FO.R.T.H. Institute of Electronic Structure and Laser

Ultrafast dynamics in highly excited semiconductors

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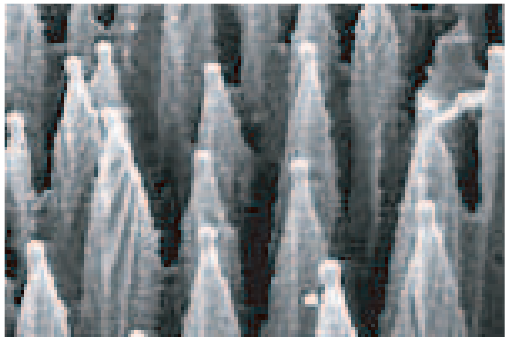
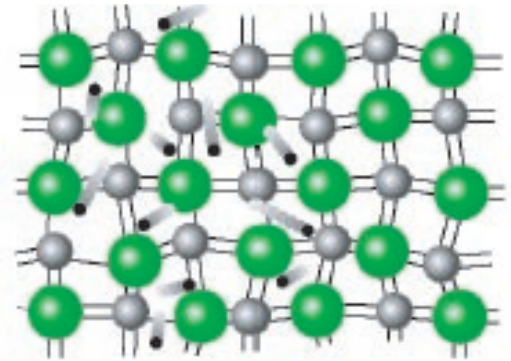
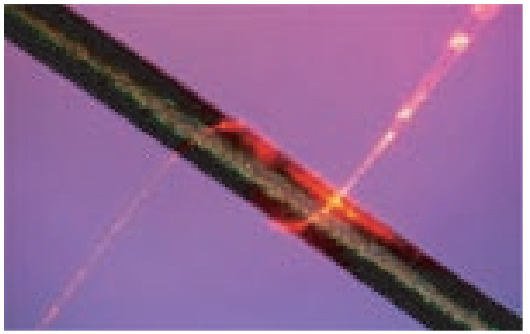
black silicon



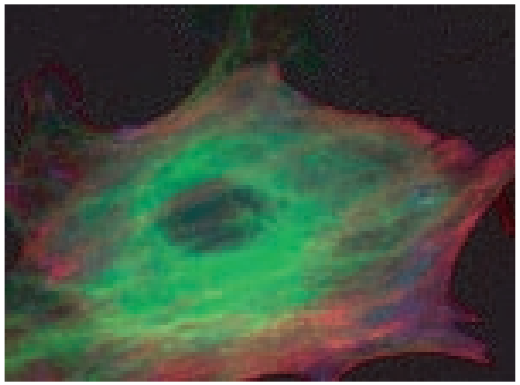
ultrafast dynamics



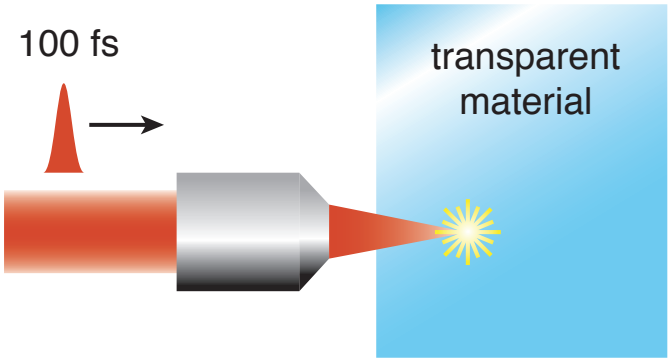
silica nanowires



nanosurgery



micromachining



science education

Outline

1. Introduction - Interaction of light with solids

2. Experimental process and technique

3. Excitation of high amplitude coherent phonons

4. Optical control of coherent phonons

Outline

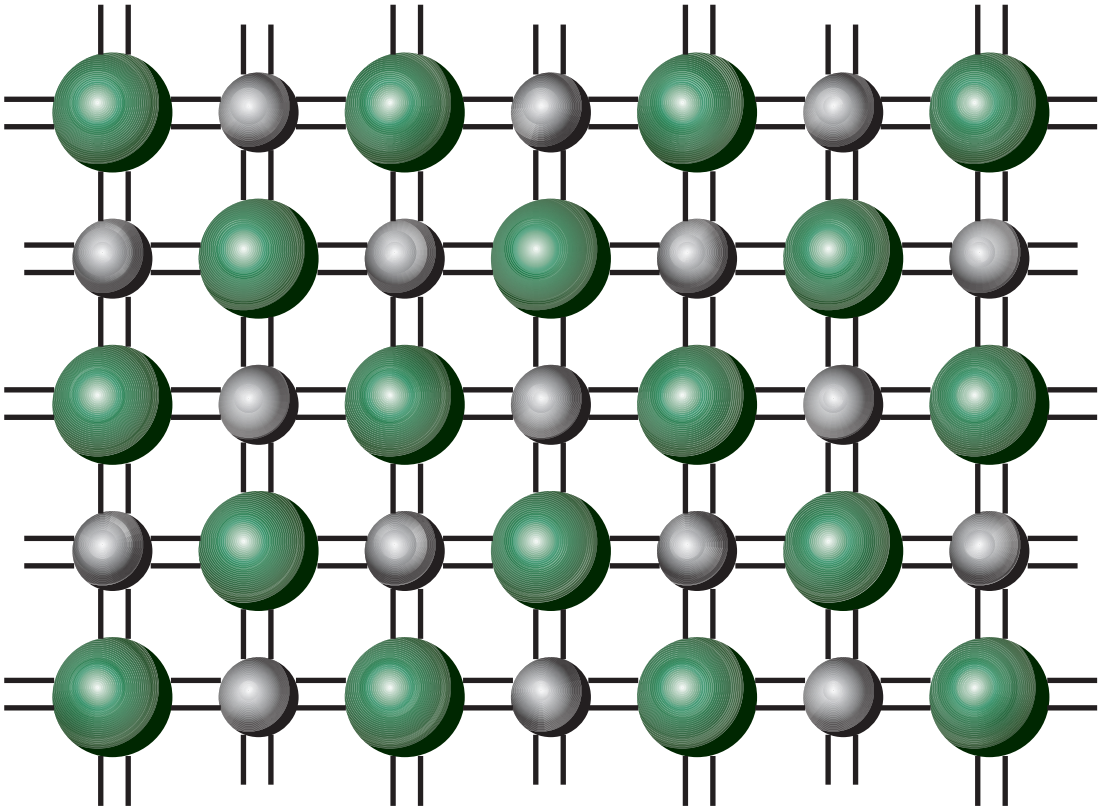
1. **Introduction - Interaction of light with solids**

2. **Experimental process and technique**

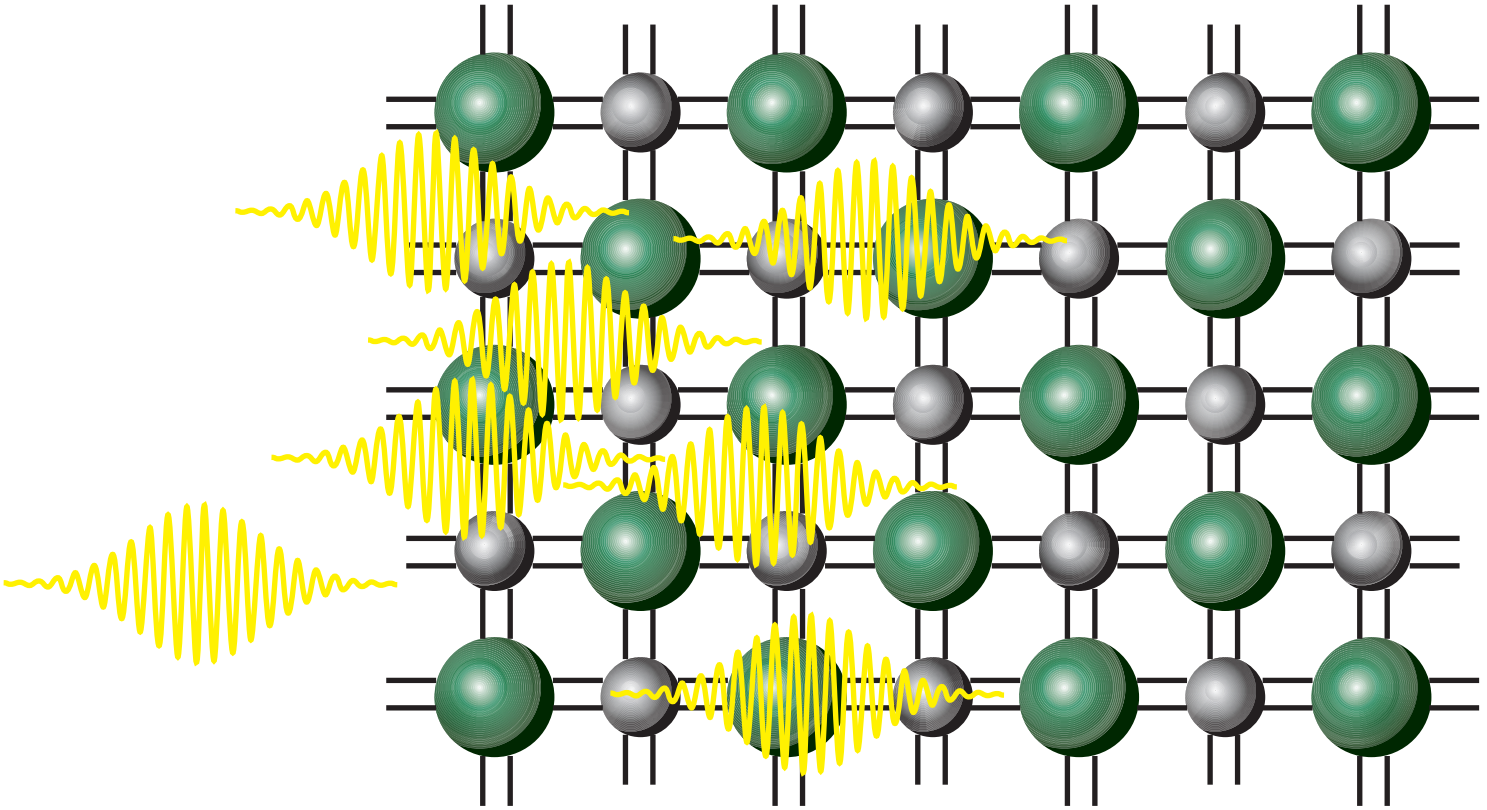
3. **Excitation of high amplitude coherent phonons**

4. **Optical control of coherent phonons**

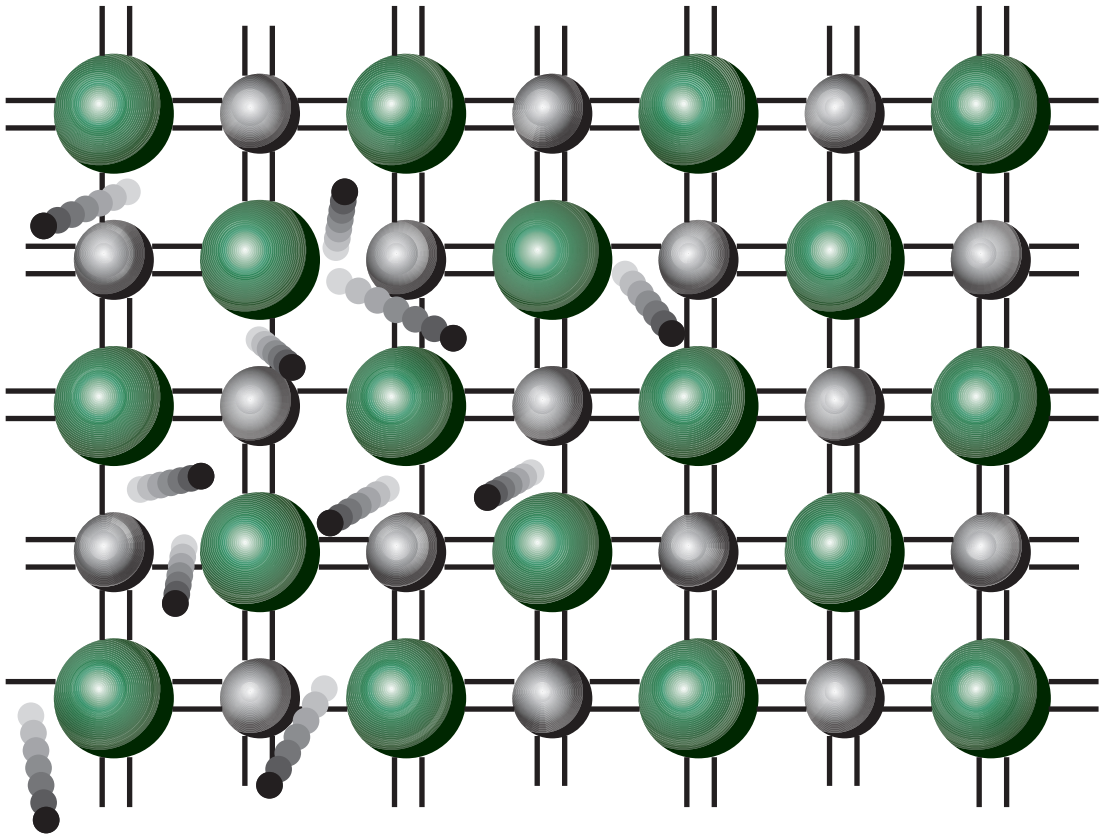
Interaction of femtosecond pulses with solids



