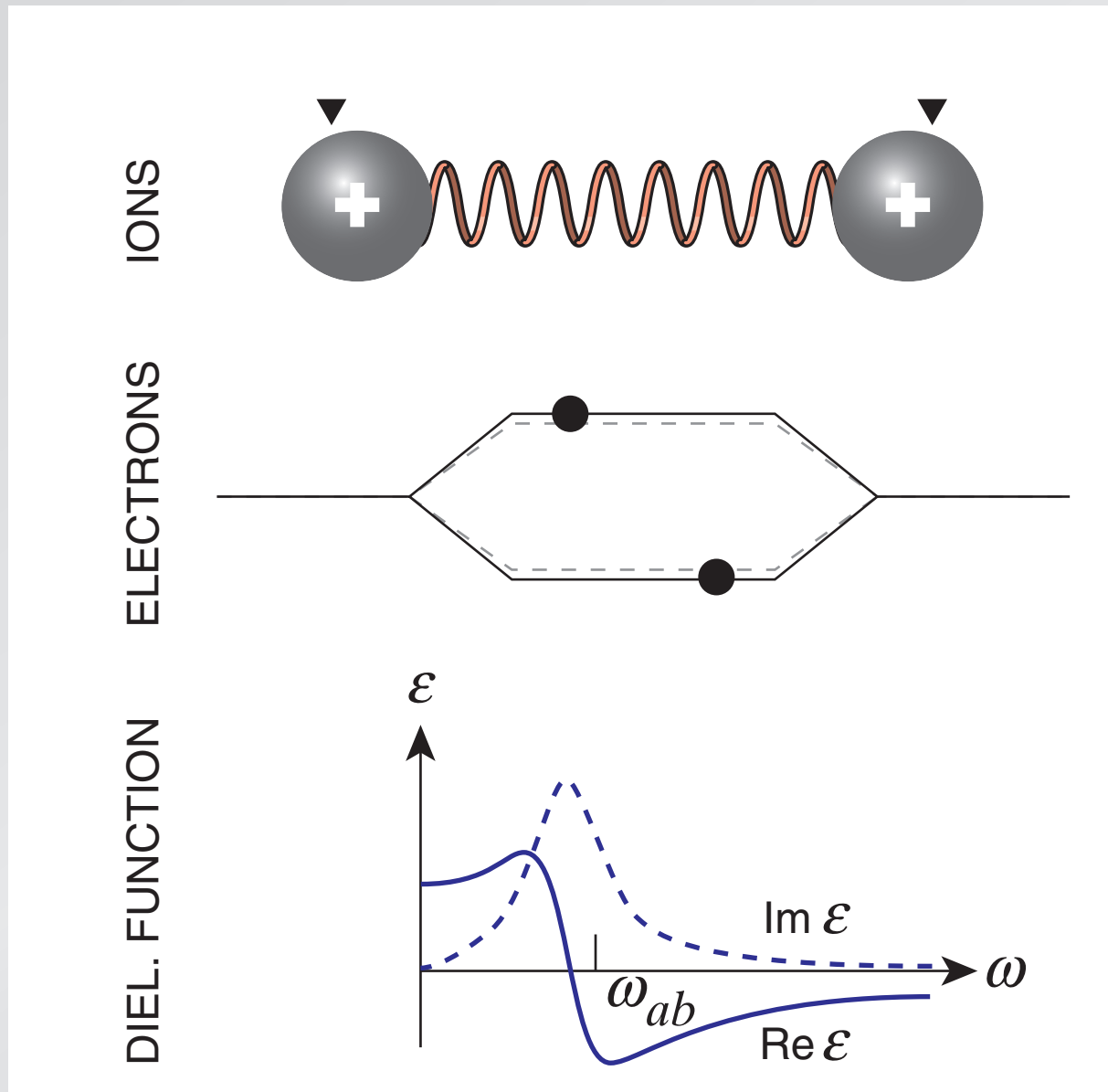
...and red-shifting the dielectric function

Coherent phonons



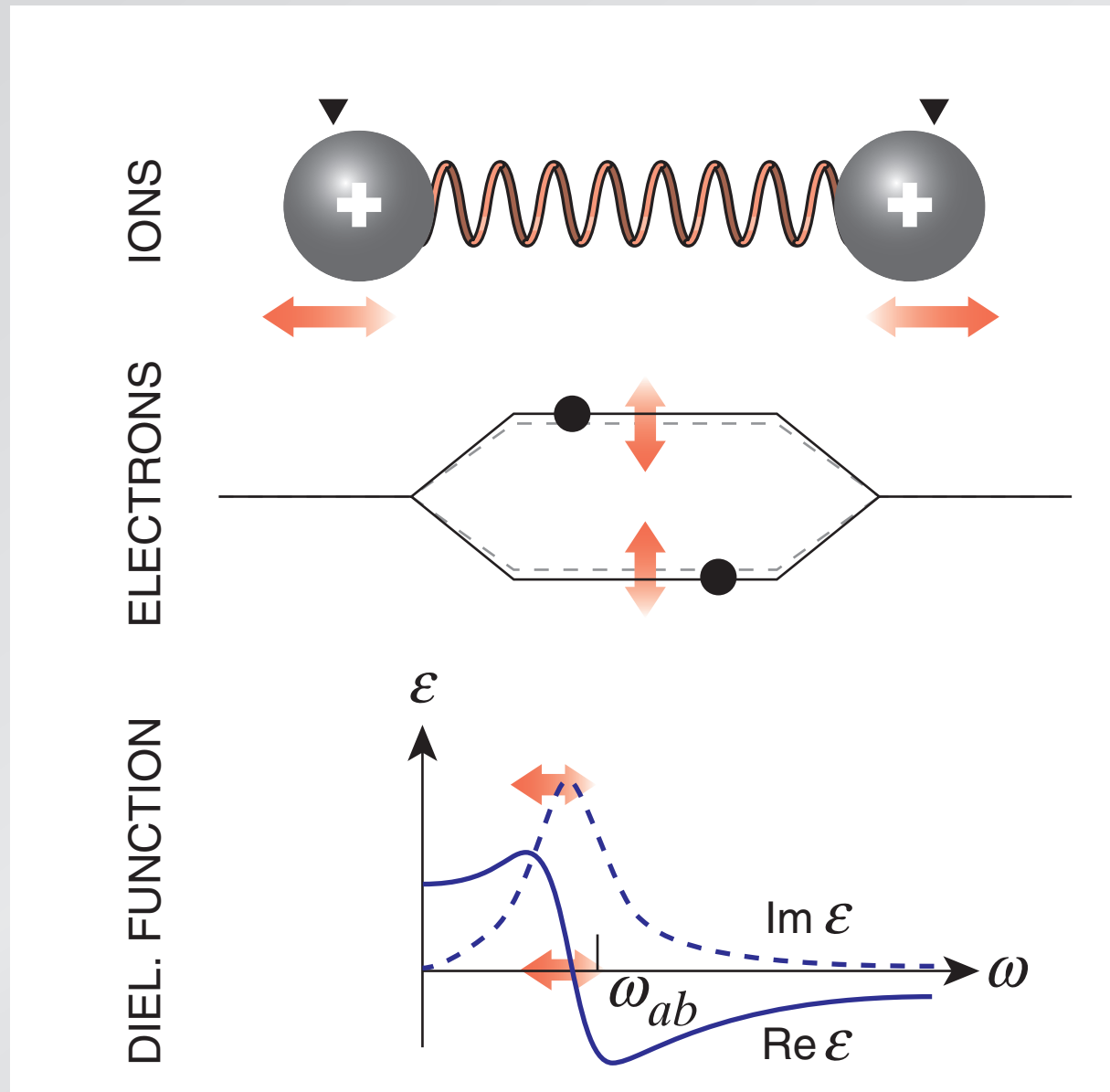
ions overshoot equilibrium position...

Coherent phonons



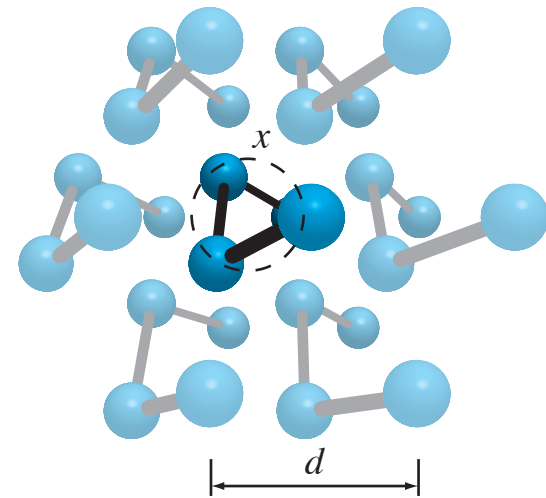
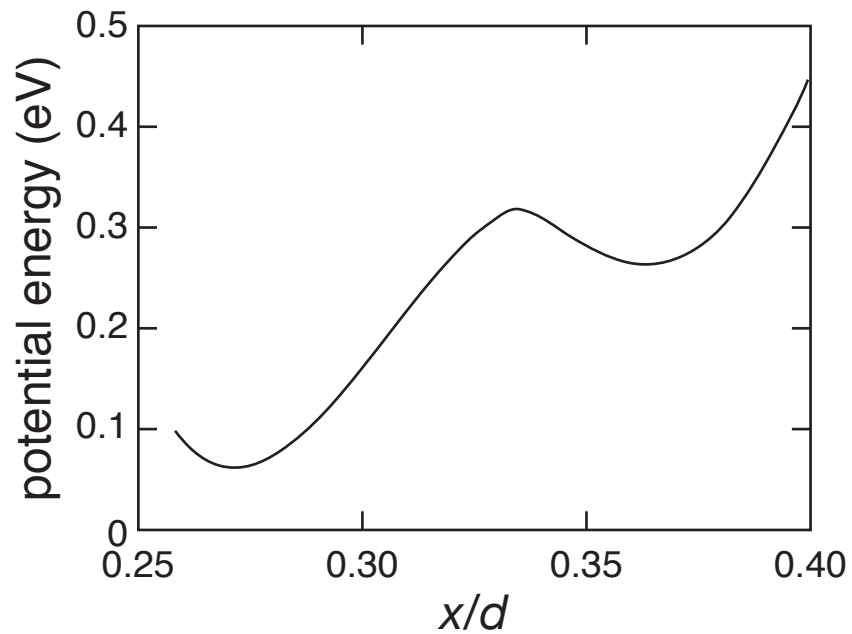
...reversing travel and overshooting again

Coherent phonons



oscillation around "displaced" equilibrium

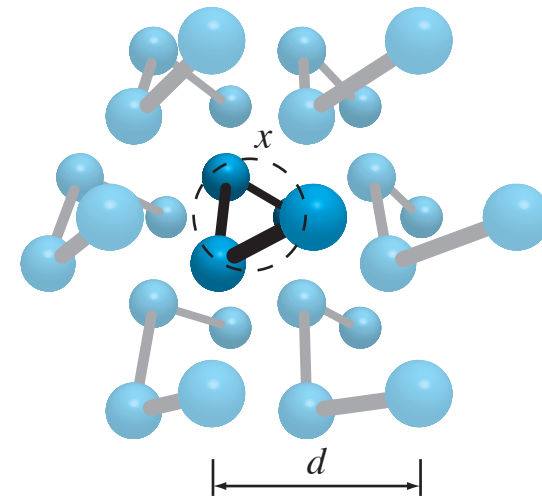
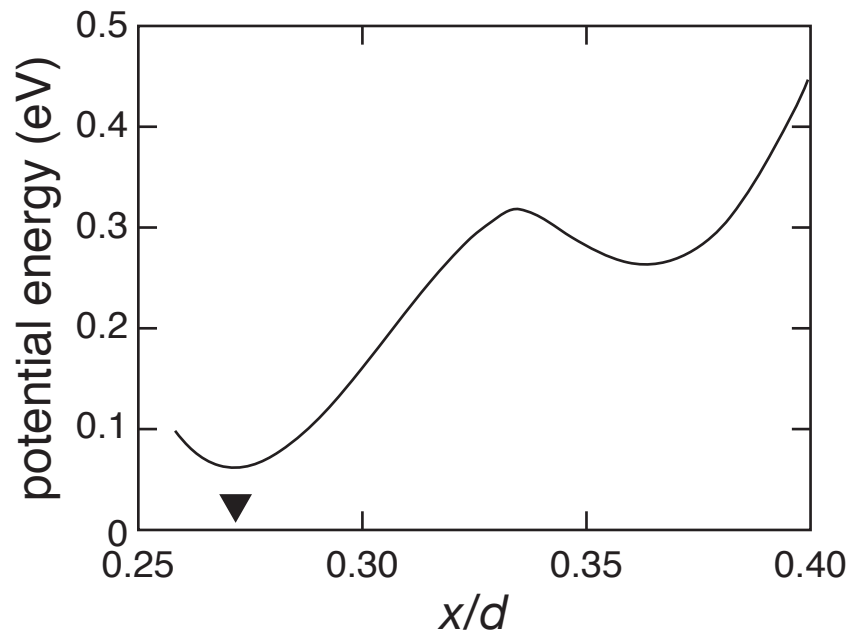
Coherent phonons



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

Coherent phonons

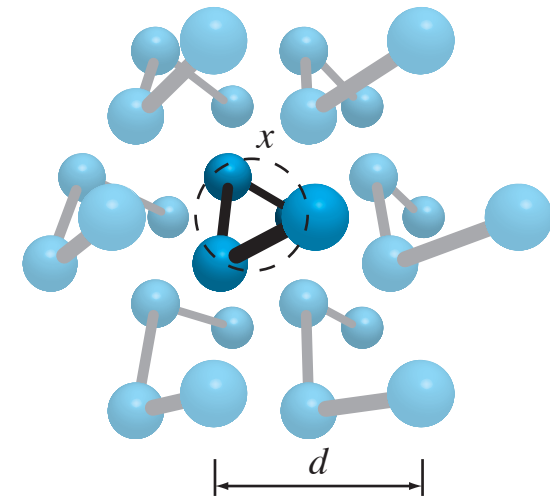
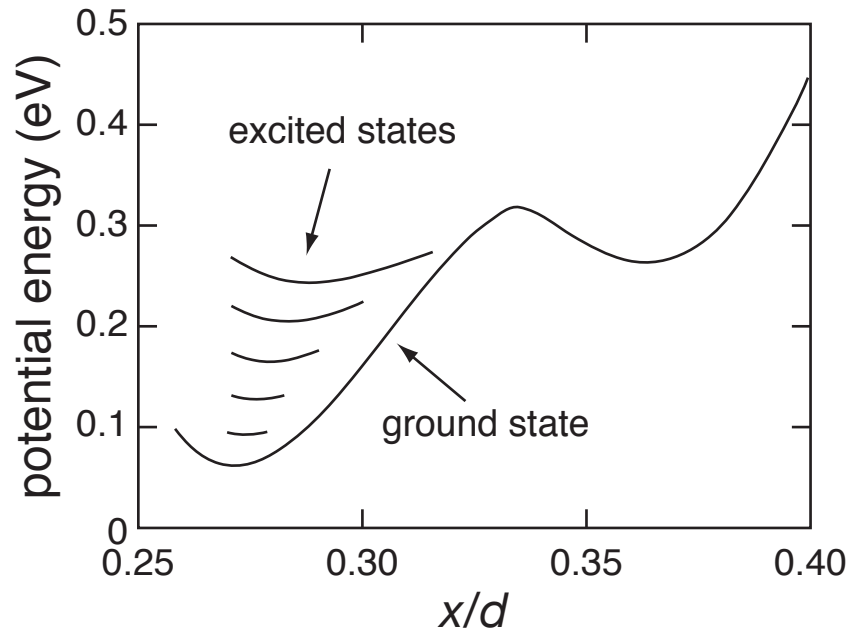
ground state equilibrium at $x/d = 0.27$