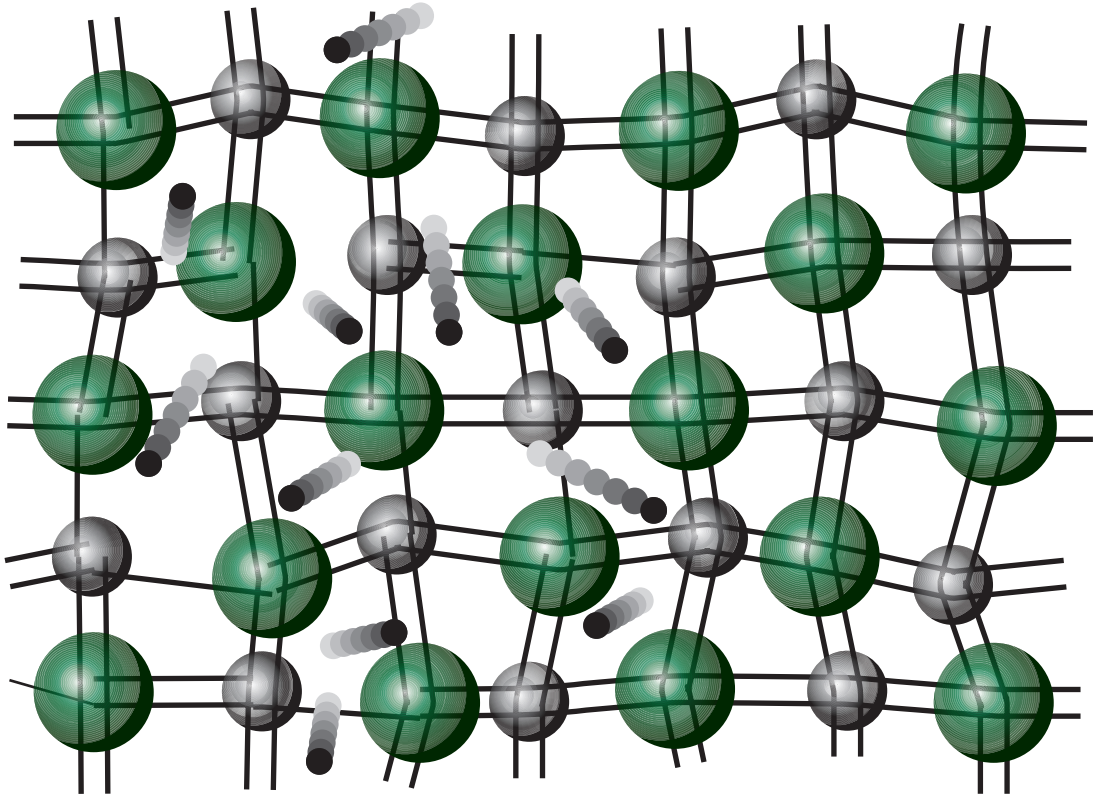
Photons excite valence electrons...



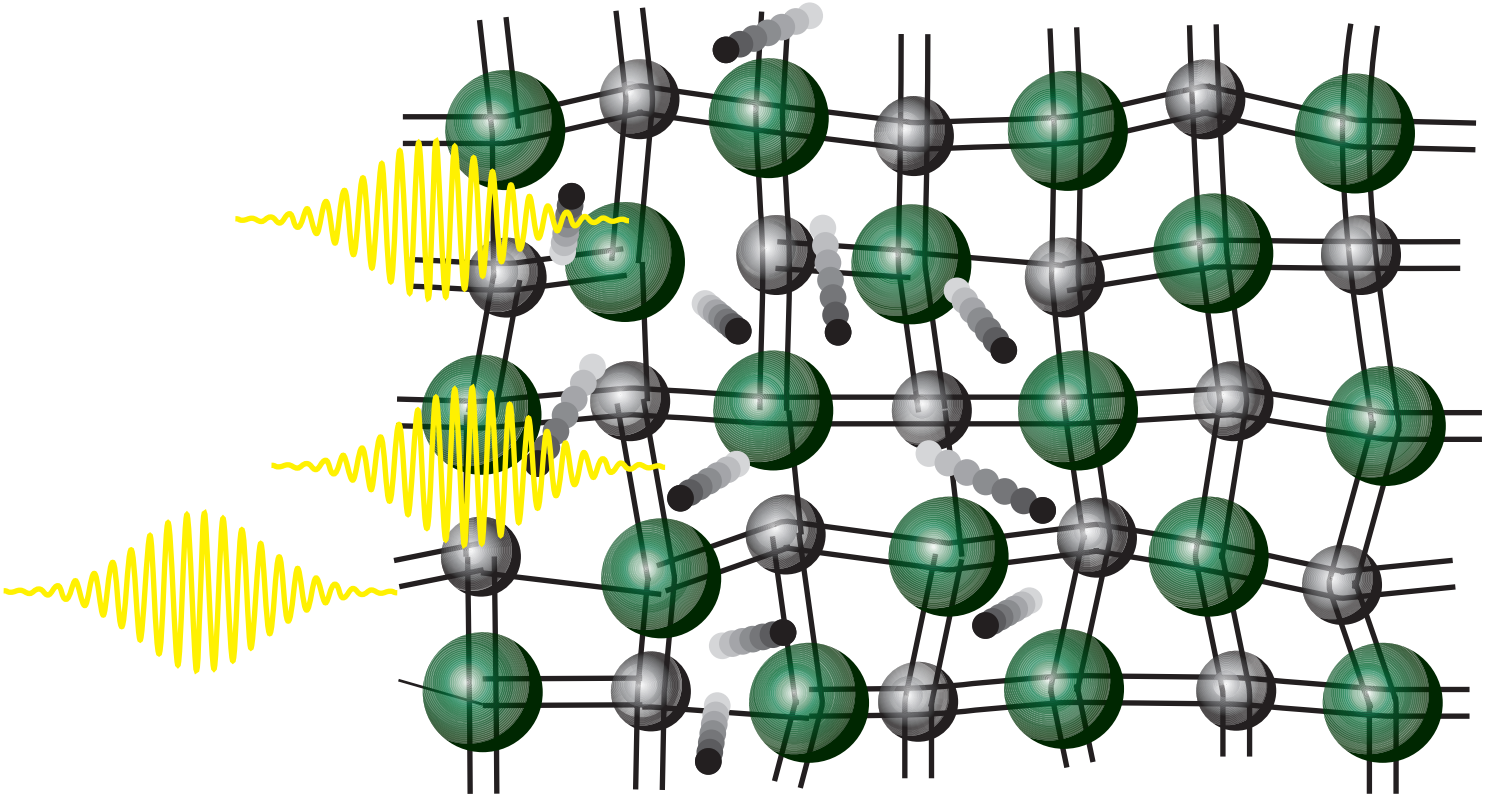
...and create free carriers...



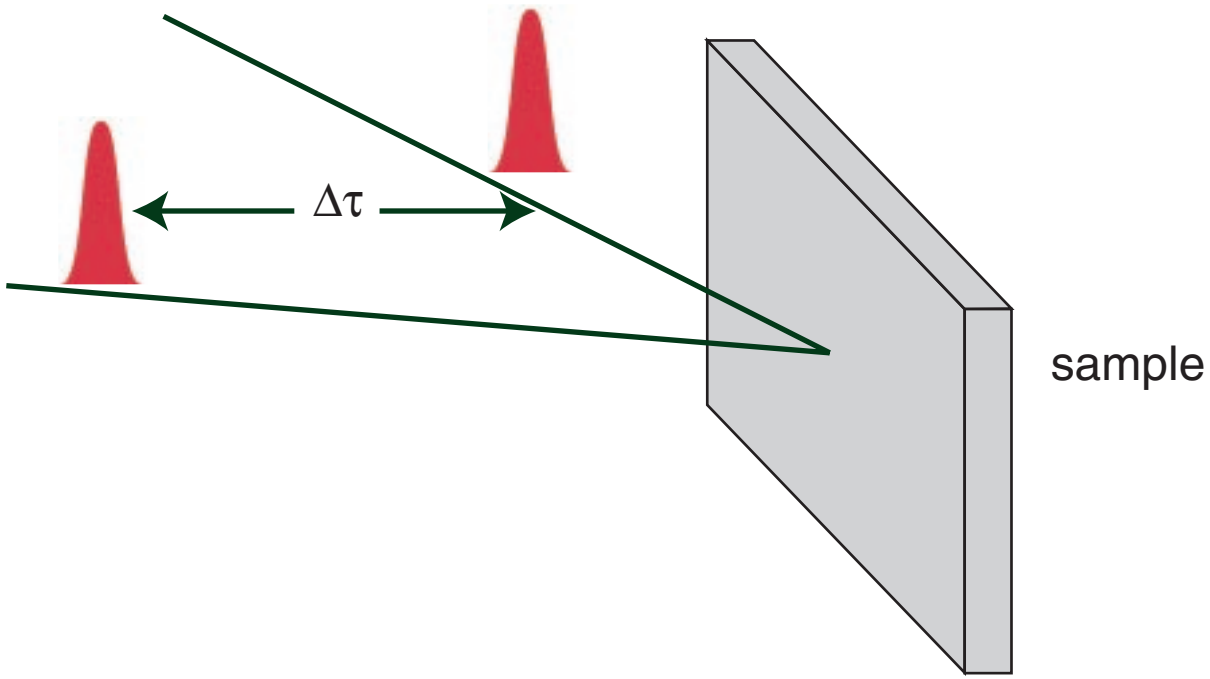
...causing electronic and structural changes...



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

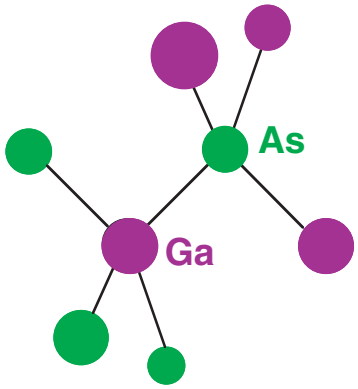


Pump - probe



Introduction

Structure and
Electronic Configuration



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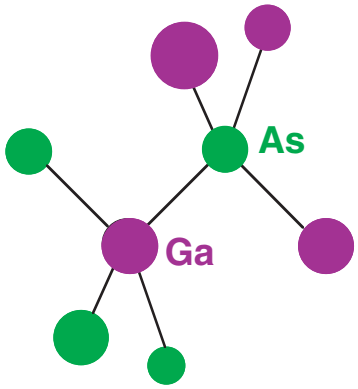
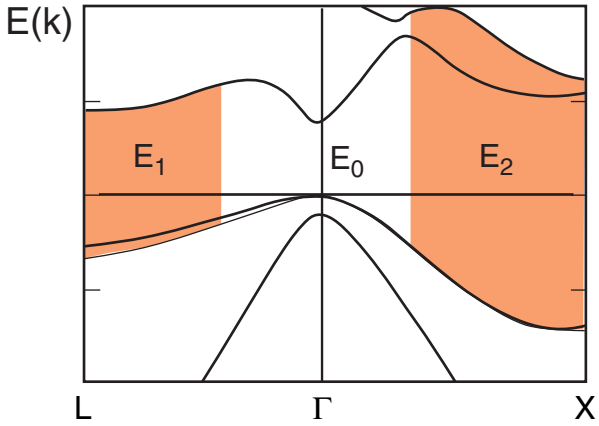


Band Structure
and Occupation



Introduction

Structure and
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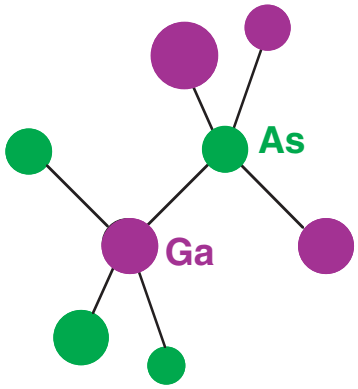

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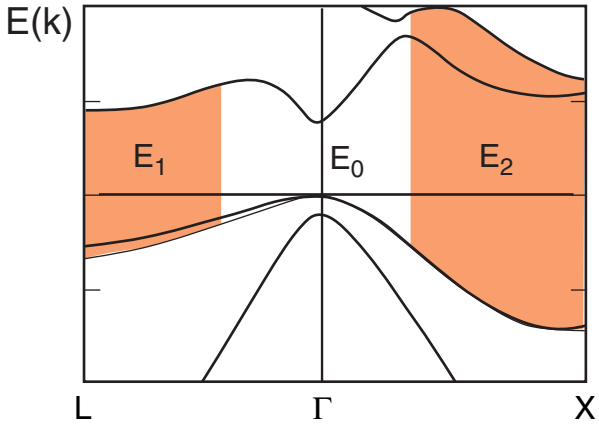
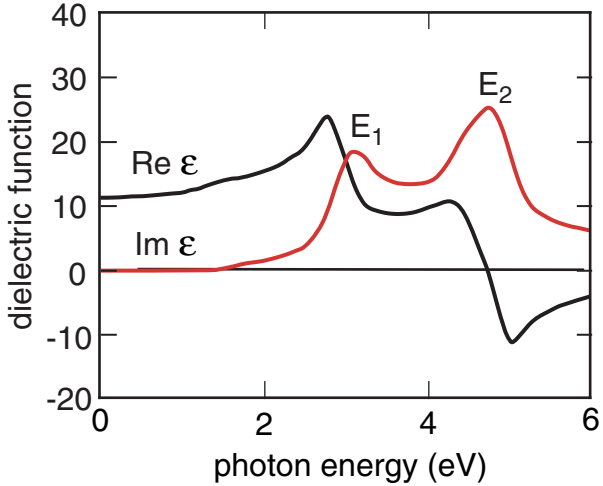


Dielectric Function



Introduction

Structure and
Electronic Configuration

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Band Structure
and Occupation

???

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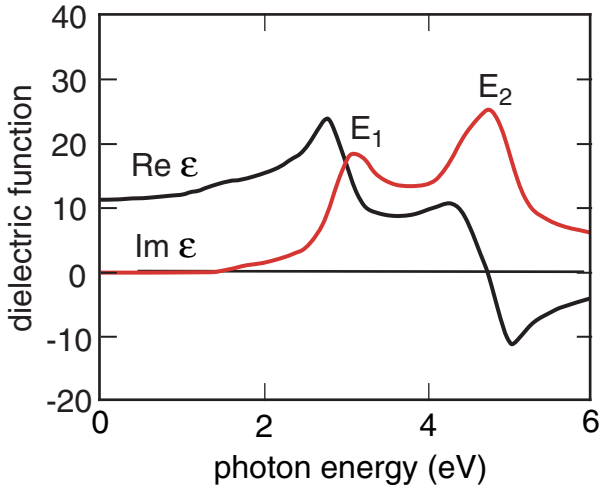
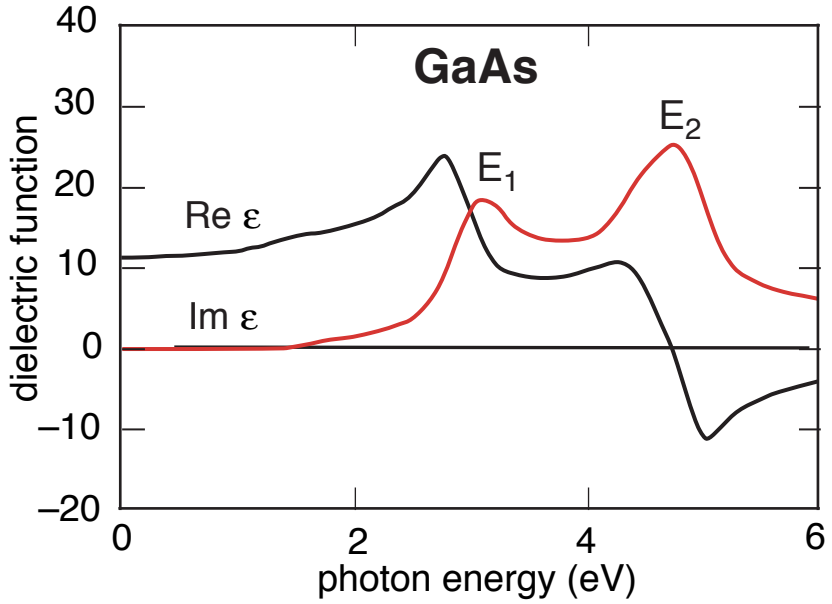
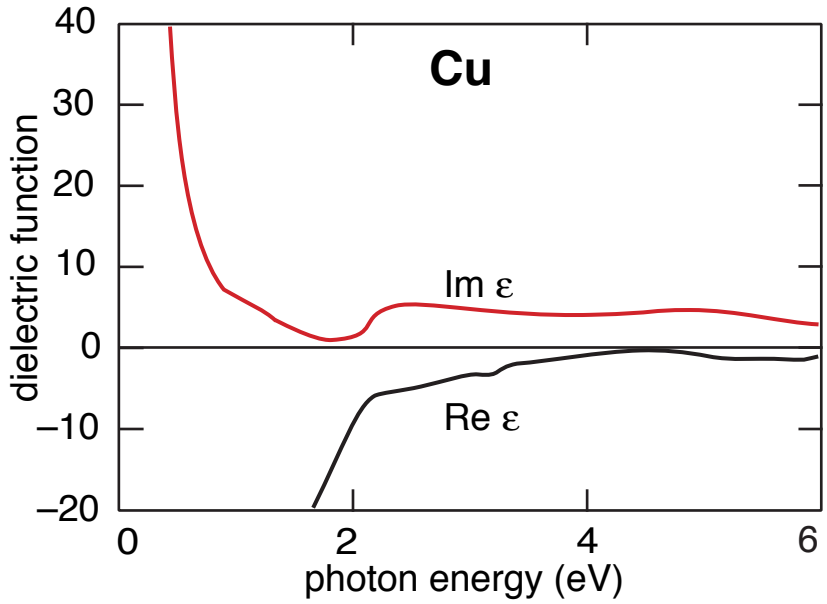
Dielectric Function



Dielectric function varies among different materials





Outline

1. Introduction - Interaction of light with solids

2. **Experimental process and technique**

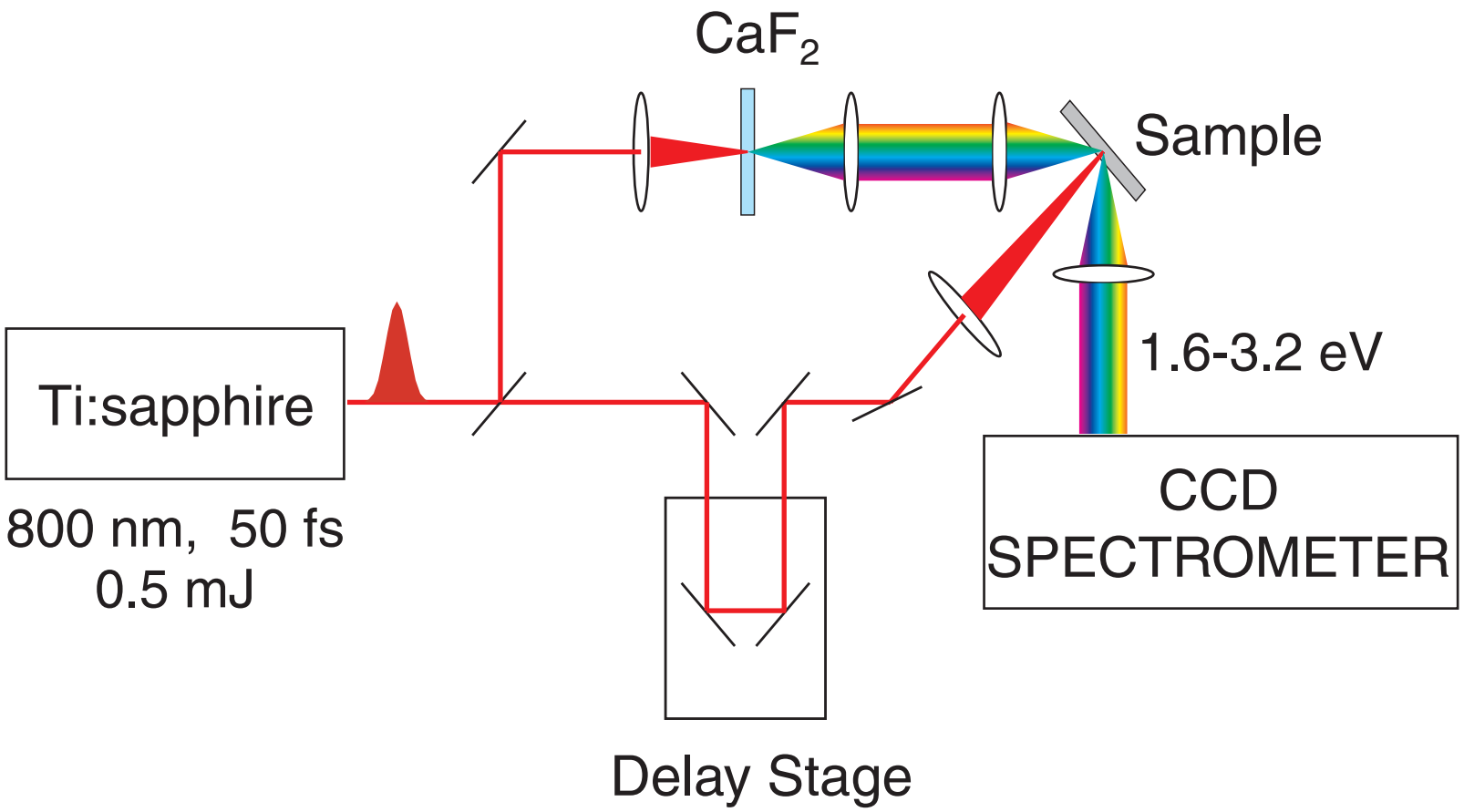
3. Excitation of high amplitude coherent phonons