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

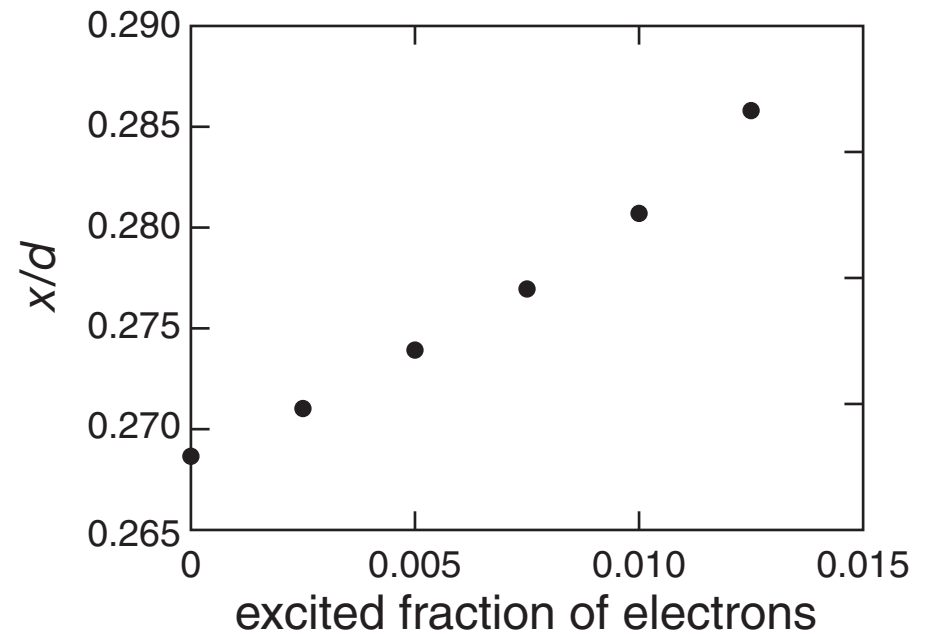
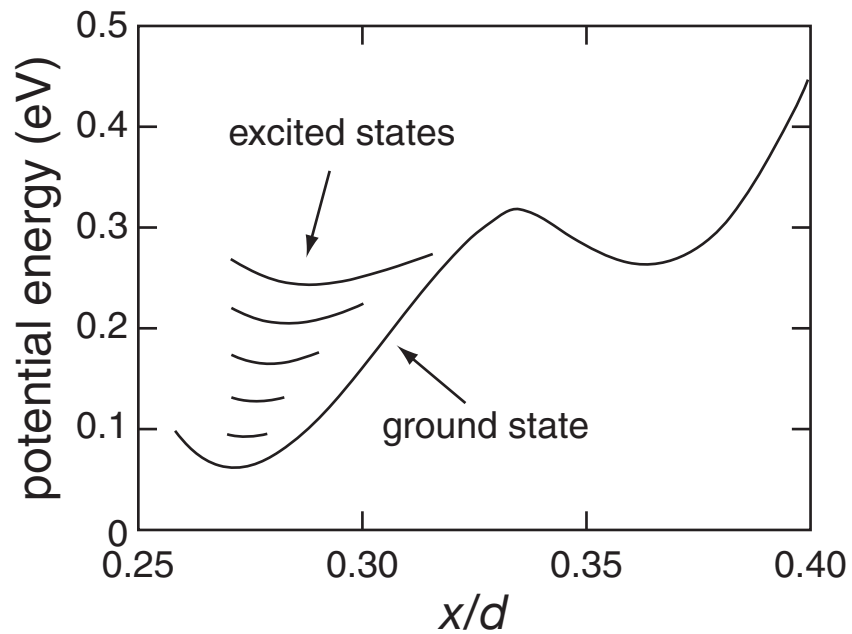
Coherent phonons

equilibrium position shifts upon excitation



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

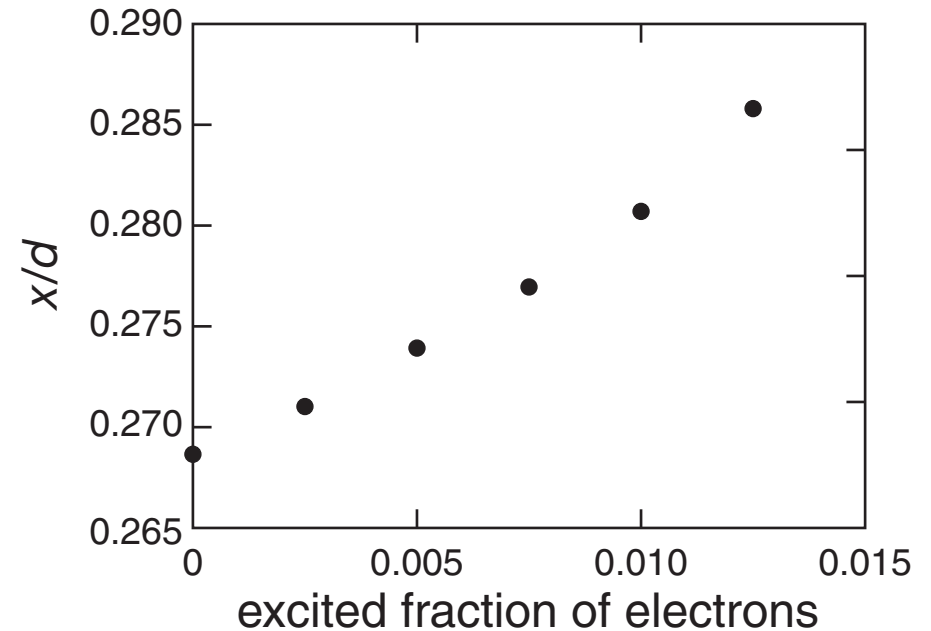
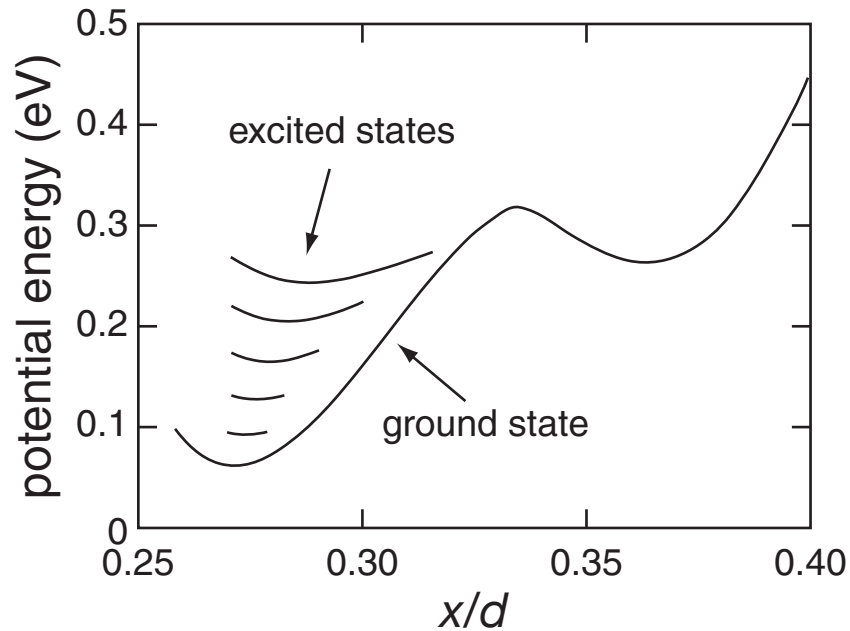
Coherent phonons



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

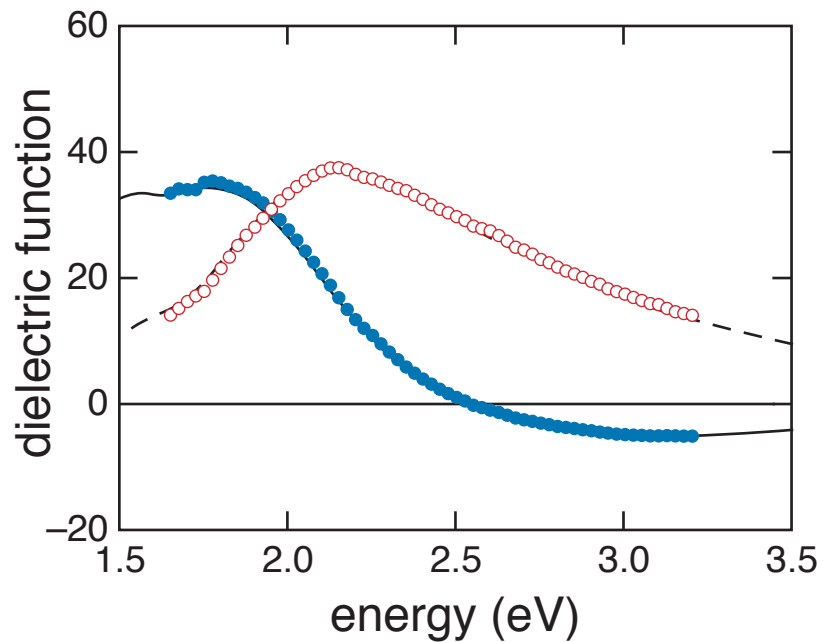
Coherent phonons

band structure depends on lattice configuration



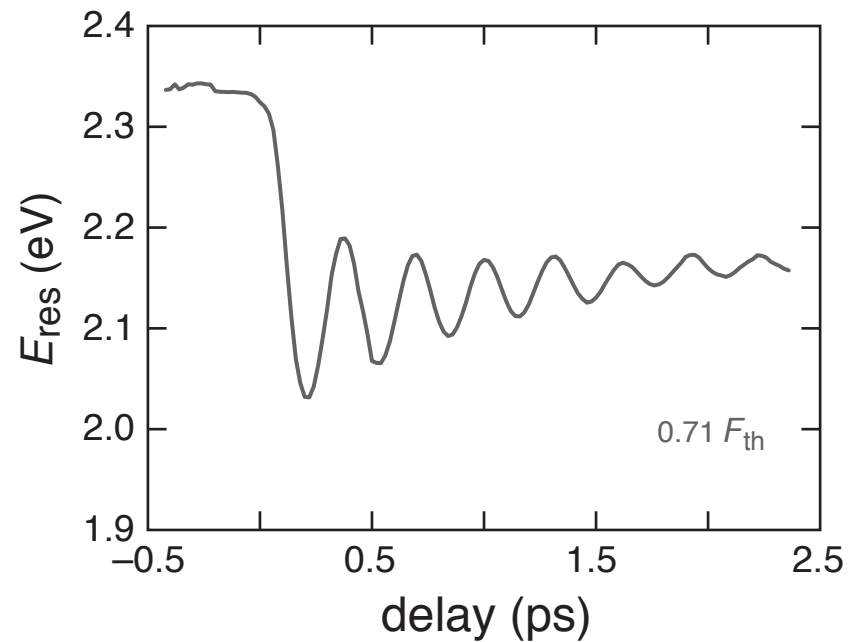
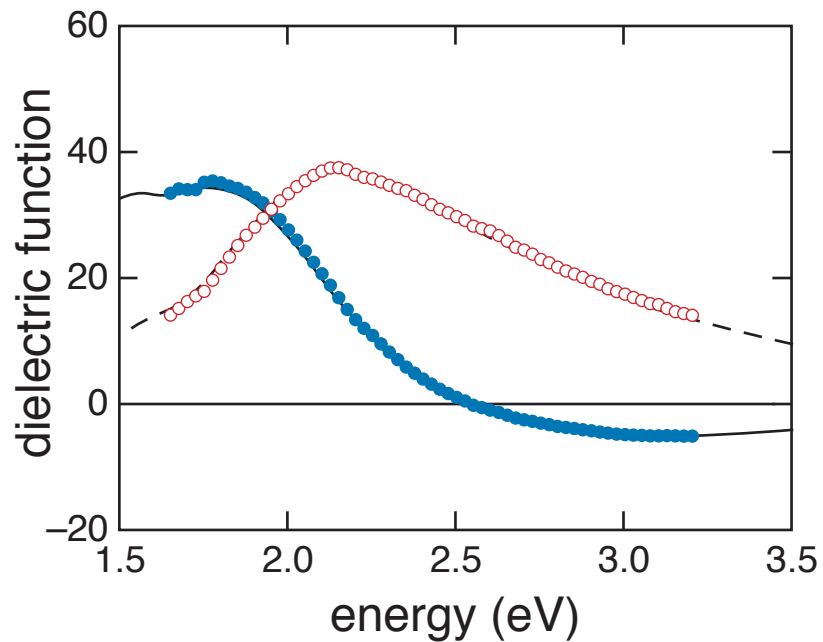
Coherent phonons

dielectric function reveals band structure changes



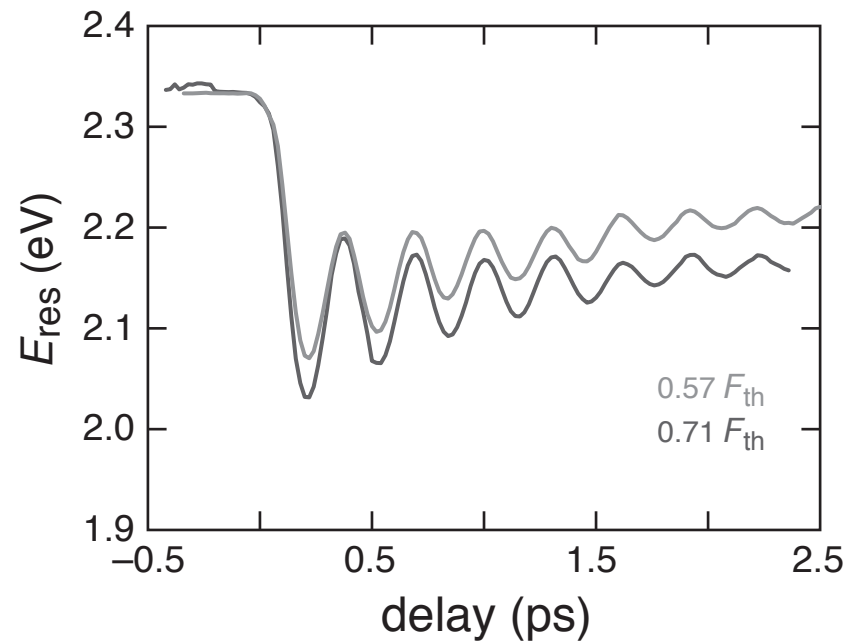
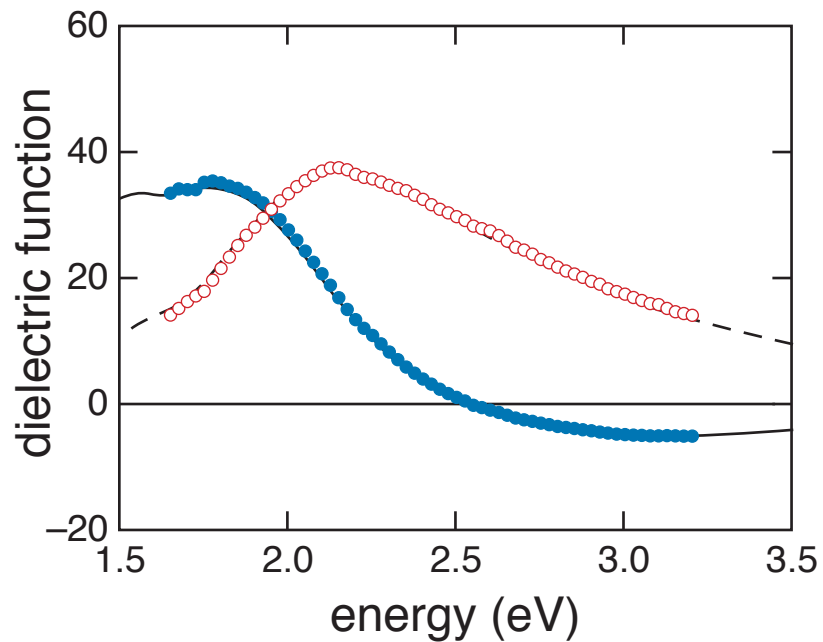
Coherent phonons

track resonance energy



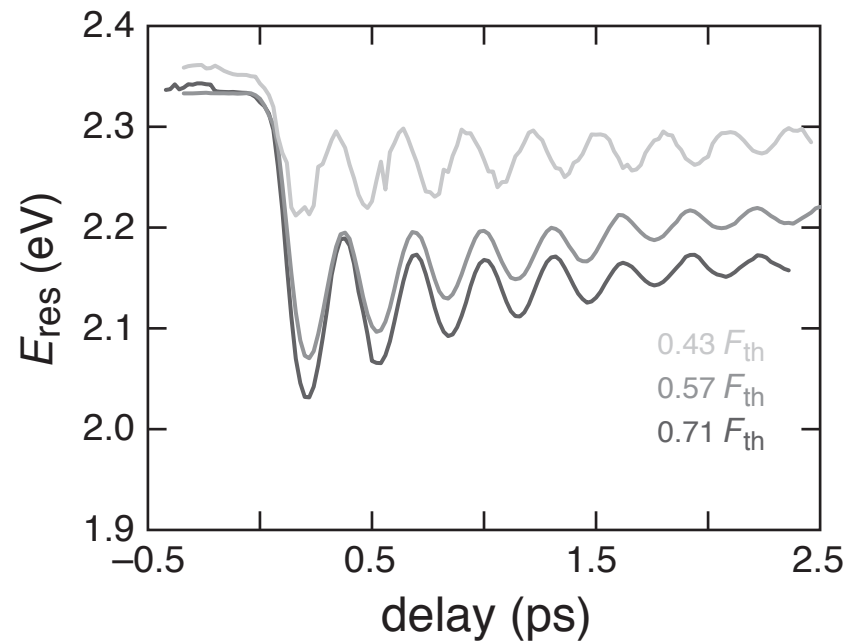
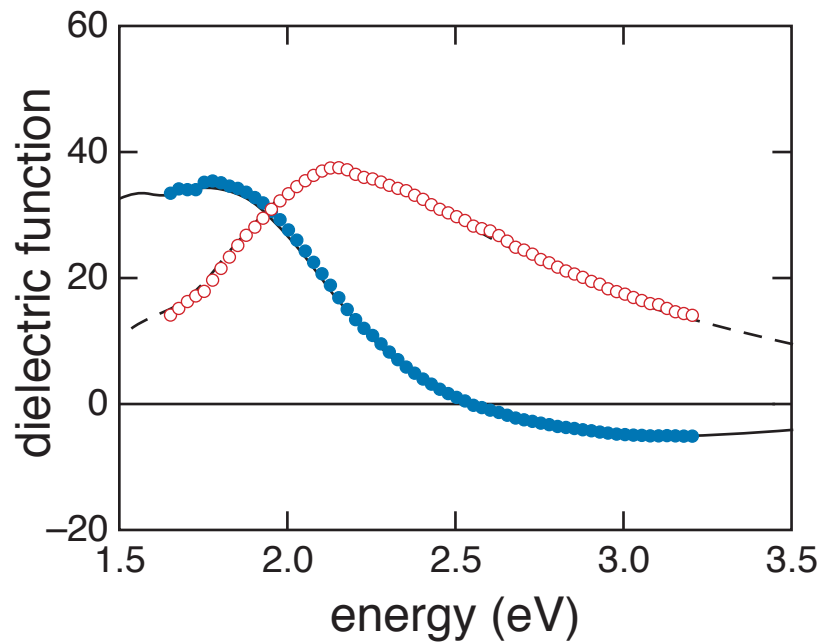
Coherent phonons