4. Optical control of coherent phonons

Experimental Technique

Time-resolved reflectometry

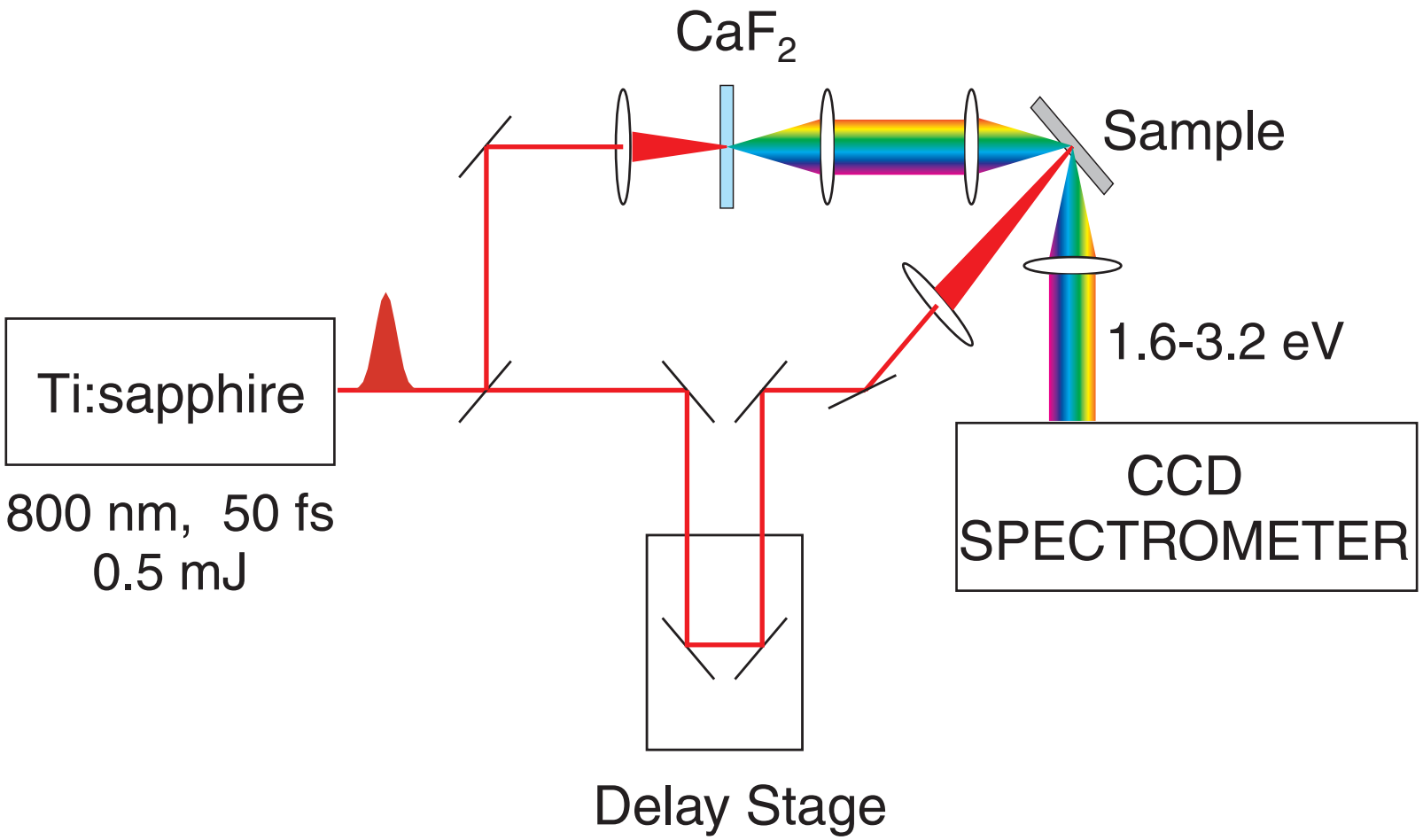
pump-probe setup



Experimental Technique

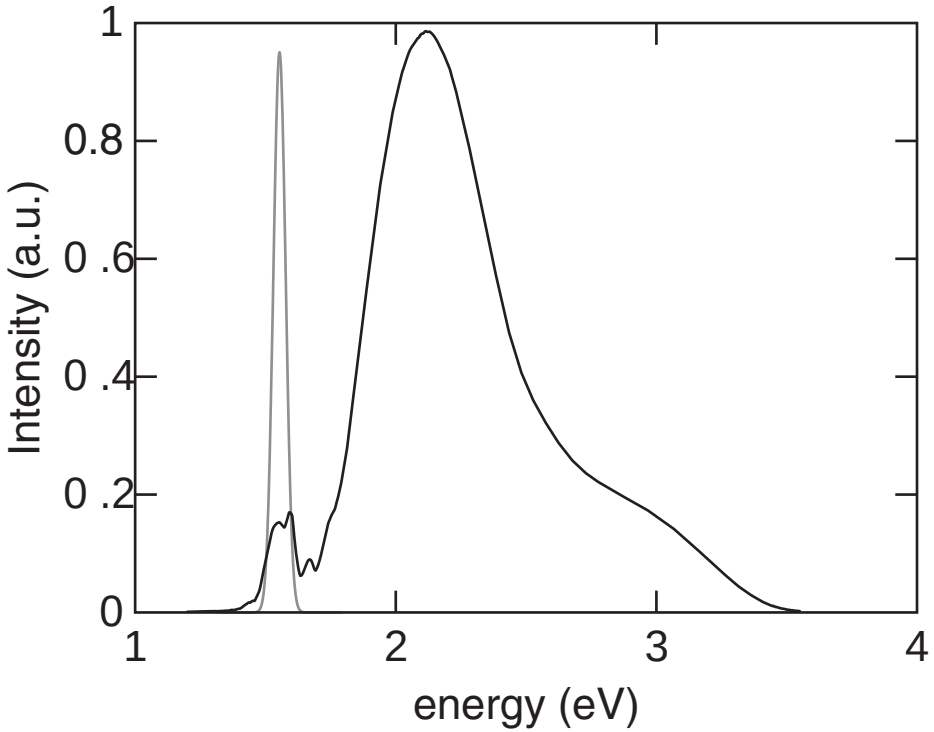
Time-resolved reflectometry

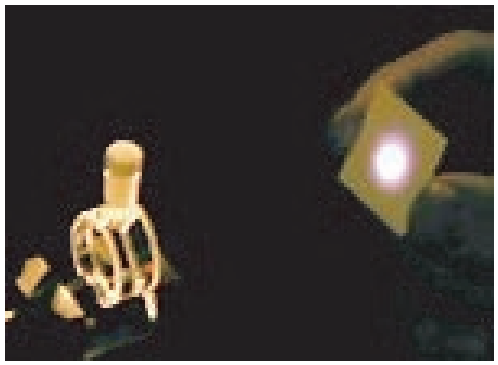
pump-probe setup



Experimental Technique

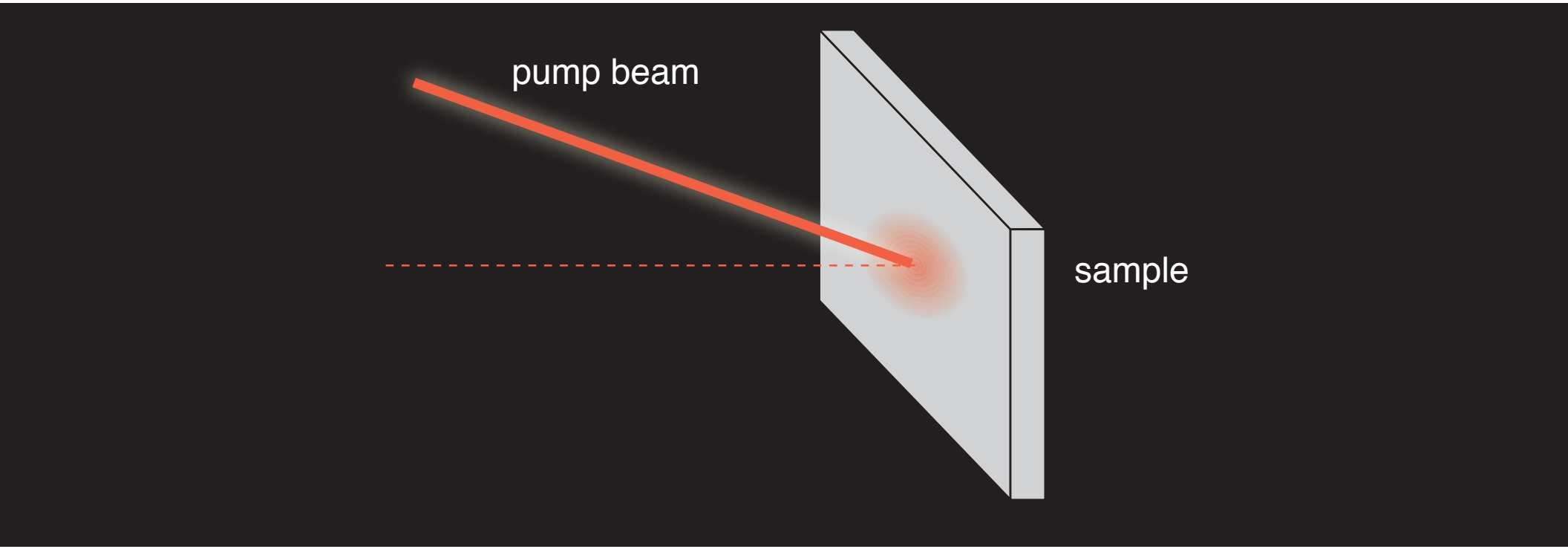
White light spectrum





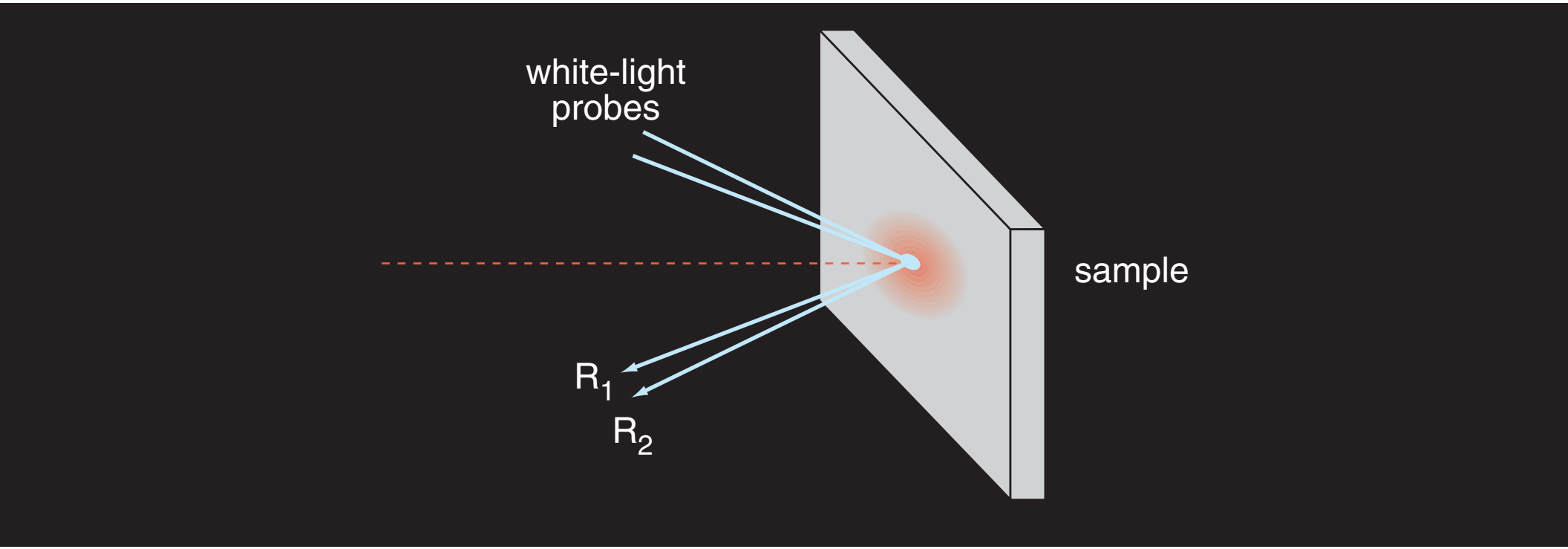
Experimental Technique

Excite sample



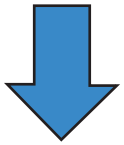
Experimental Technique

Measuring reflectivity at two different angles



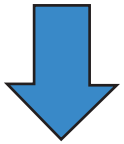
Fresnel Equations

$$\{R_1, R_2, \theta\}$$



$$r_p = \frac{\varepsilon \cos\theta - \sqrt{\varepsilon_0}\,\sqrt{\varepsilon - \varepsilon_0 \sin^2\theta}}{\varepsilon \cos\theta + \sqrt{\varepsilon_0}\,\sqrt{\varepsilon - \varepsilon_0 \sin^2\theta}}$$

$$r_s = \frac{\sqrt{\varepsilon_0}\cos\theta - \sqrt{\varepsilon - \varepsilon_0 \sin^2\theta}}{\sqrt{\varepsilon_0}\cos\theta + \sqrt{\varepsilon - \varepsilon_0 \sin^2\theta}}$$



$$\{\mathrm{Re}[\varepsilon(\omega)], \mathrm{Im}[\varepsilon(\omega)]\}$$

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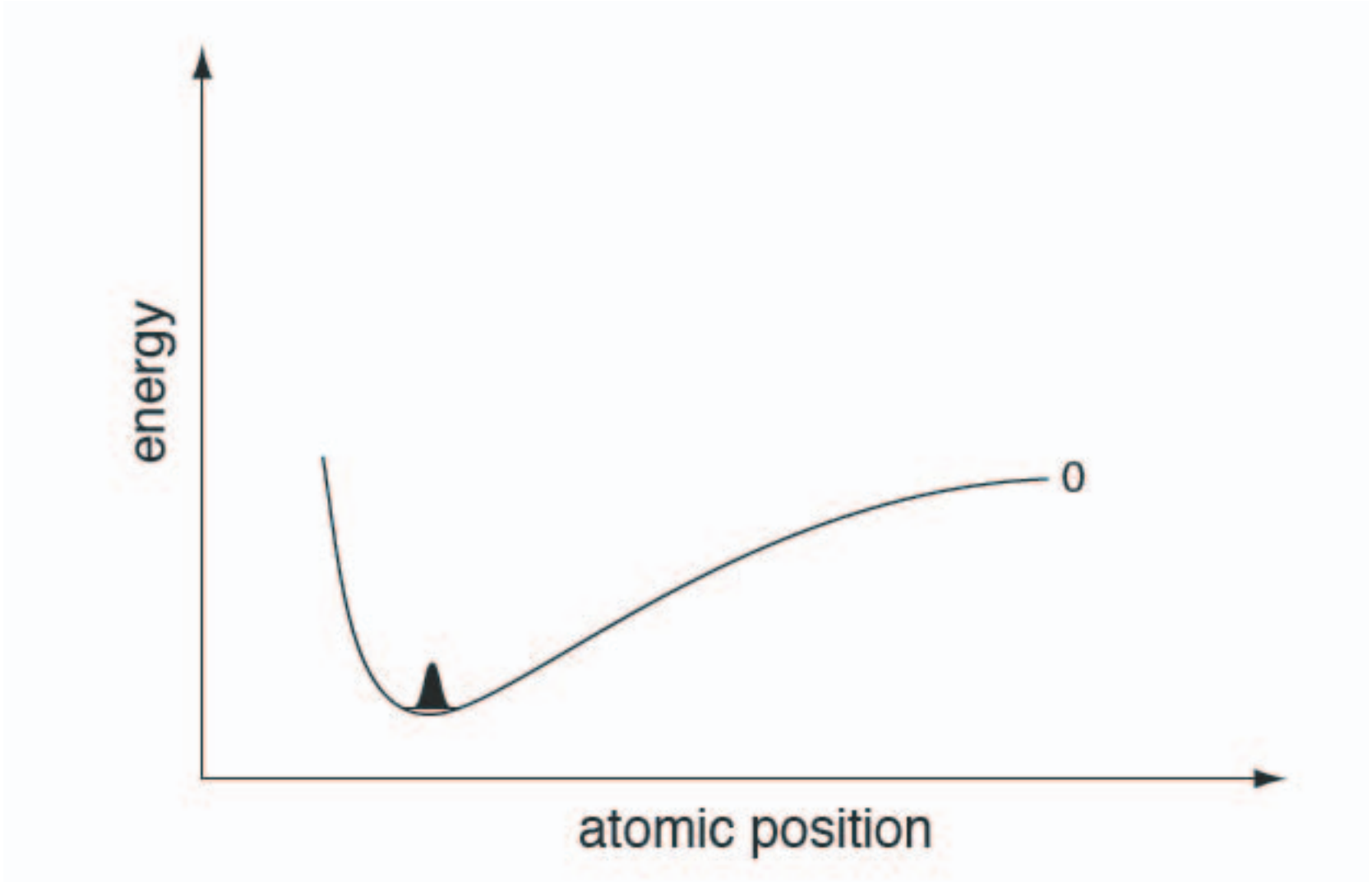
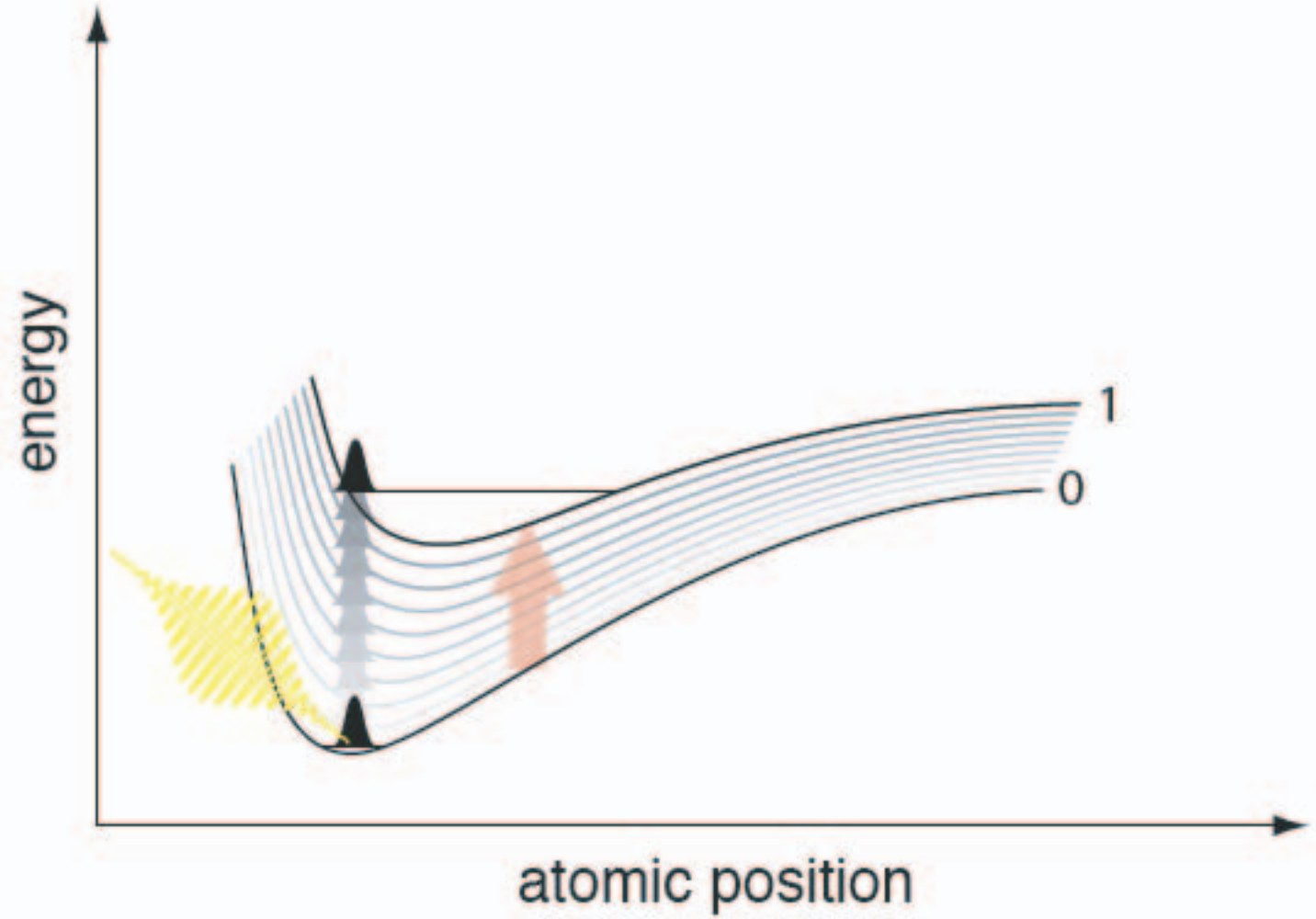
Coherent Phonons

Before irradiation nuclear wavepacket sits at potential minimum



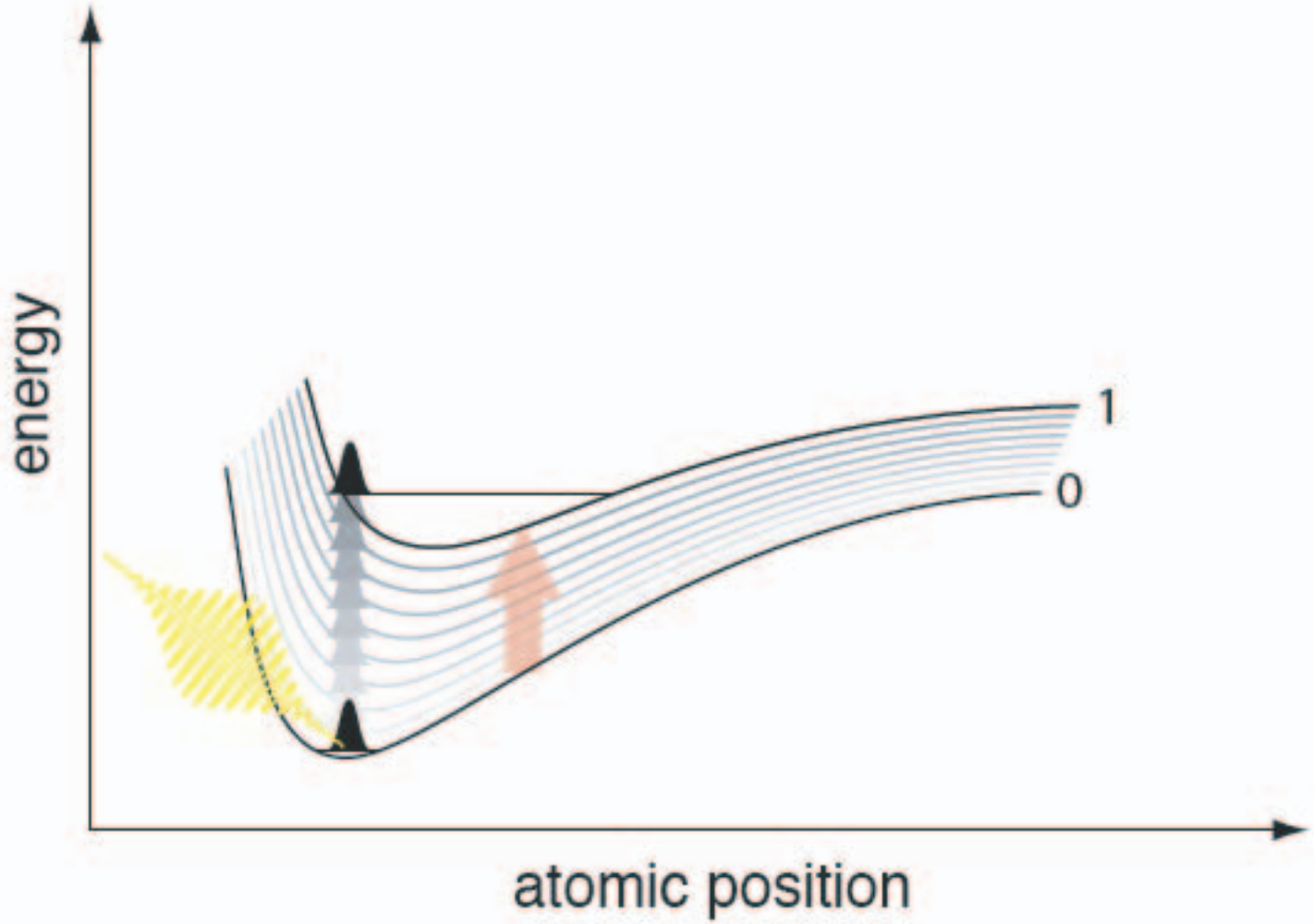
Coherent Phonons

Femtosecond pumping: electrons are almost instantaneously excited altering the potential landscape



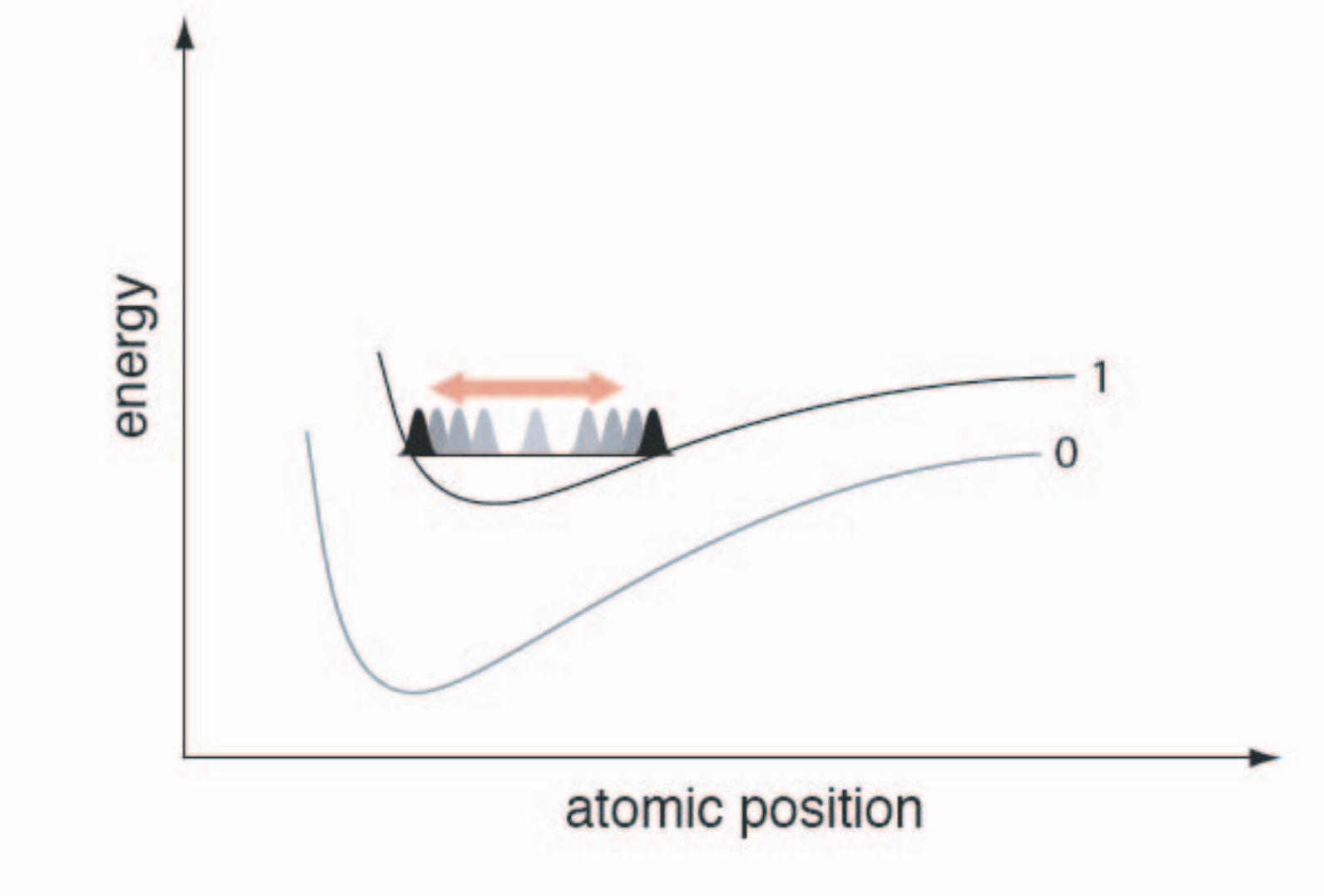
Coherent Phonons

Nuclear wavepacket is projected on new potential surface...



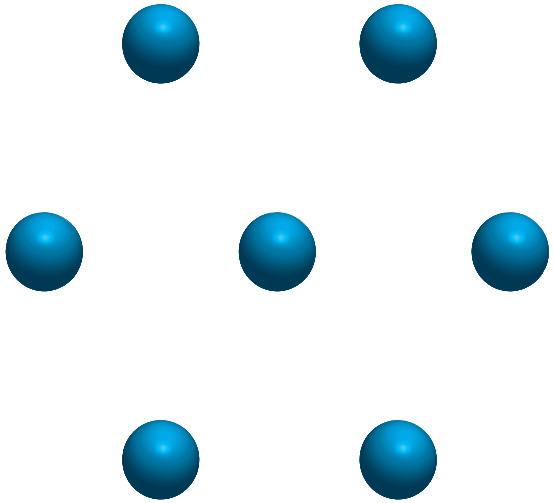
Coherent Phonons

...and starts oscillating around new equilibrium position



Coherent Phonons in Te

Te structure: 3-atom per turn helices arranged in a hexagonal array



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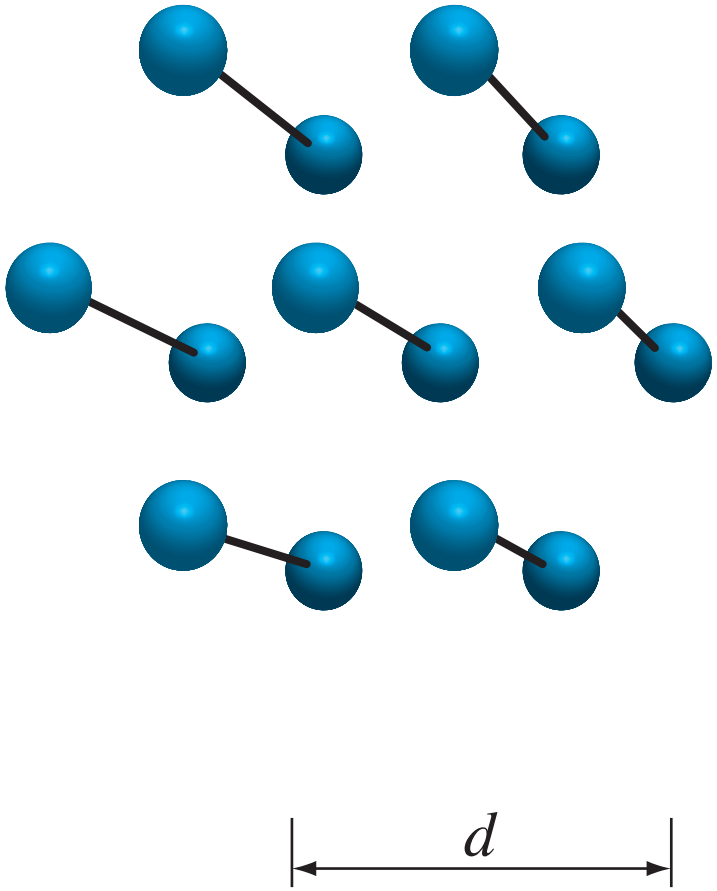

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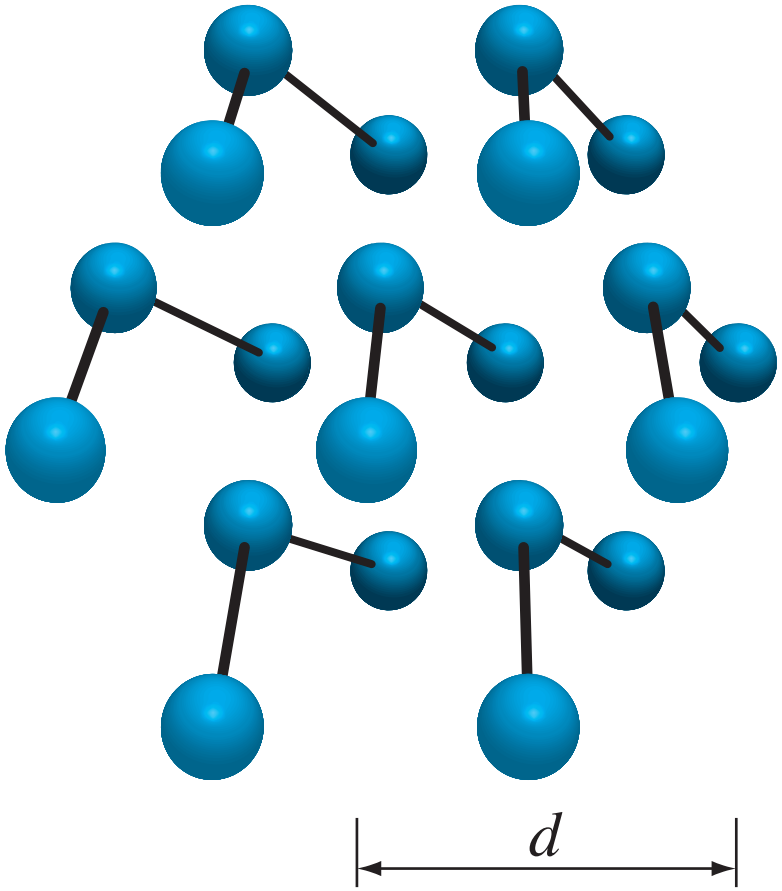