track resonance energy



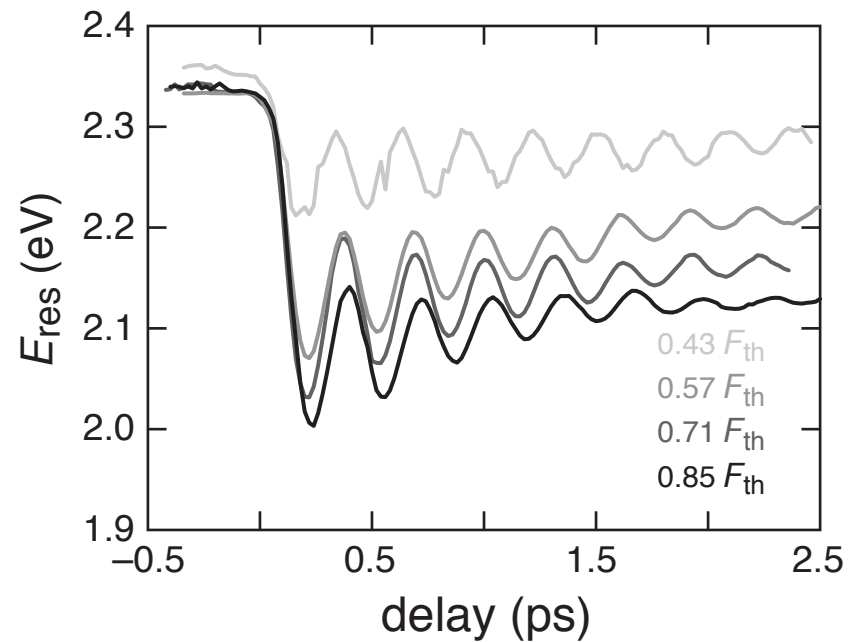
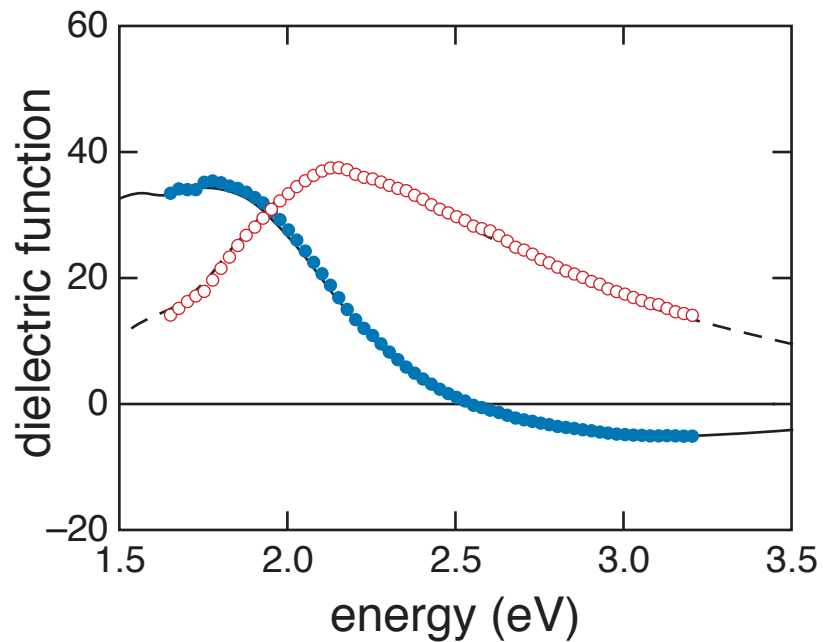
Coherent phonons

track resonance energy



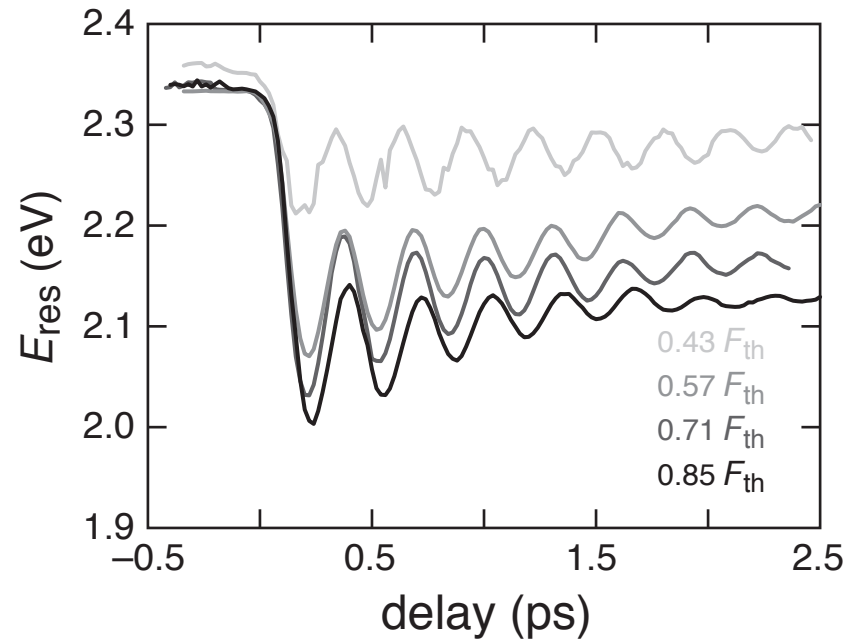
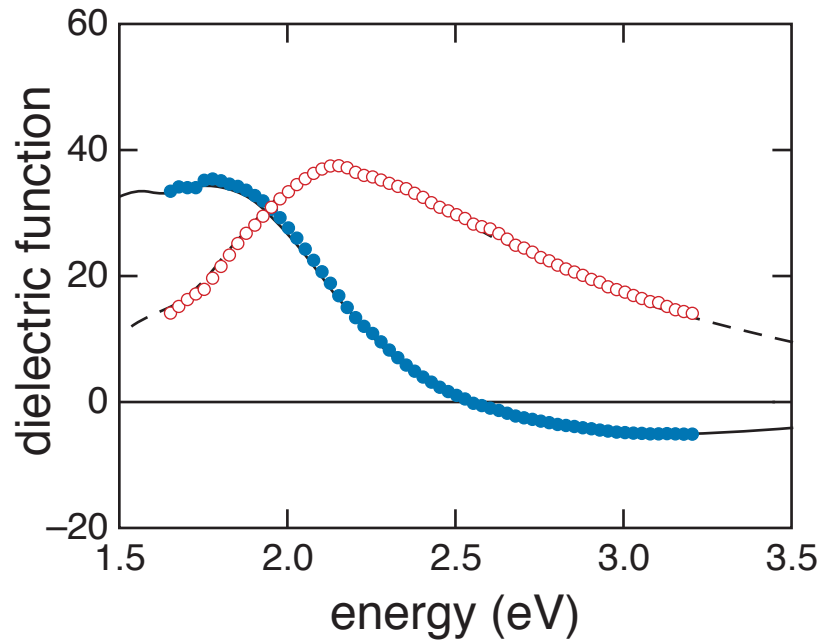
Coherent phonons