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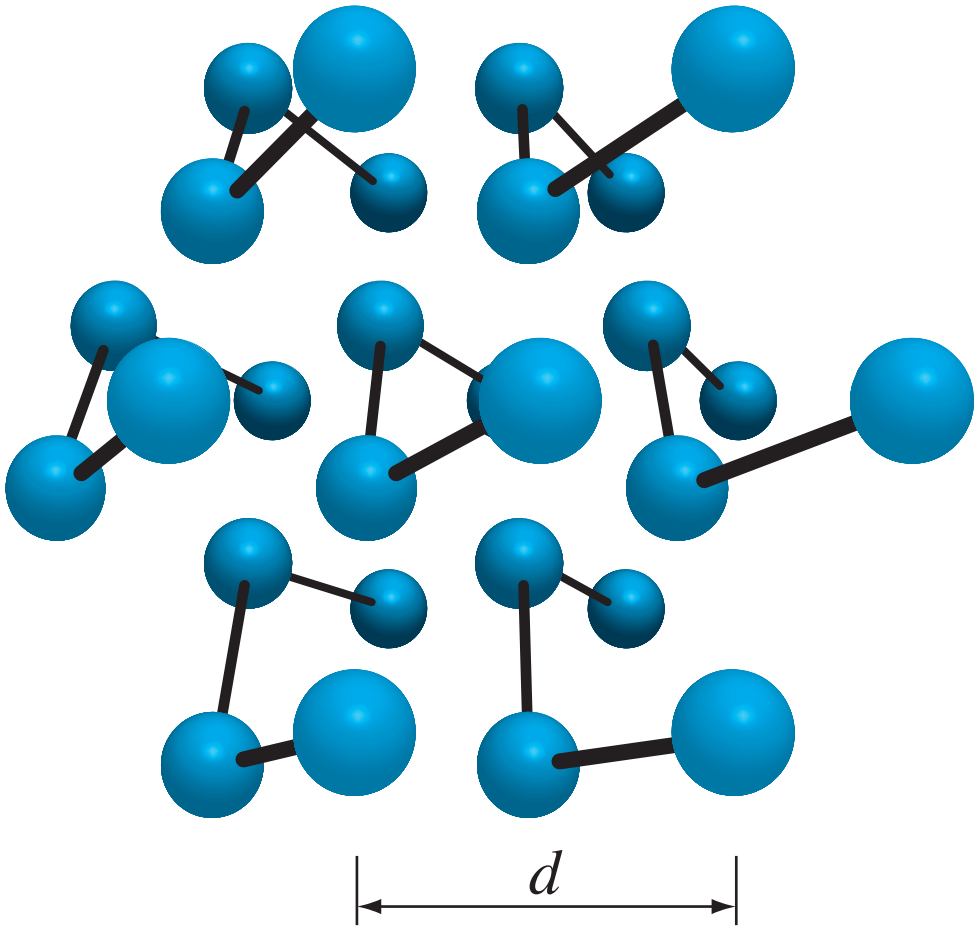
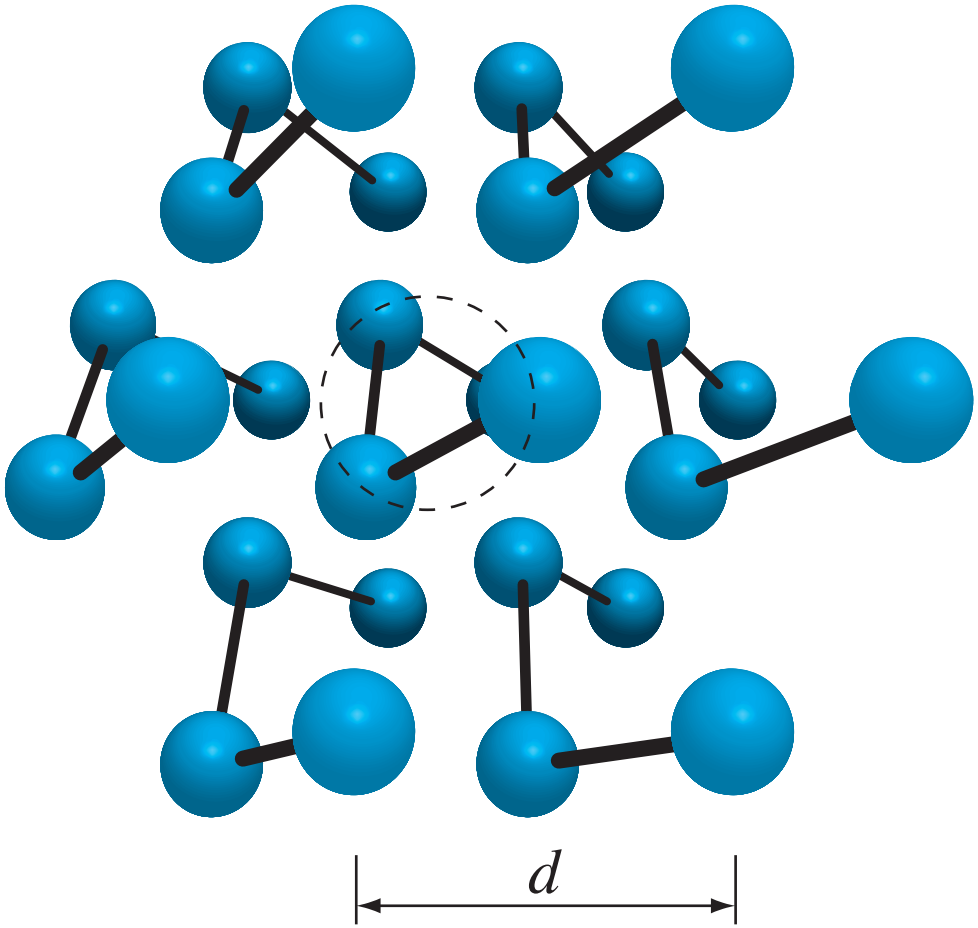


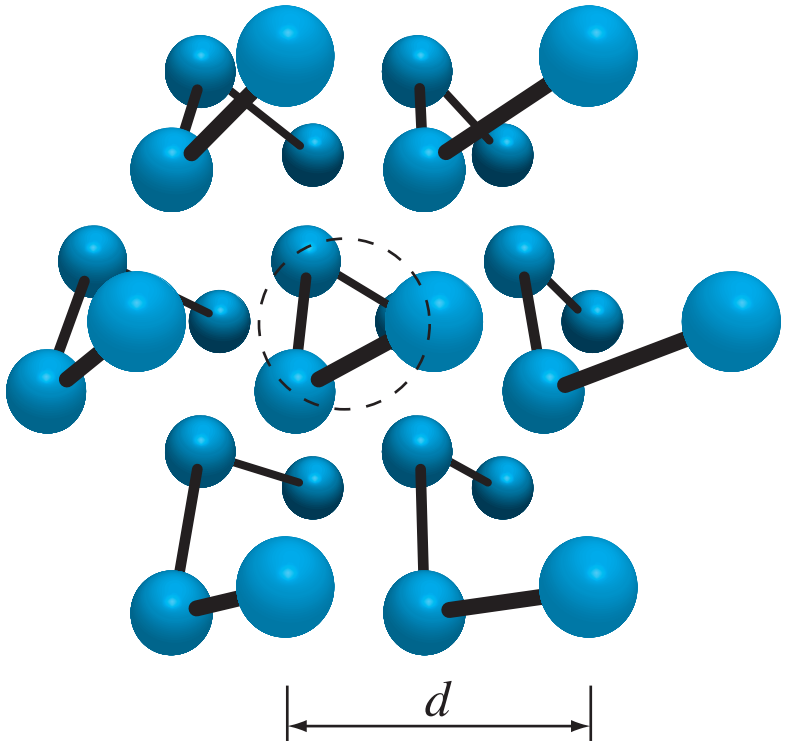
Coherent Phonons in Te

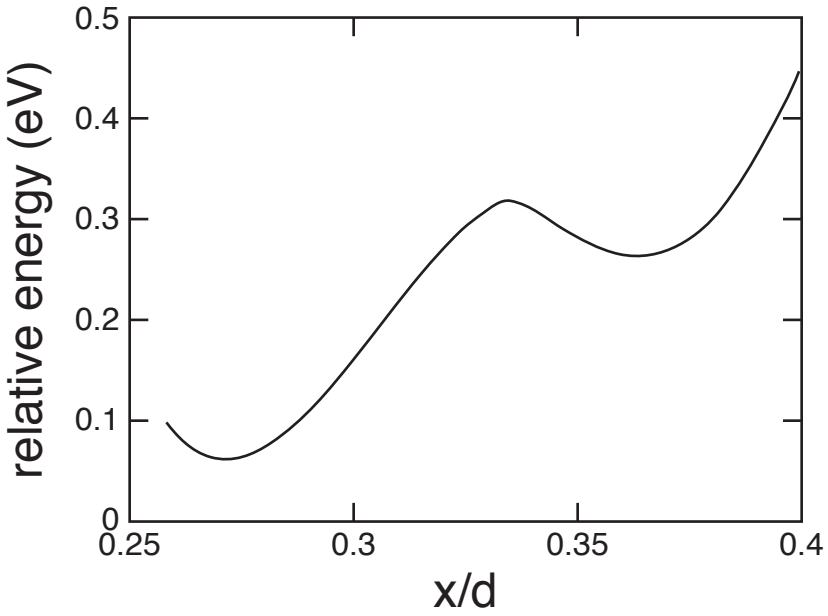
helical radius x = 0.27d



Coherent Phonons in Te

Equilibrium helical radius $x_0 = 0.27d$

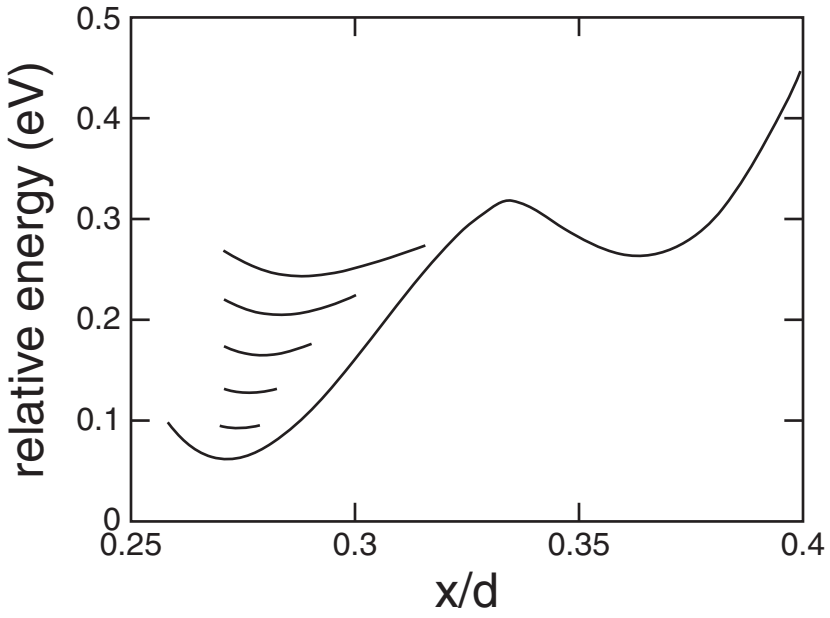


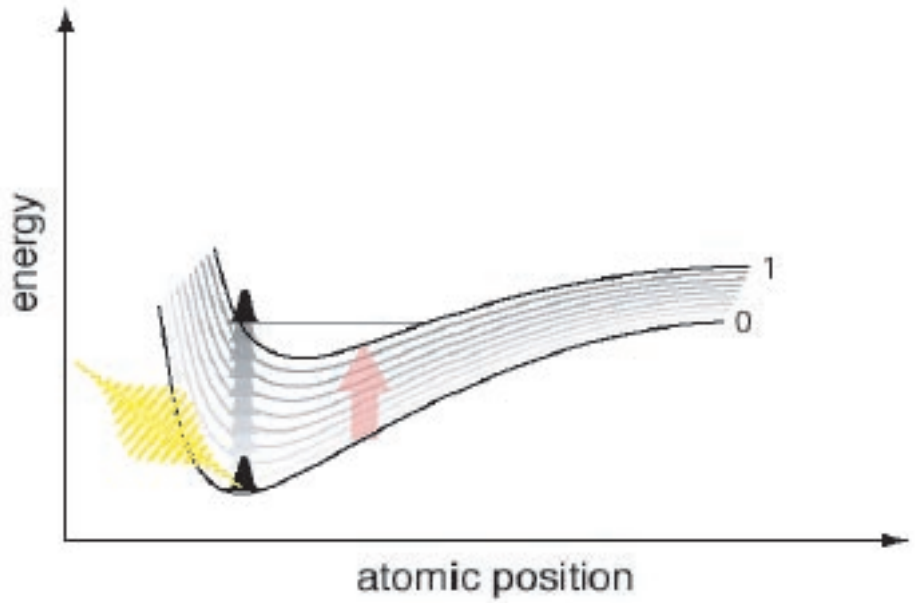


Tangney and Fahy, *Phys. Rev. B* **65**, 054302 (2002)