track resonance energy



Coherent phonons

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

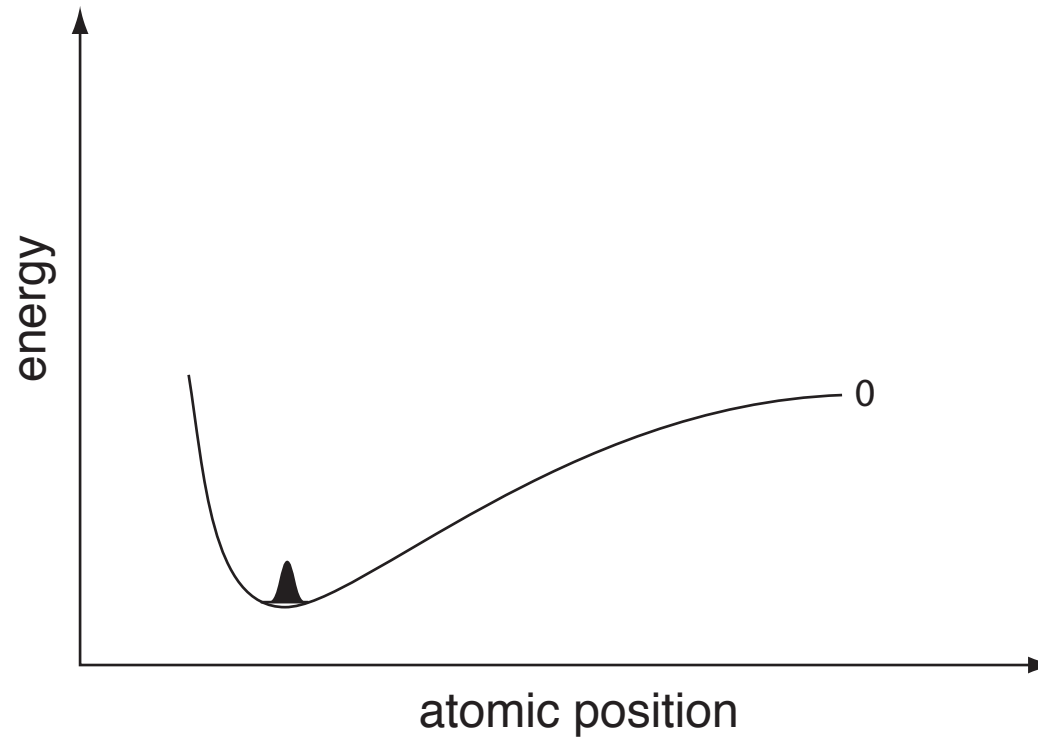


Outline

- experimental
- coherent phonons
- optical control

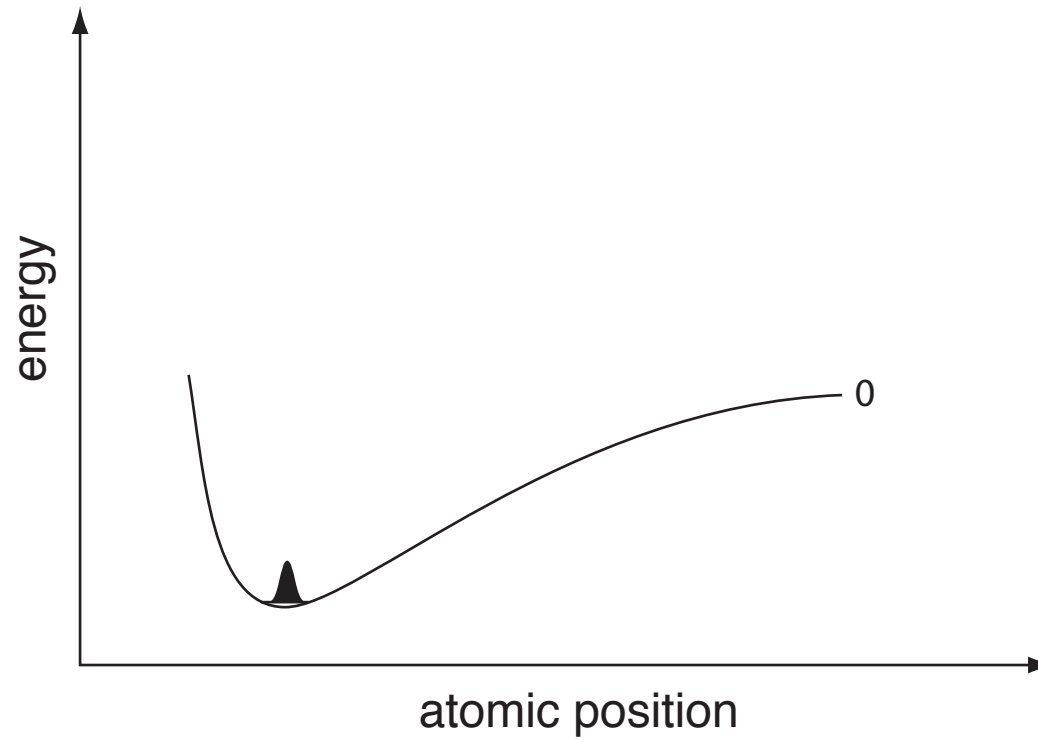
Optical control

semiclassical model of nuclear motion



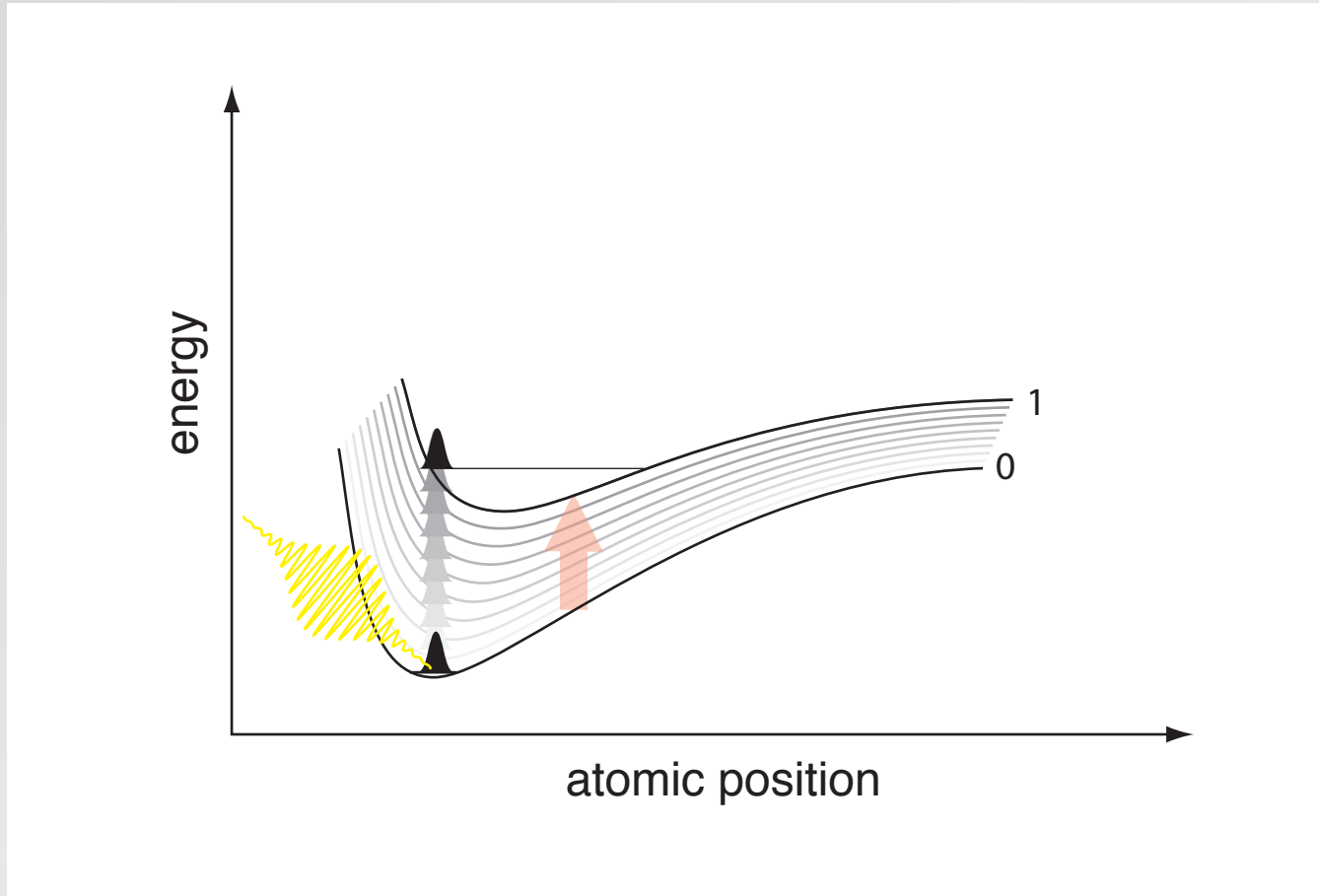
Optical control

nuclear wave packet sits at minimum



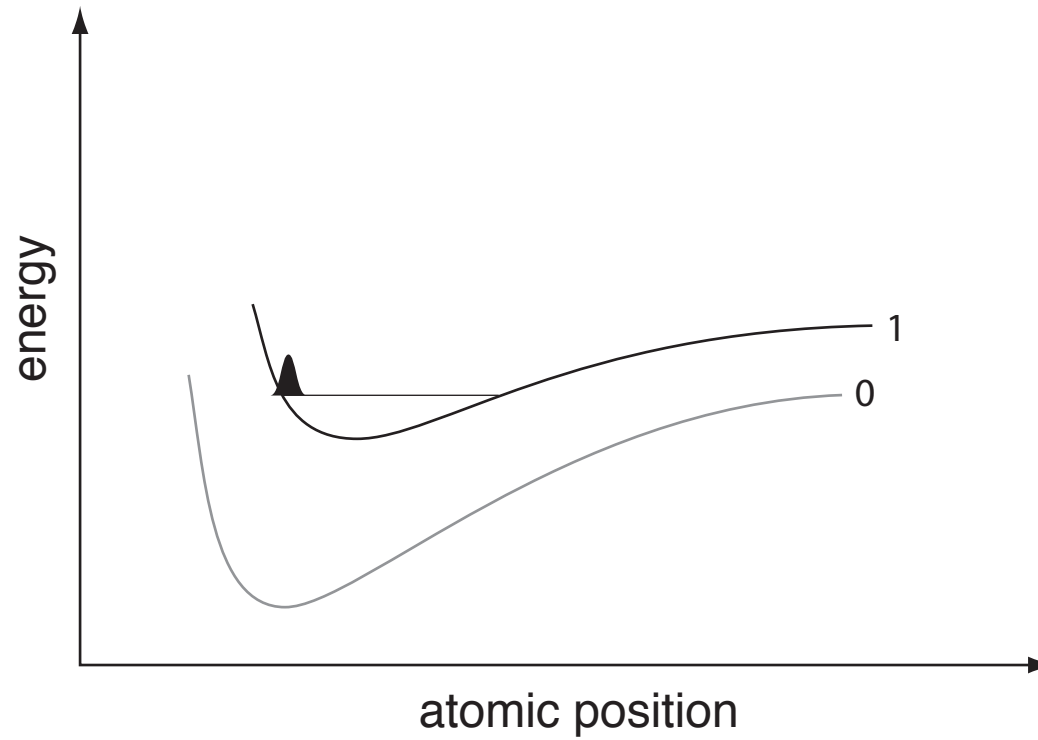
Optical control

laser pulse excites electrons, alters potential



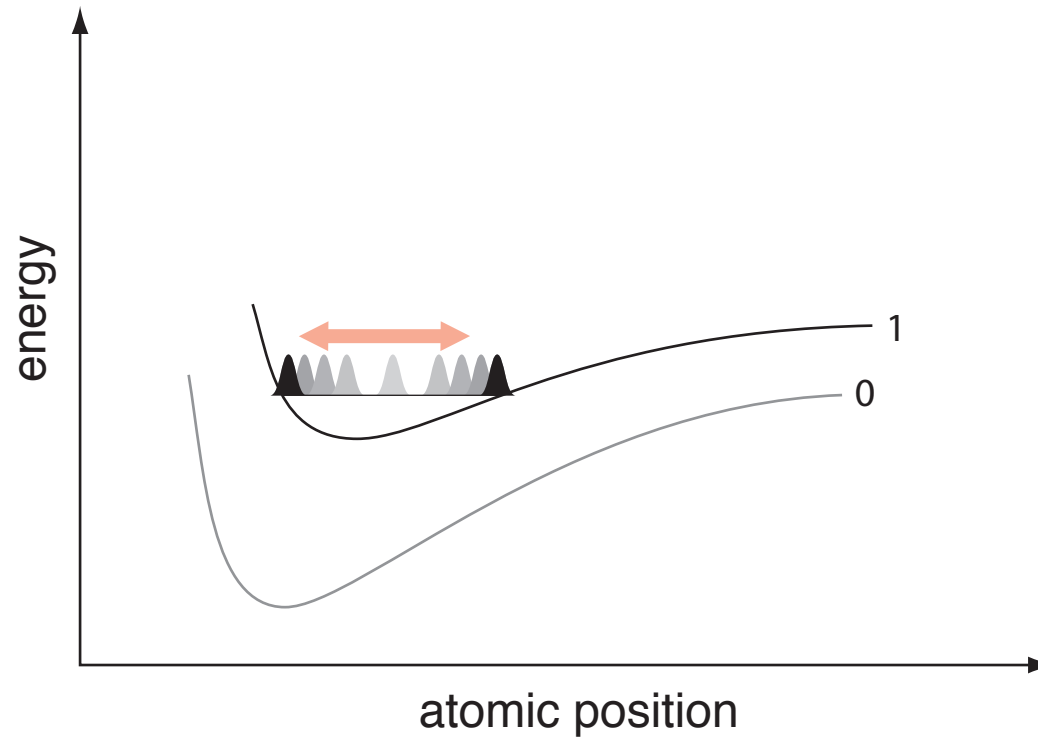
Optical control

nuclear wave packets on new potential



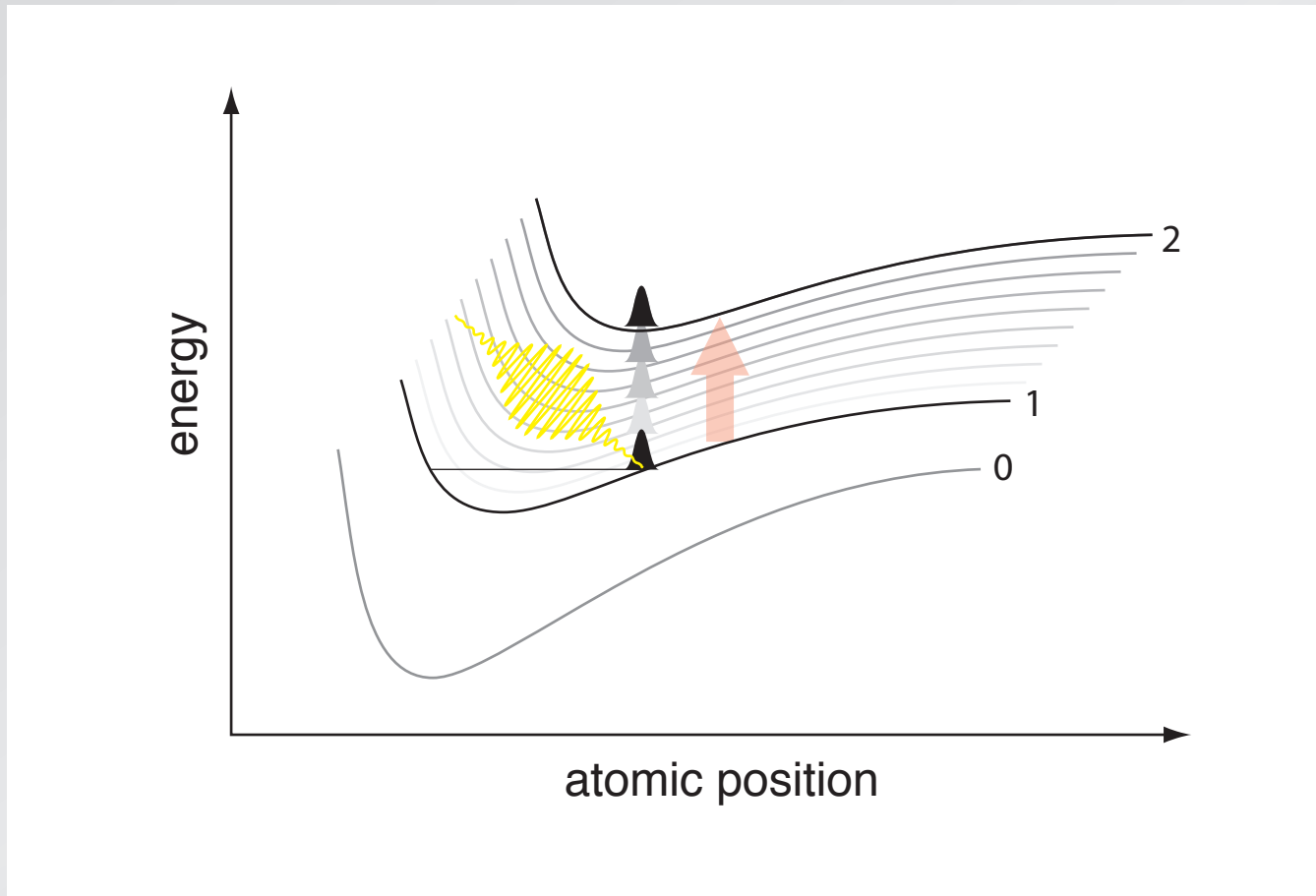
Optical control

wave packet oscillates on new potential



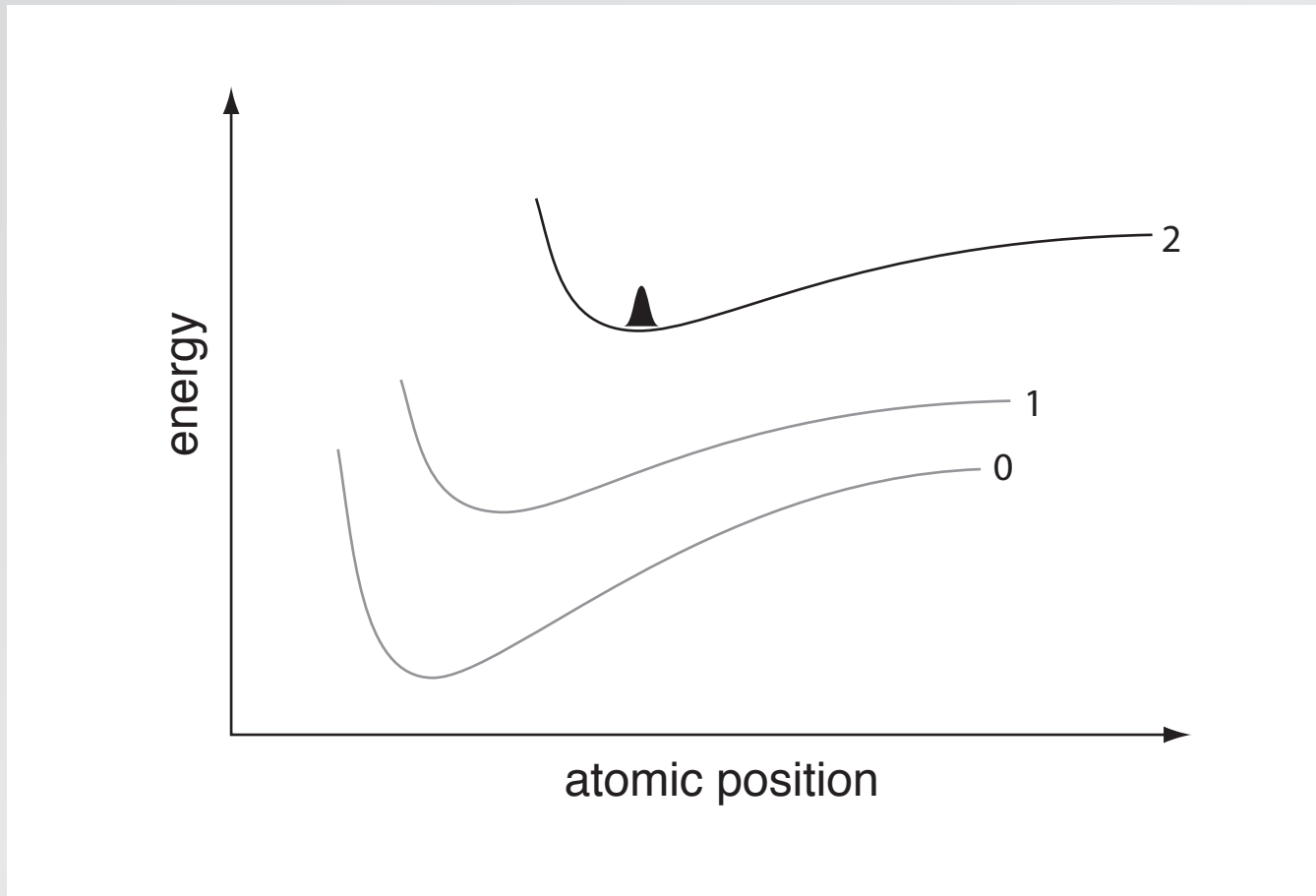
Optical control

excite again at turning point...



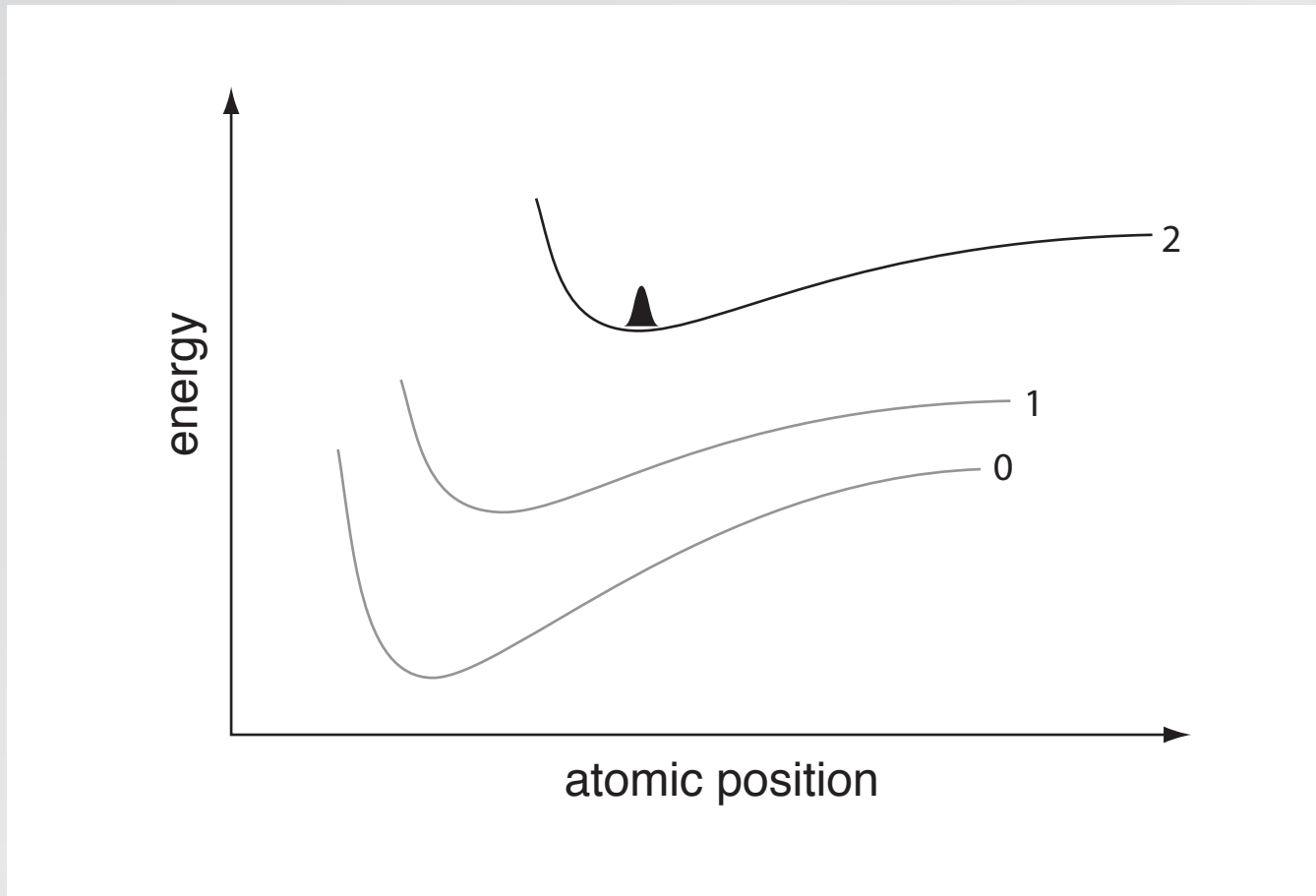
Optical control

...so wave packet lands at minimum in new potential



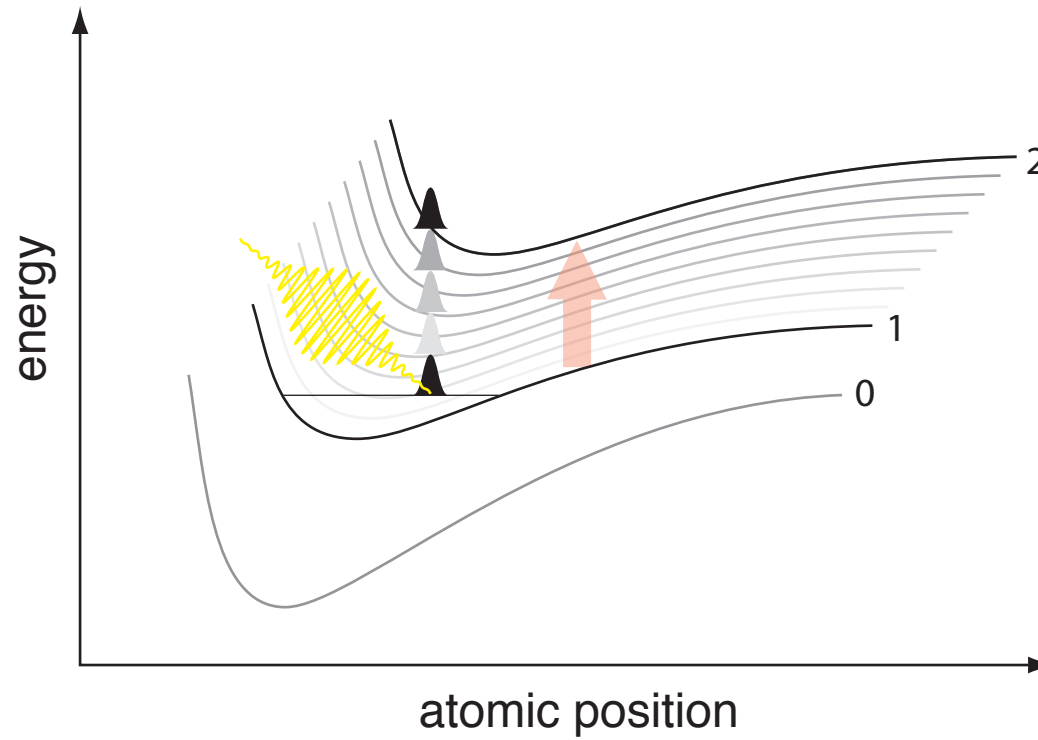
Optical control

leaving lattice displaced (without oscillations)



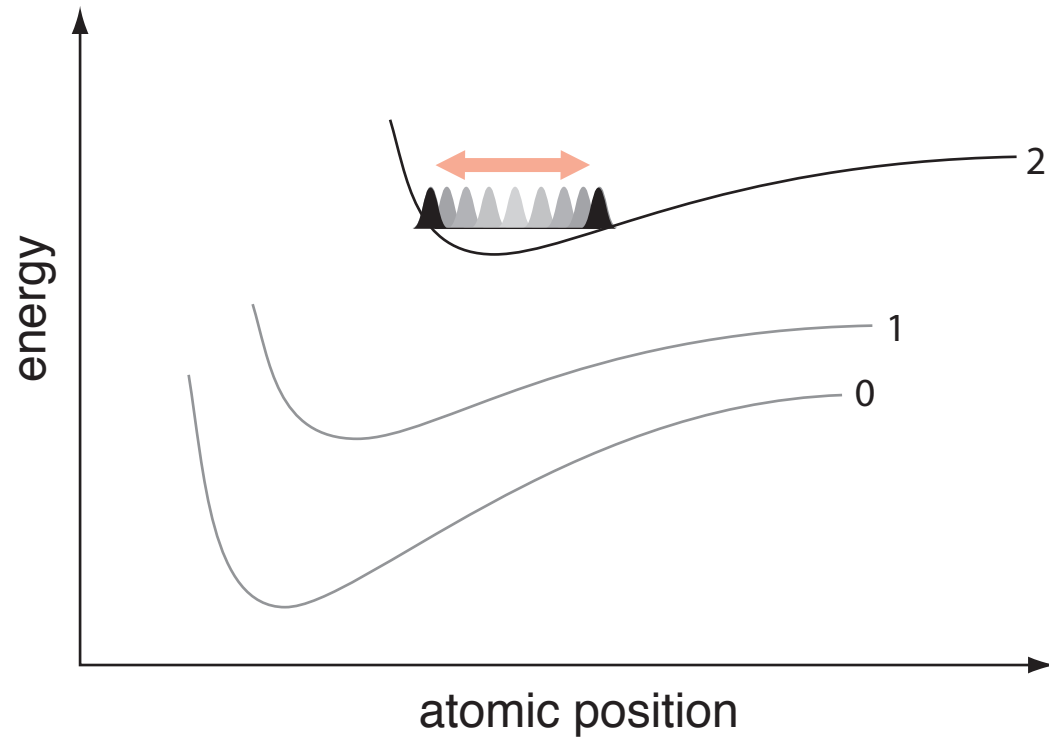
Optical control

if timing wrong...



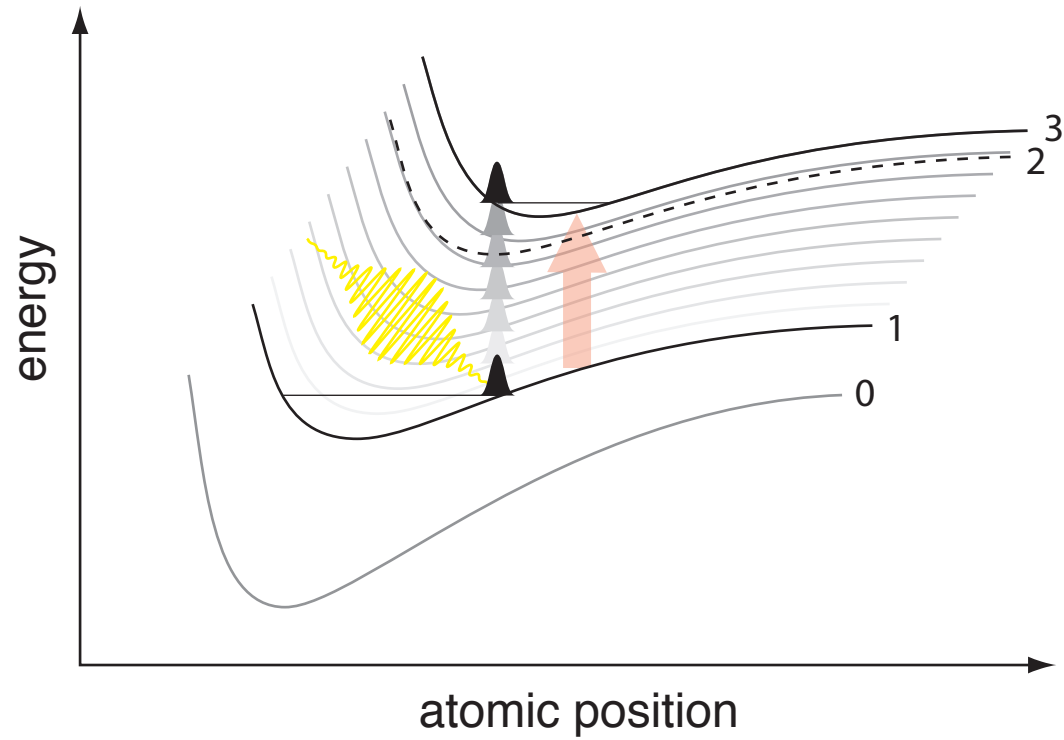
Optical control

...we get oscillations on the new potential



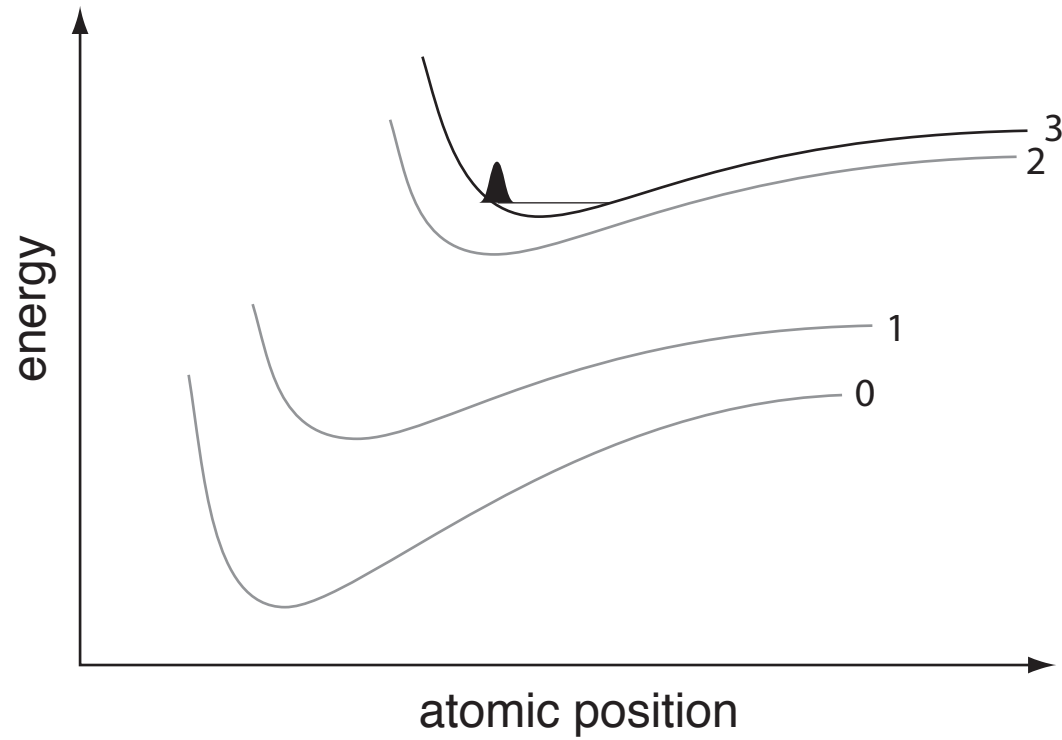
Optical control

if fluence wrong...



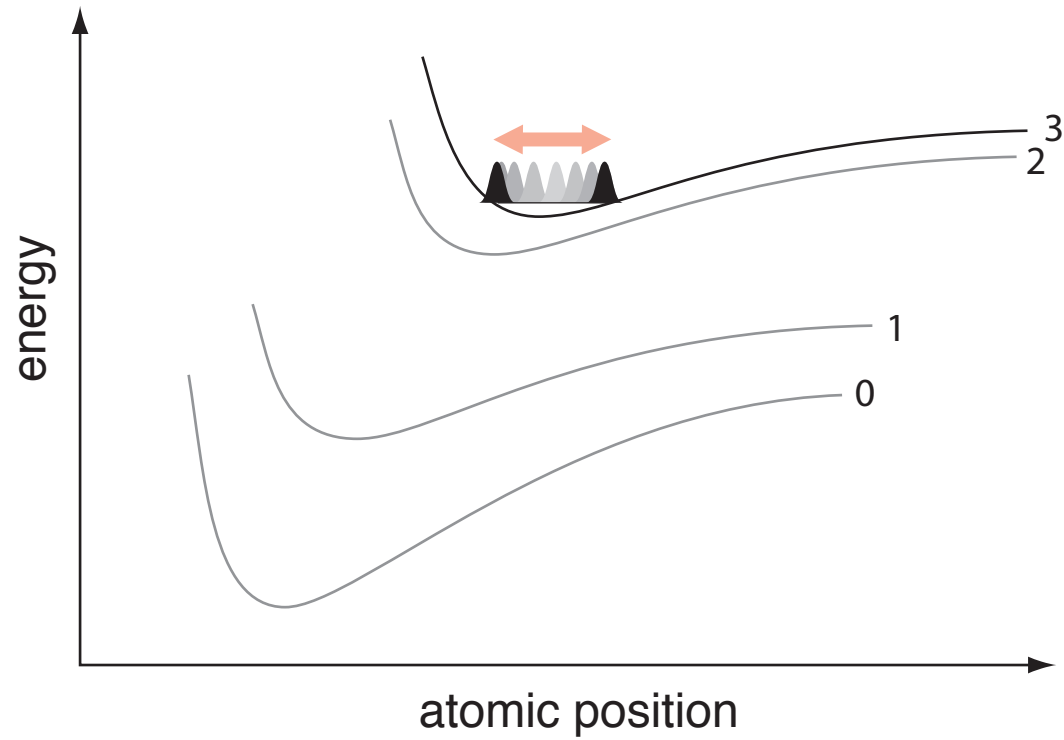
Optical control

excite to other potential surface...