Coherent Phonons in Te

Equilibrium position shifts upon photoexcitation

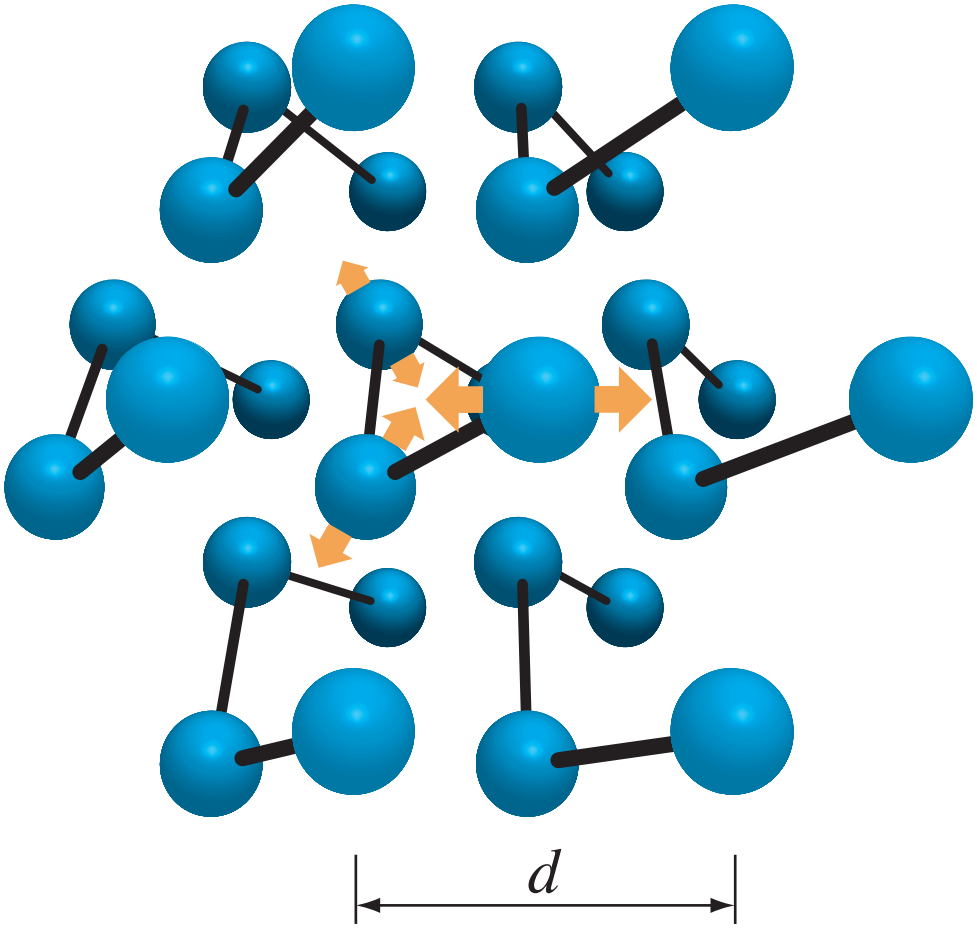




Tangney and Fahy, *Phys. Rev. B* **65**, 054302 (2002)

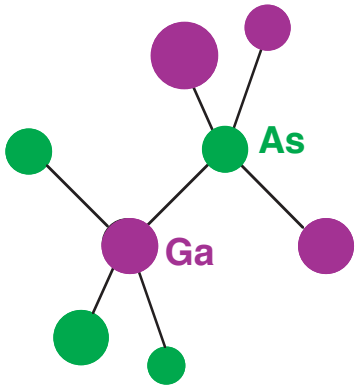
Coherent Phonons in Te

A_1 phonon mode is excited

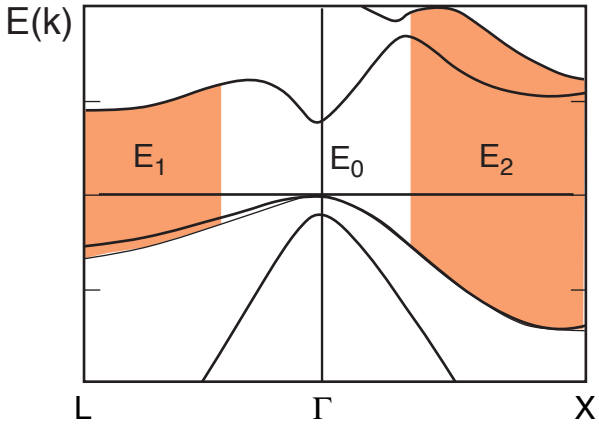


Coherent Phonons in Te

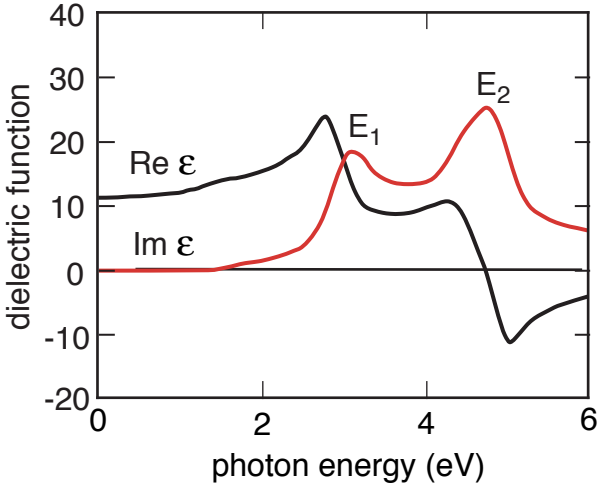
Structure and
Electronic Configuration



Band Structure
and Occupation

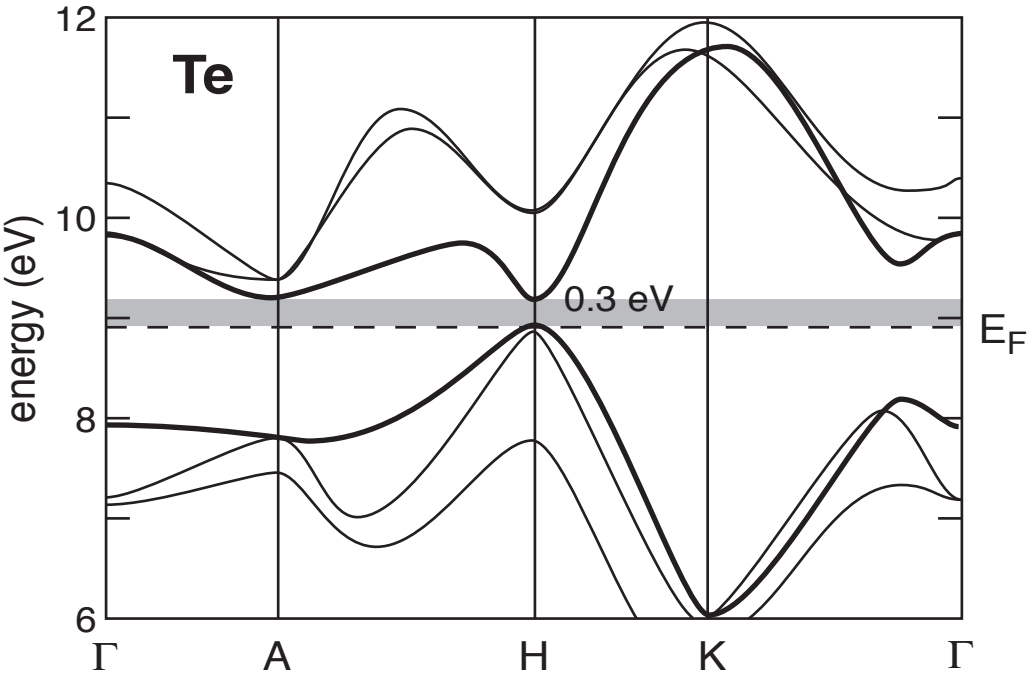


Dielectric Function



Coherent Phonons in Te

Te bandstructure



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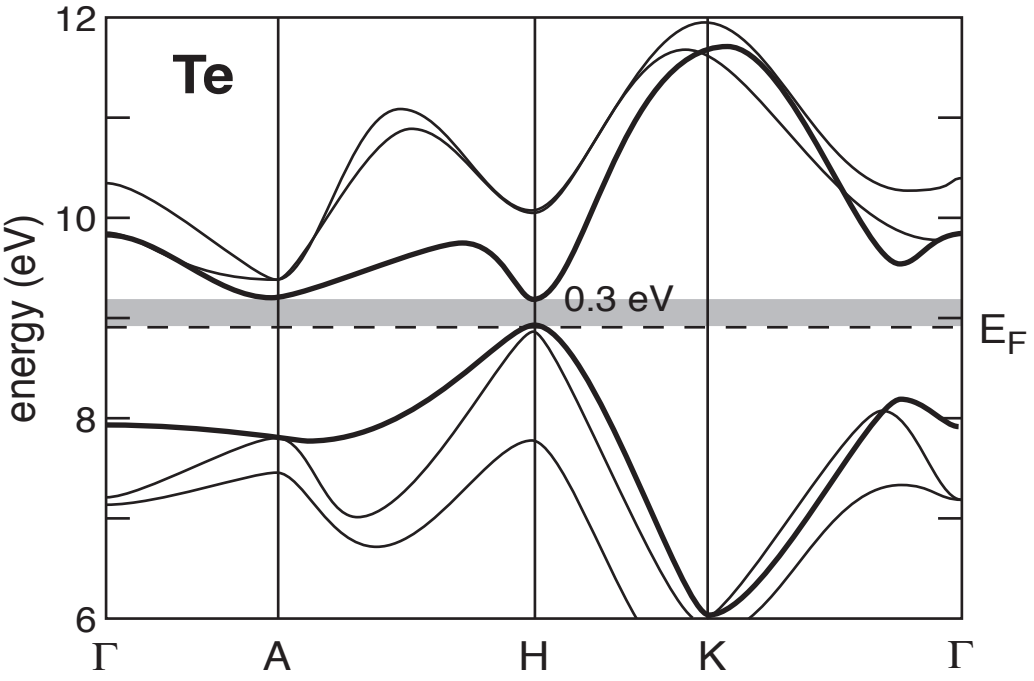
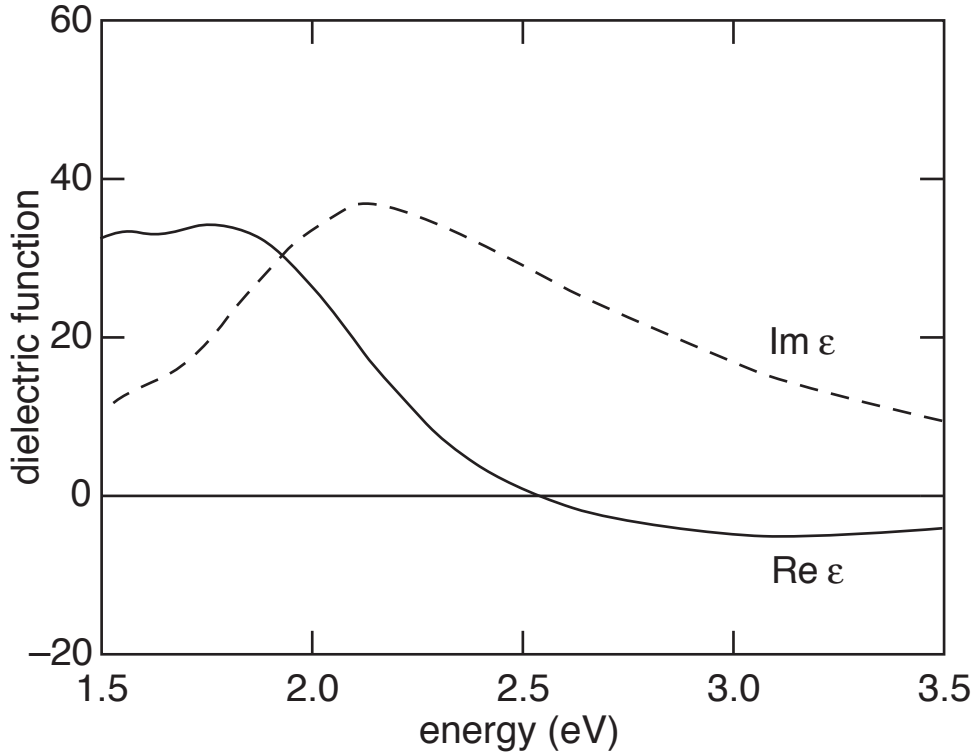
Coherent Phonons in Te

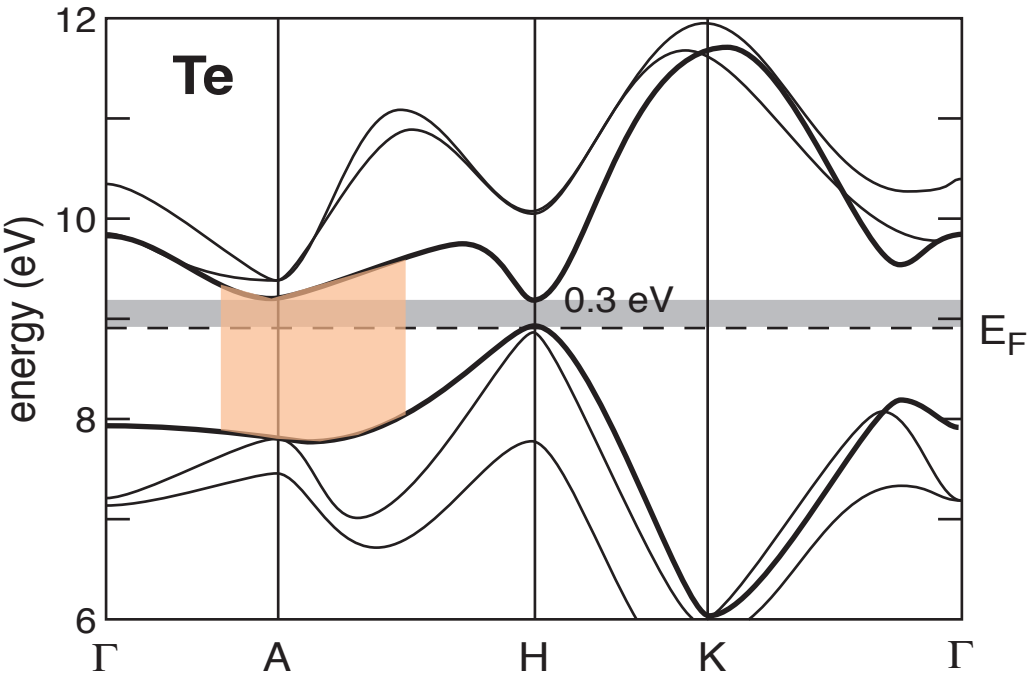
Te bandstructure and dielectric function