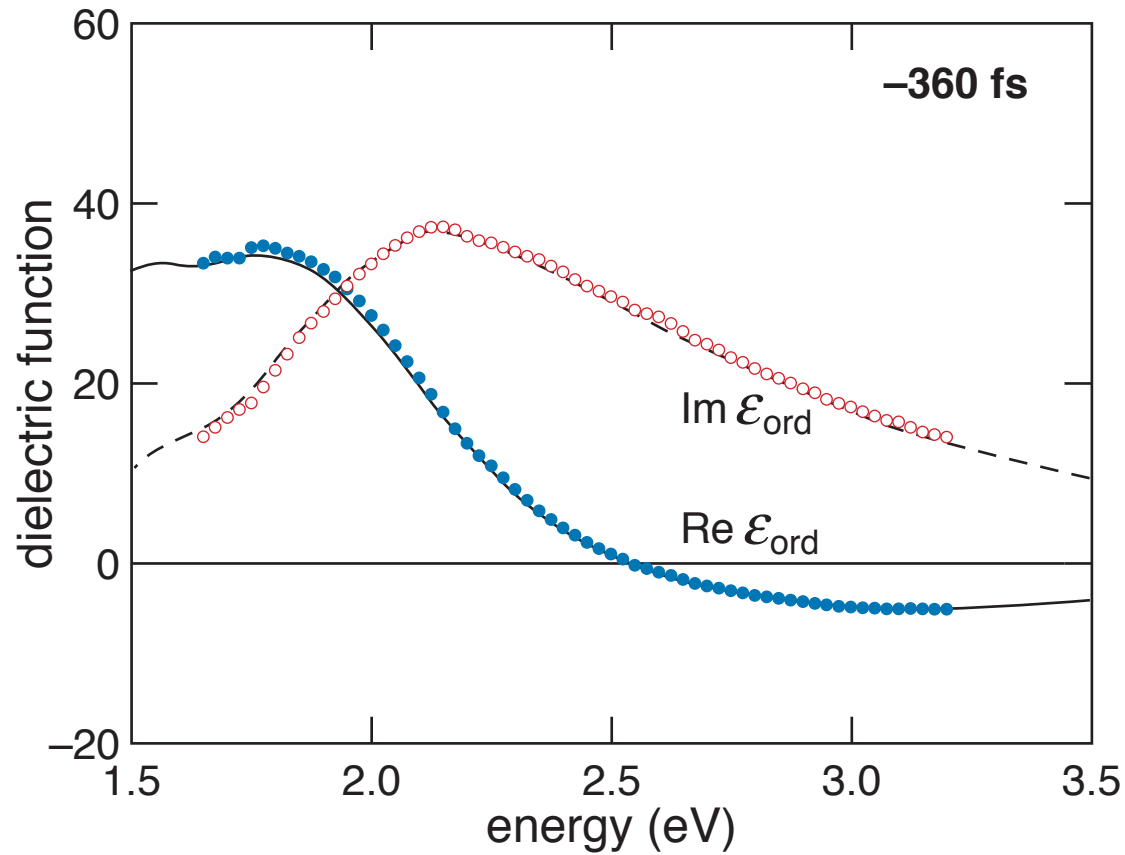
Optical control

...and wave packet oscillates



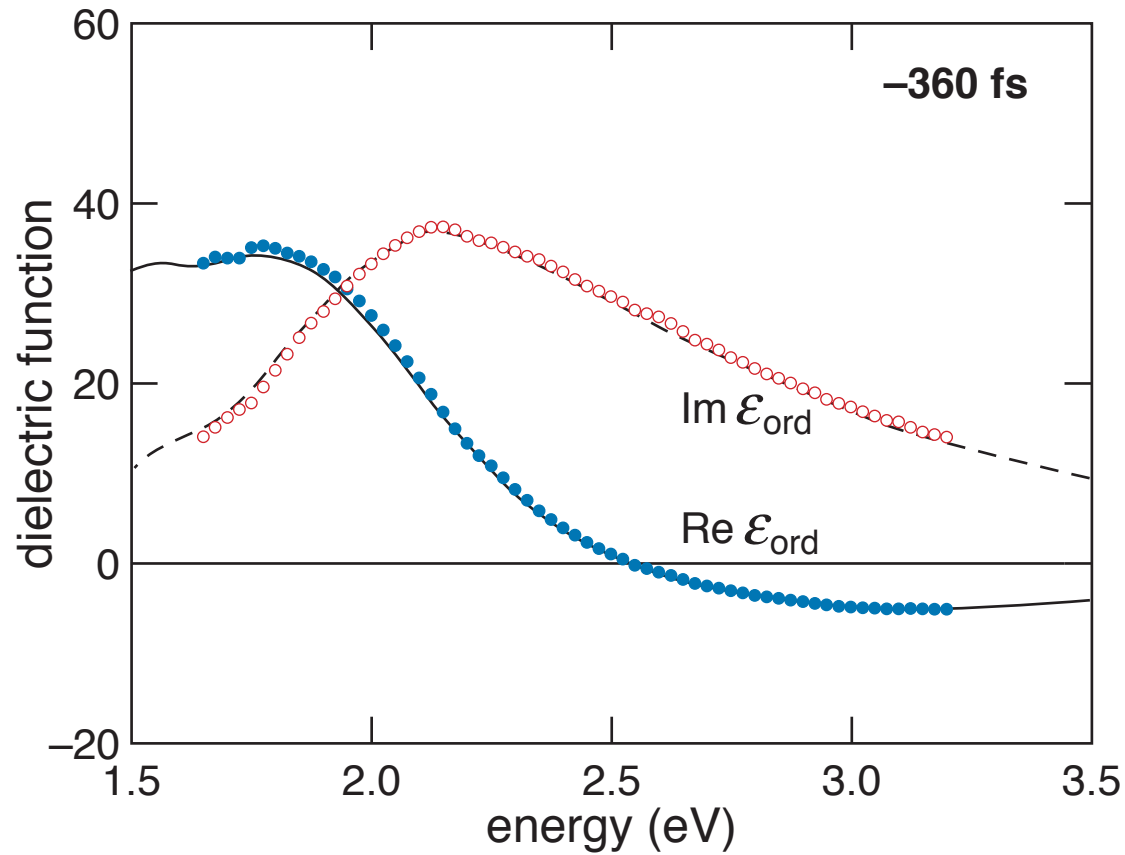
Optical control

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



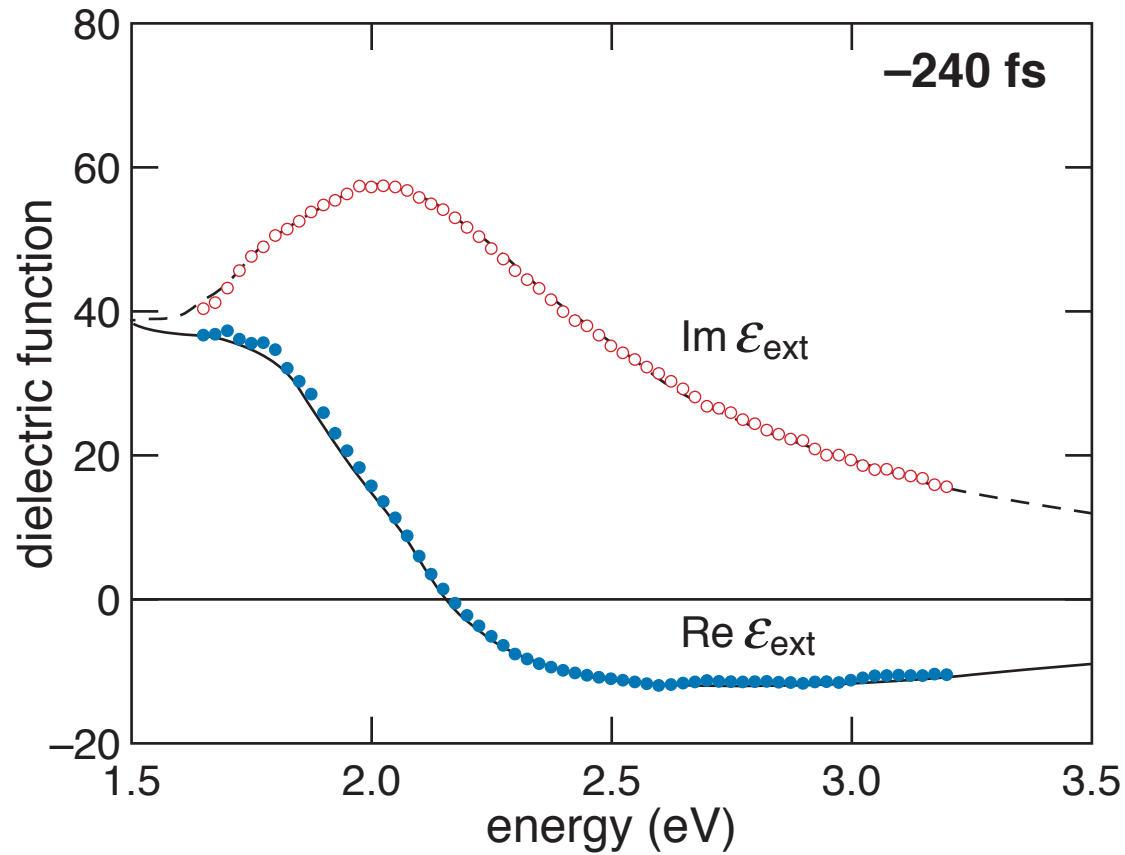
Optical control

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



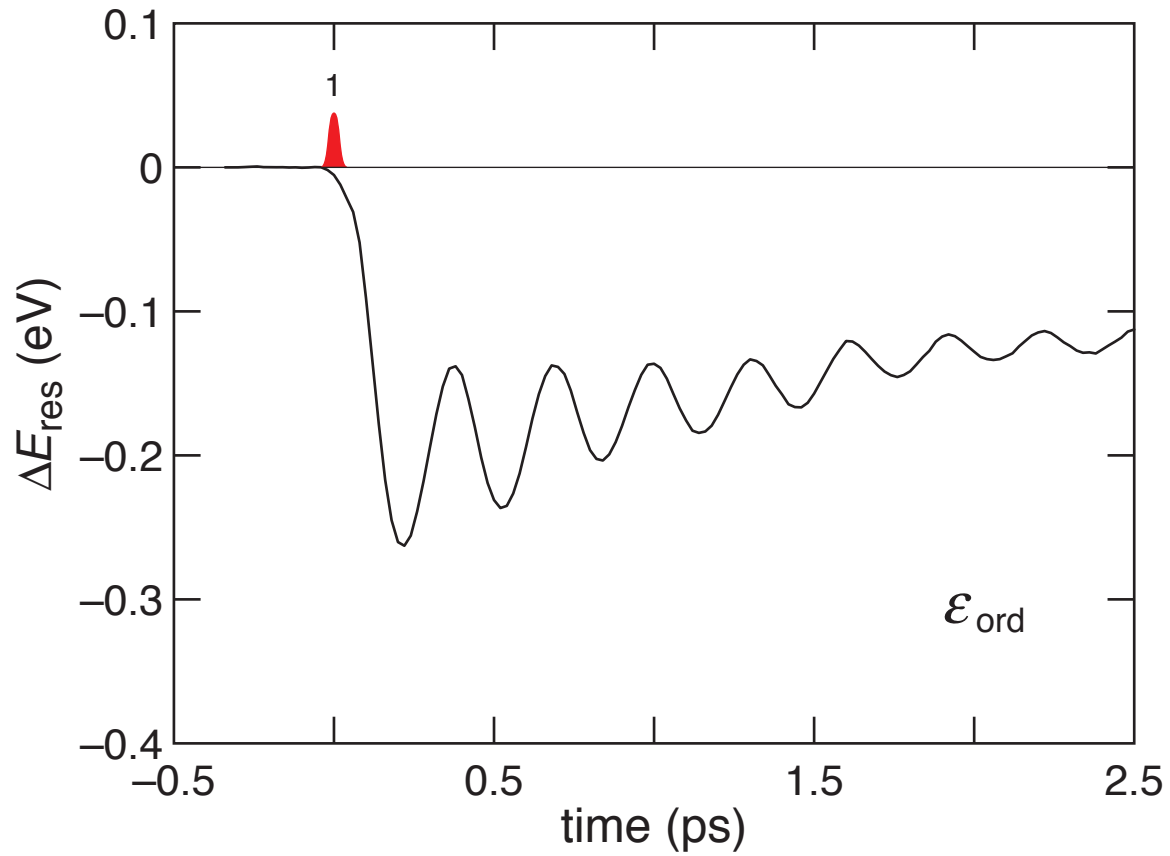
Optical control

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



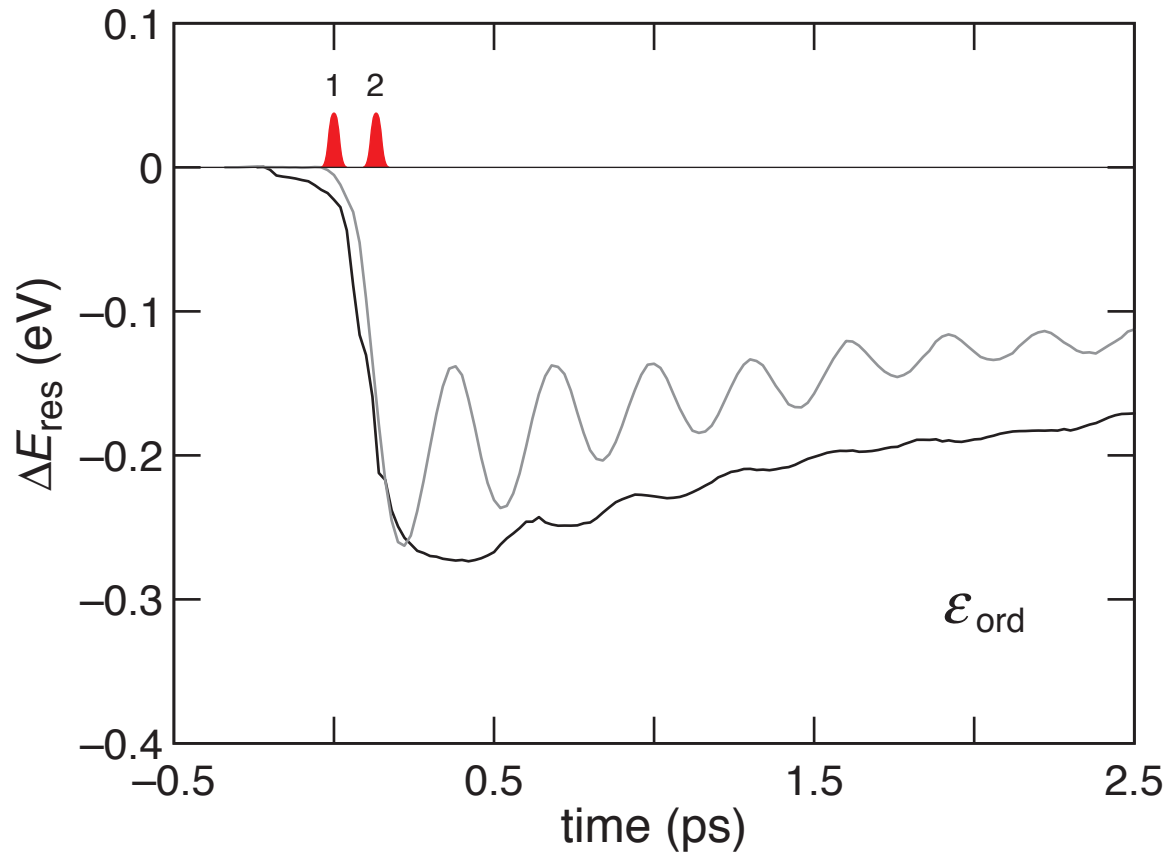
Optical control

$$F_1 = 0.57 F_{th}$$



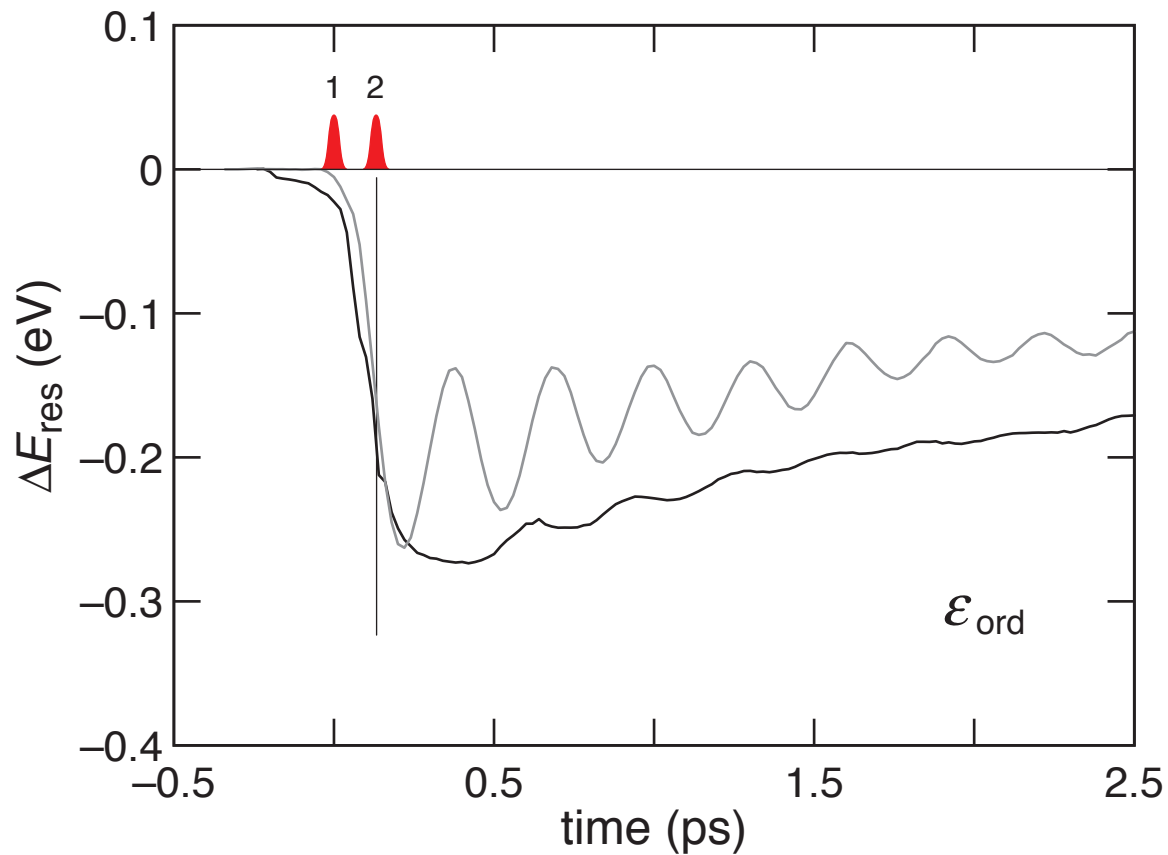
Optical control

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



Optical control

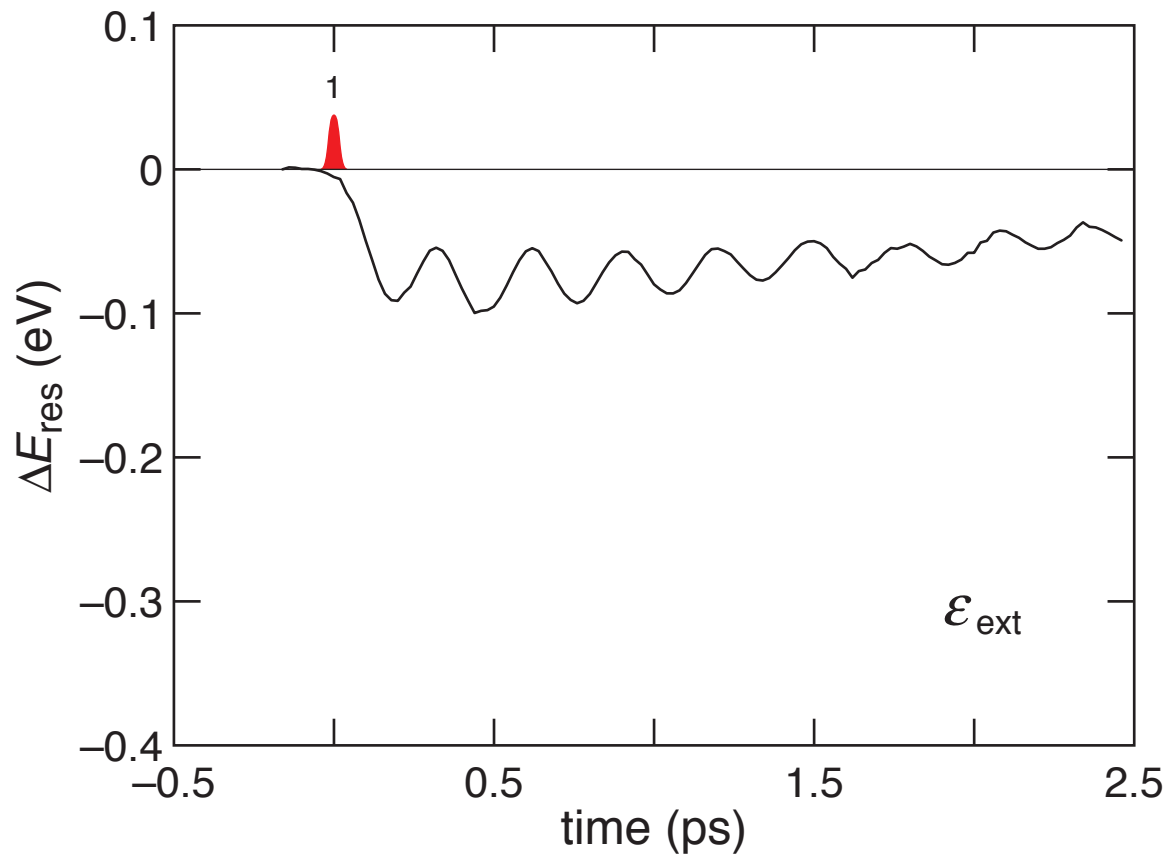
$$F_1 = 0.57 F_{\text{th}} \quad F_2 = 0.46 F_{\text{th}} \quad \tau = 133 \text{ fs}$$



...but delay a bit less than half a period

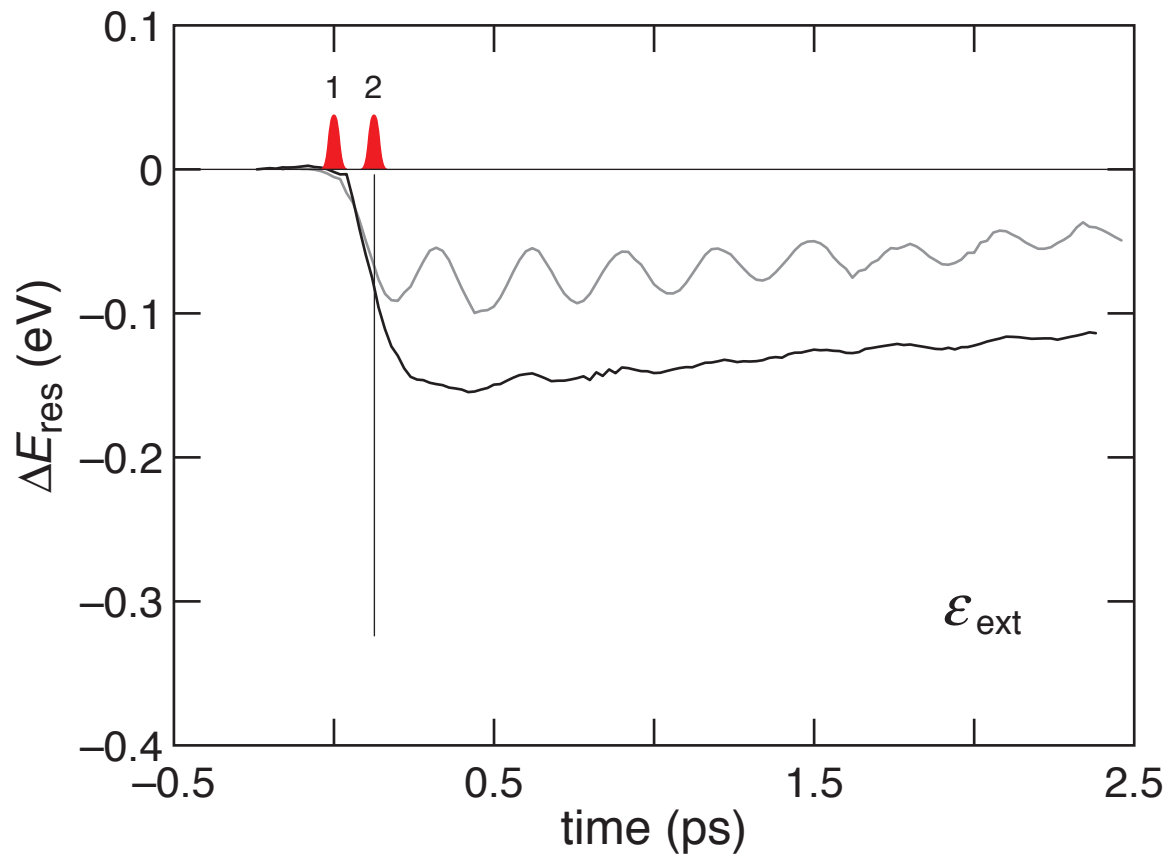
Optical control

$$F_1 = 0.43 F_{\text{th}}$$



Optical control

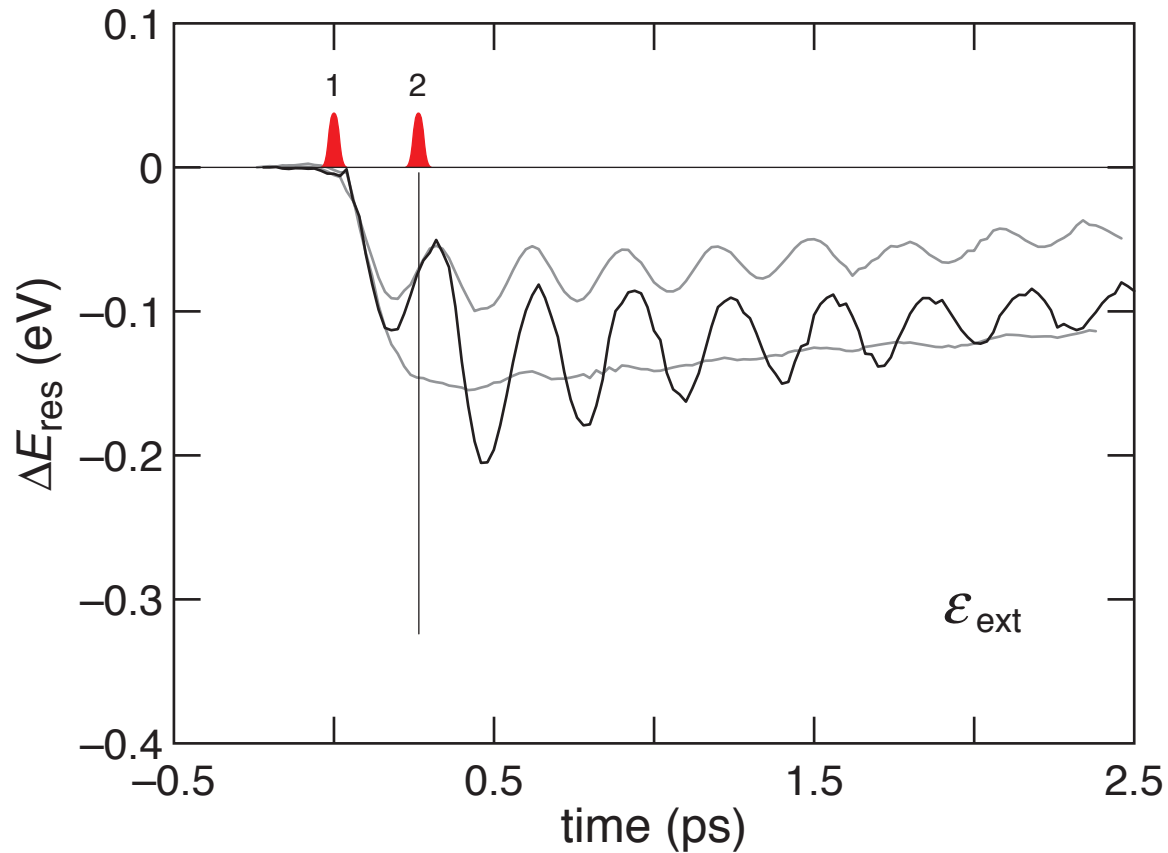
$$F_1 = 0.43 F_{th} \quad F_2 = 0.33 F_{th} \quad \tau = 127 \text{ fs}$$



delay again less than half a period

Optical control

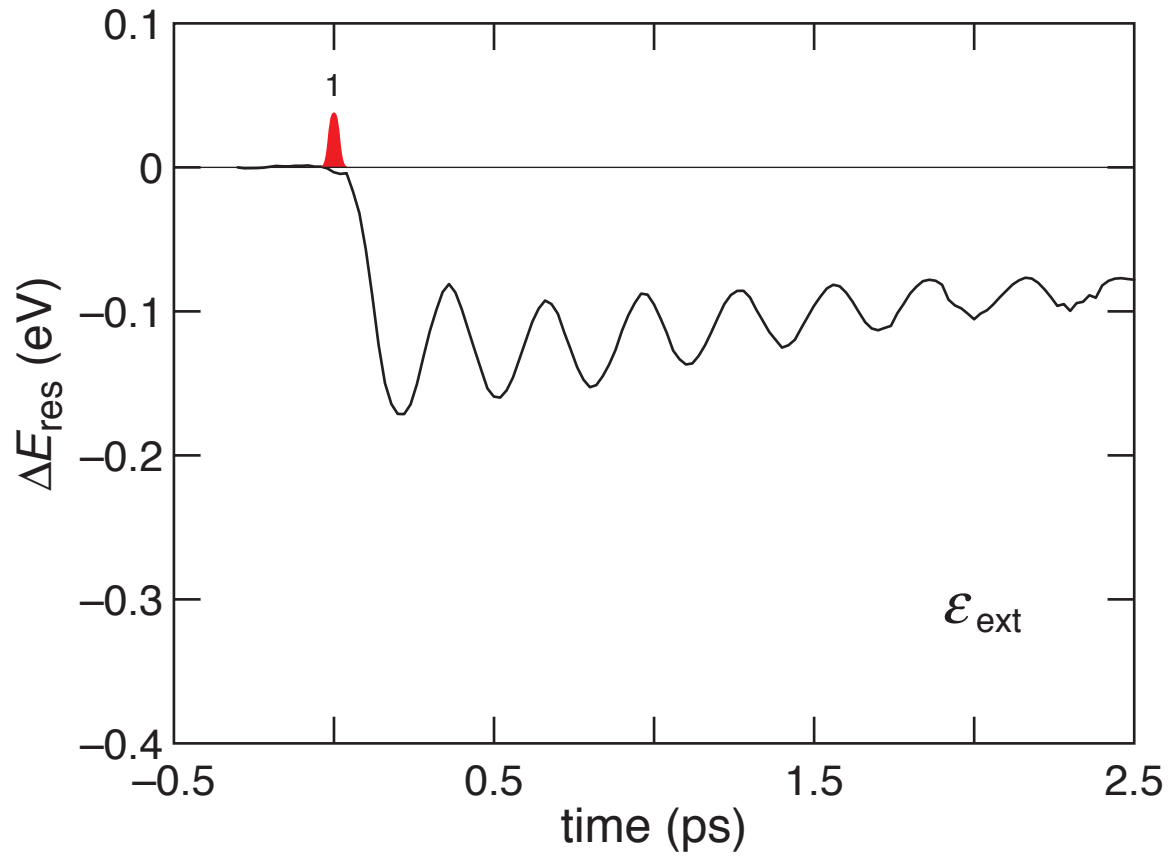
$$F_1 = 0.43 F_{\text{th}} \quad F_2 = 0.33 F_{\text{th}} \quad \tau = 267 \text{ fs}$$



delay a bit less than a period

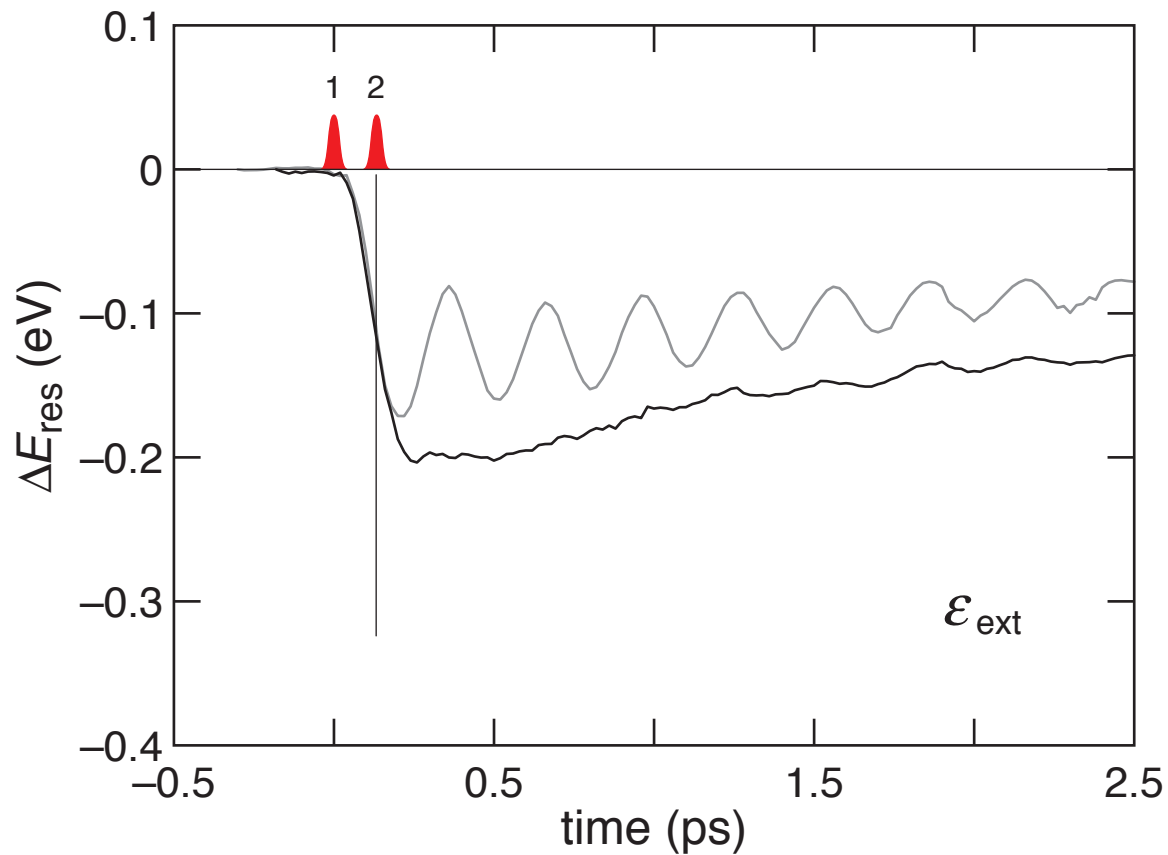
Optical control

$$F_1 = 0.57 F_{\text{th}}$$



Optical control

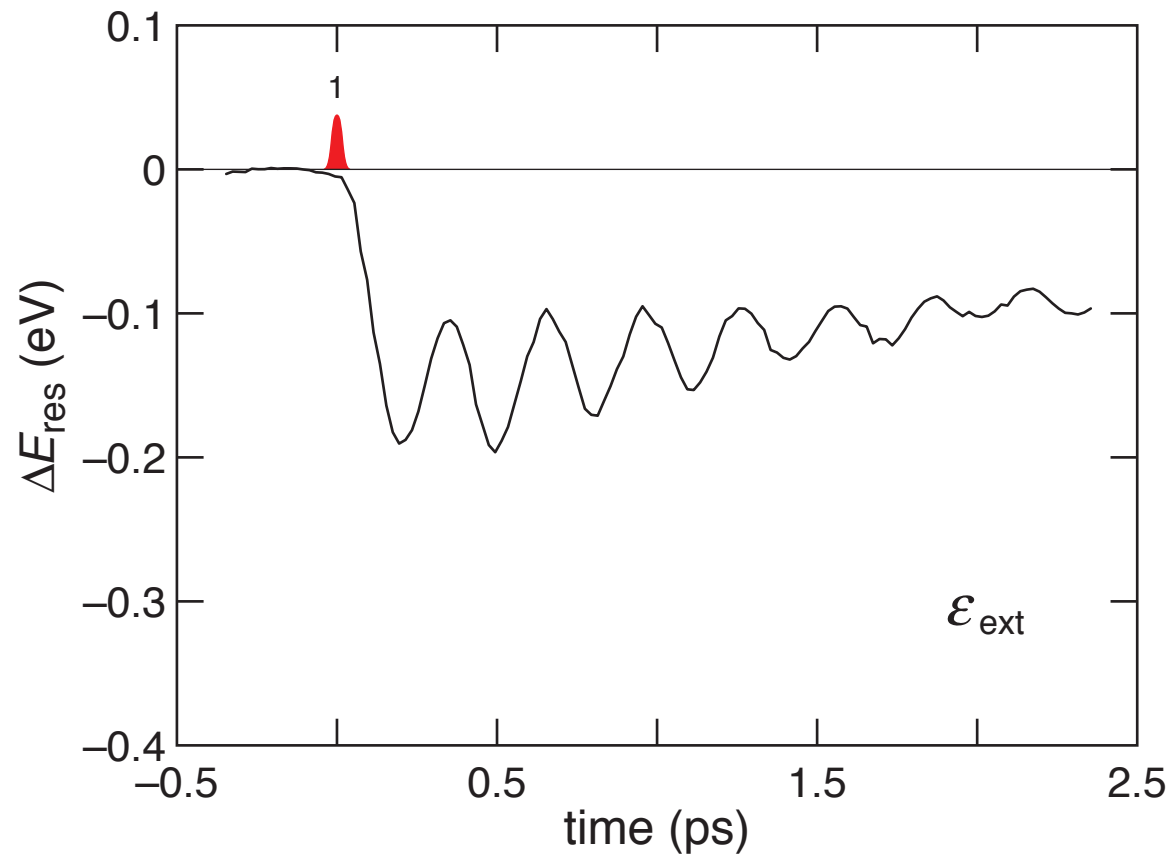
$$F_1 = 0.57 F_{\text{th}} \quad F_2 = 0.45 F_{\text{th}} \quad \tau = 133 \text{ fs}$$



cancellation on first swing

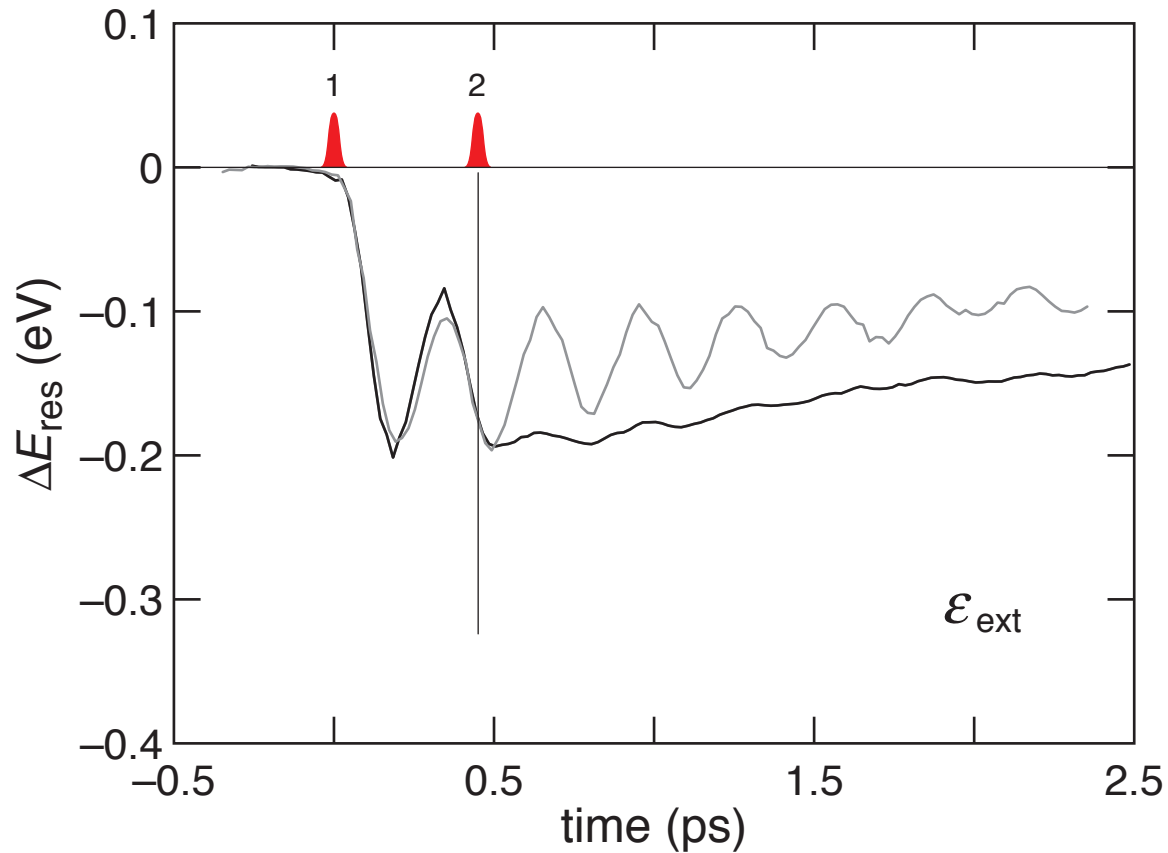
Optical control

$$F_1 = 0.71 F_{\text{th}}$$



Optical control

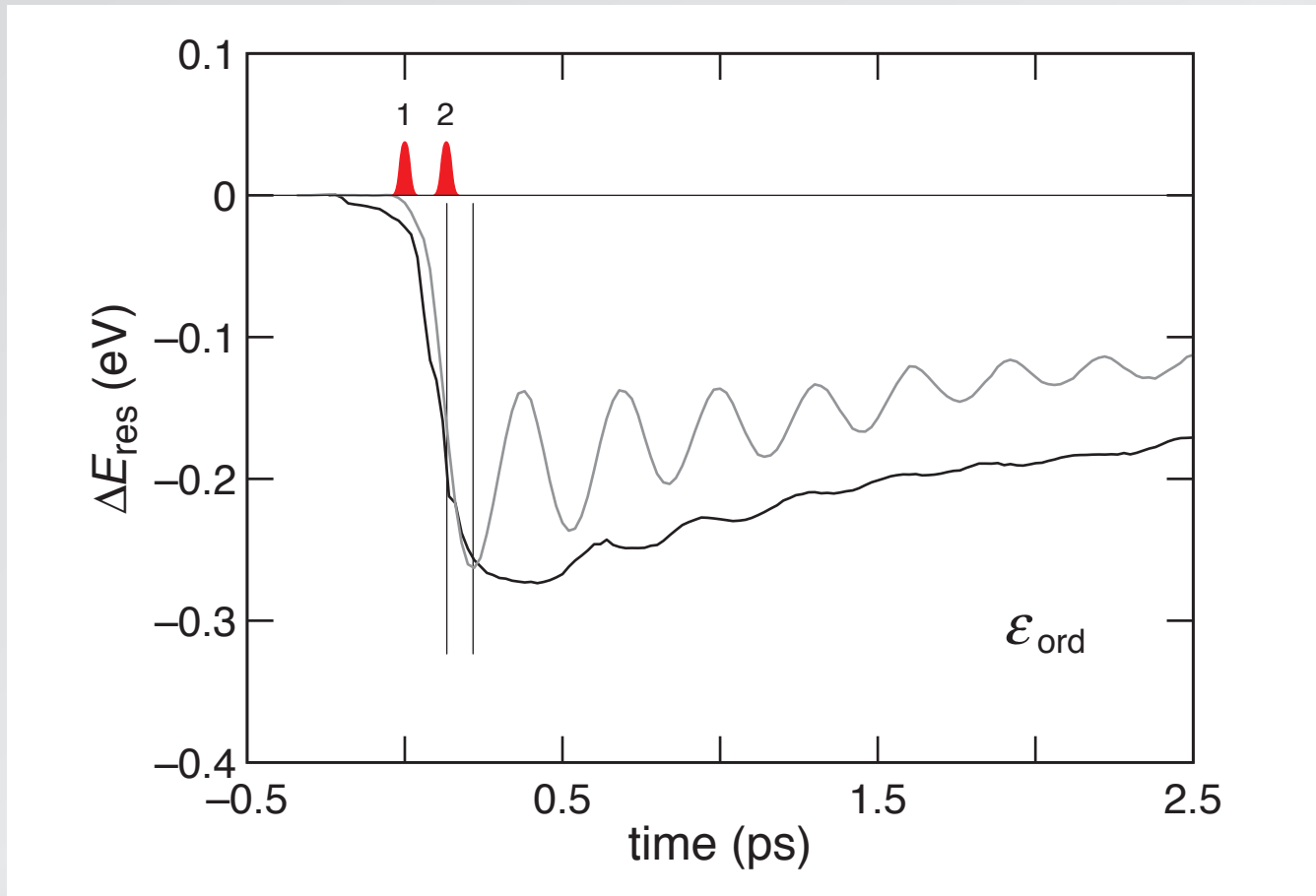
$$F_1 = 0.71 F_{\text{th}} \quad F_2 = 0.34 F_{\text{th}} \quad \tau = 467 \text{ fs}$$



cancellation on second swing

Optical control

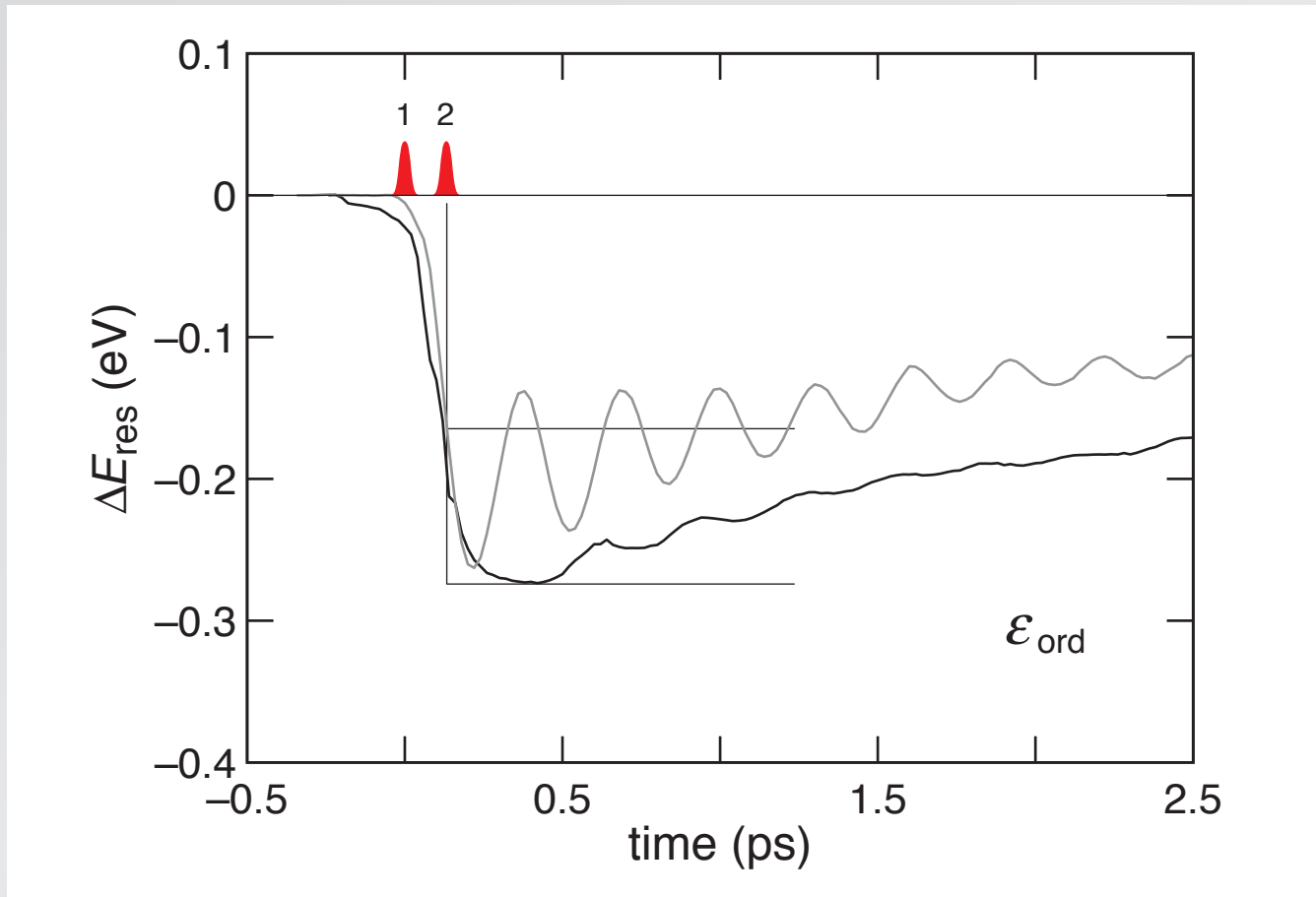
two things to note:



second pulse always earlier than expected, and...

Optical control

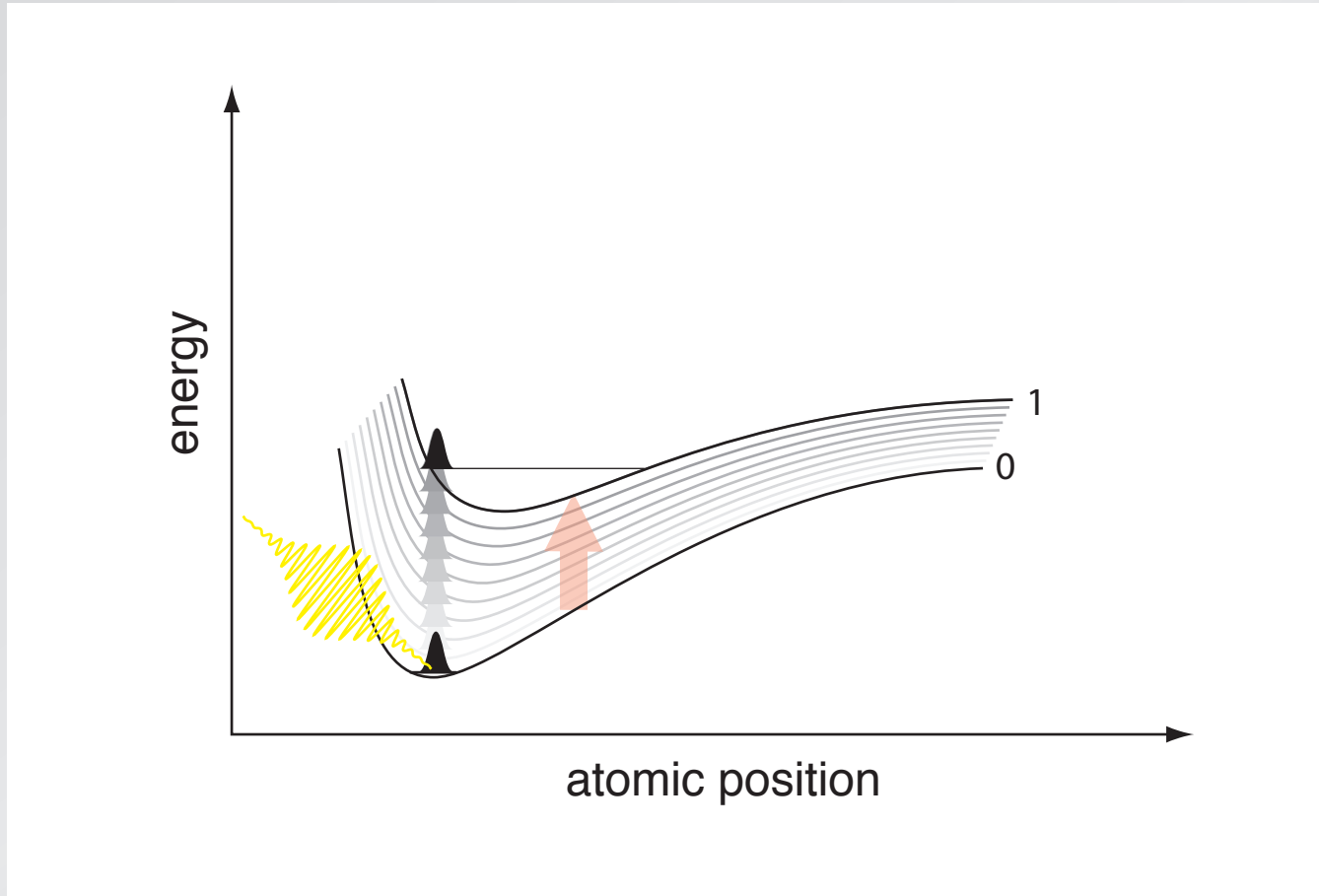
two things to note:



... resonance continues to shift after second pulse

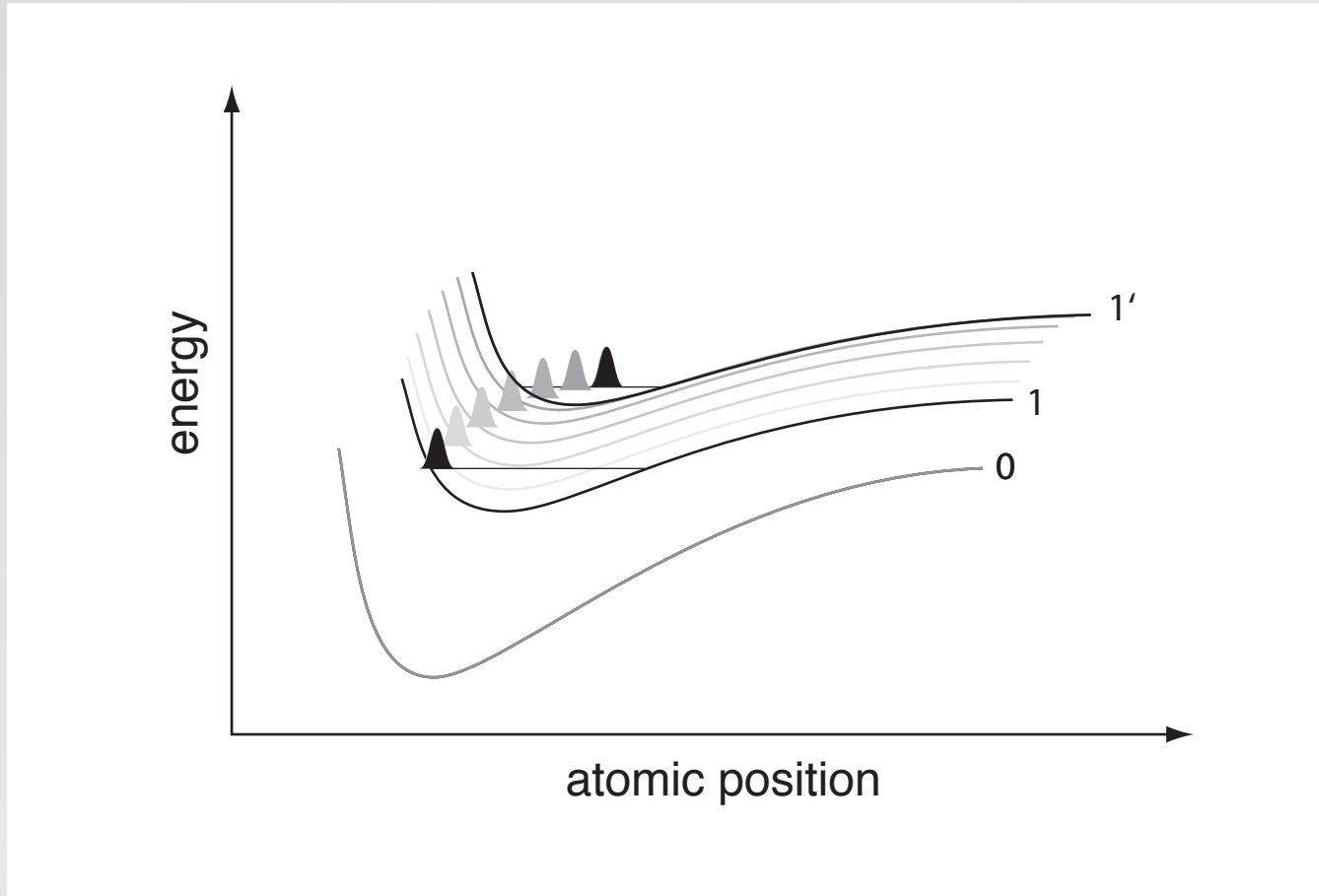
Optical control

excited electrons 'instantaneously' alter potential



Optical control

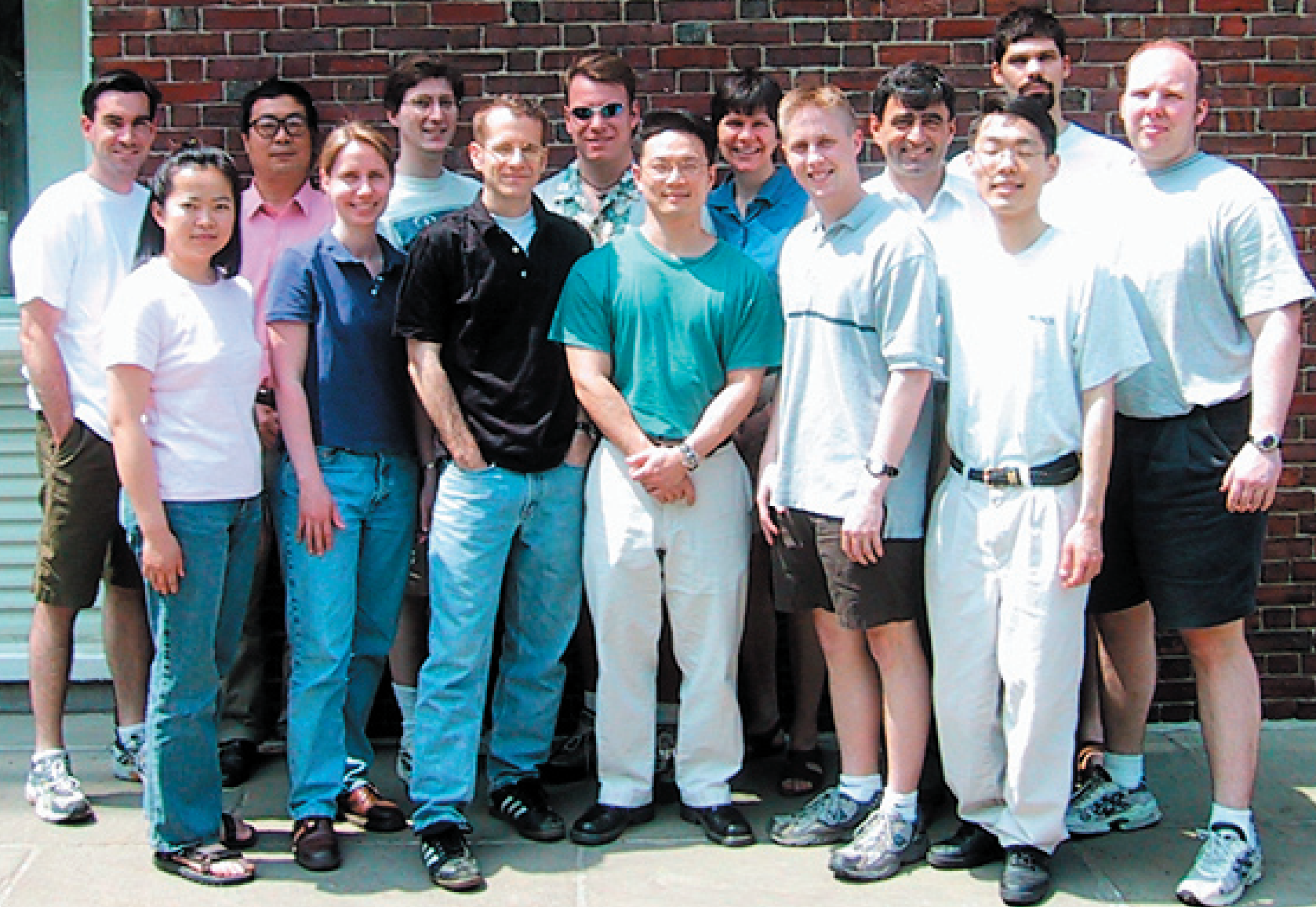
but nuclear rearrangement also alters potential



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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