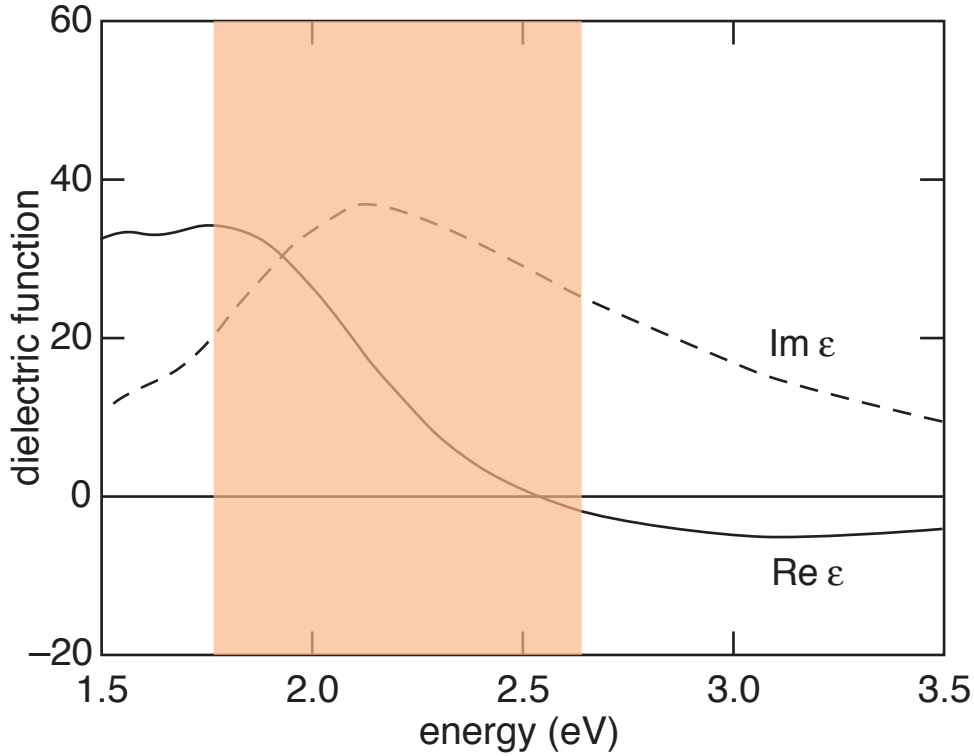




Coherent Phonons in Te

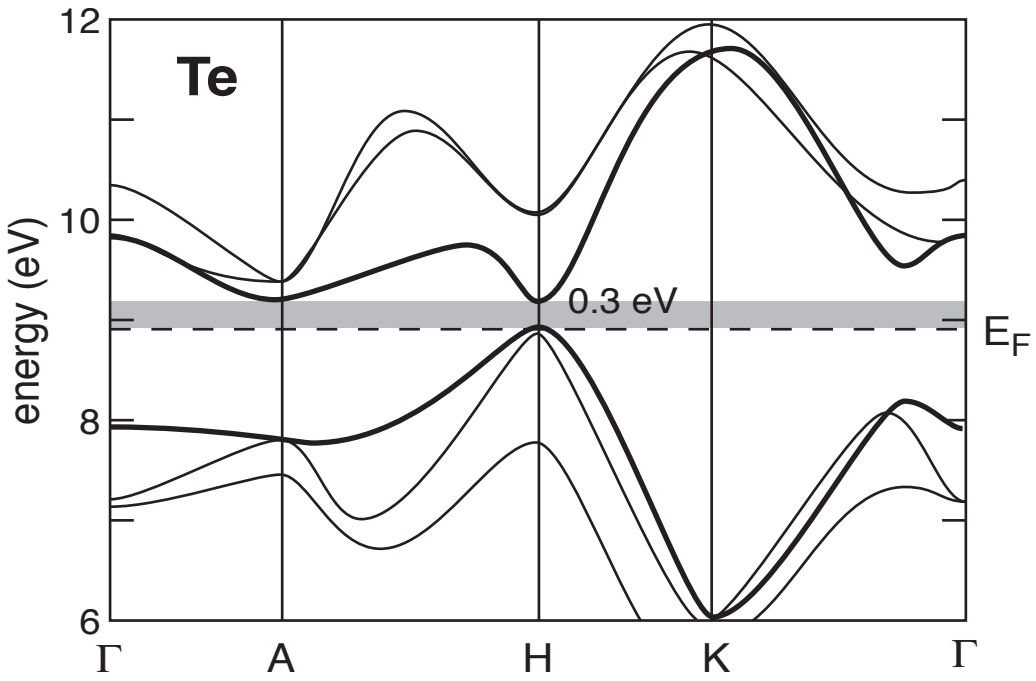
Resonance of dielectric function mainly due to transitions around the A point

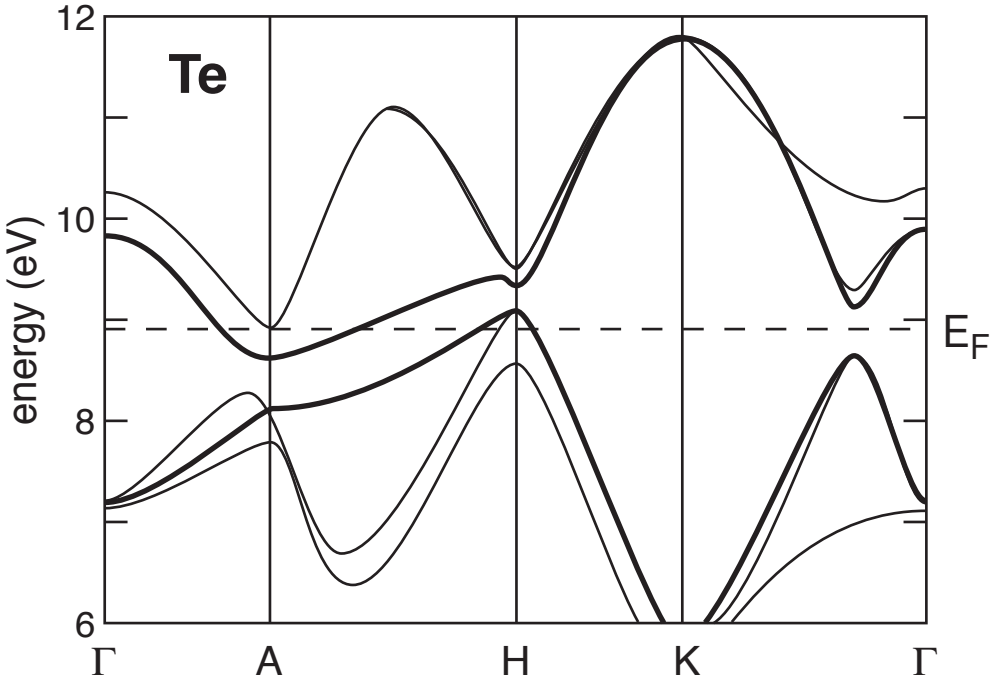




Coherent Phonons in Te

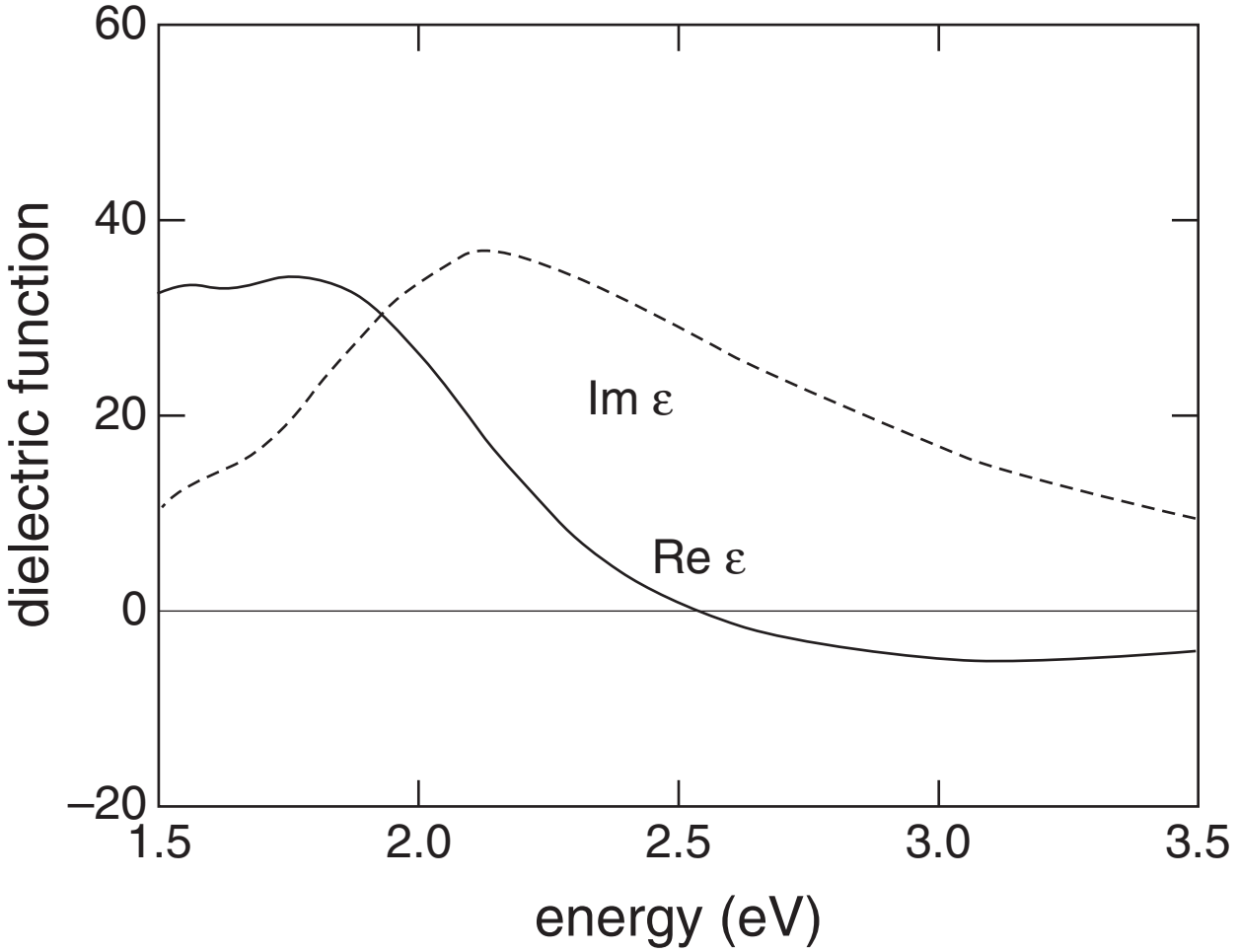
Lattice displacement redshifts resonance





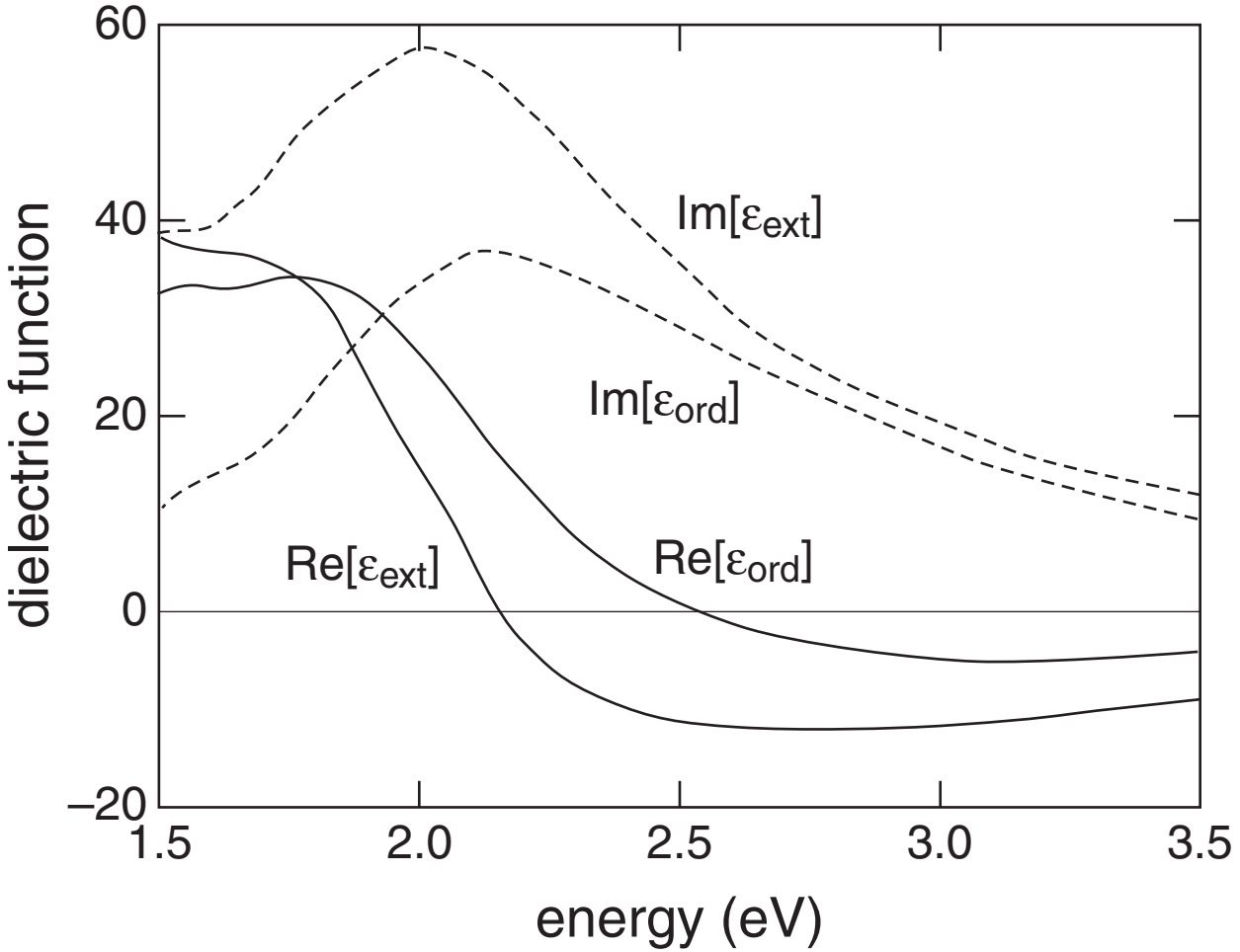
Tangney and Fahy, *Phys. Rev. B* **65**, 054302 (2002)

Literature values for dielectric function



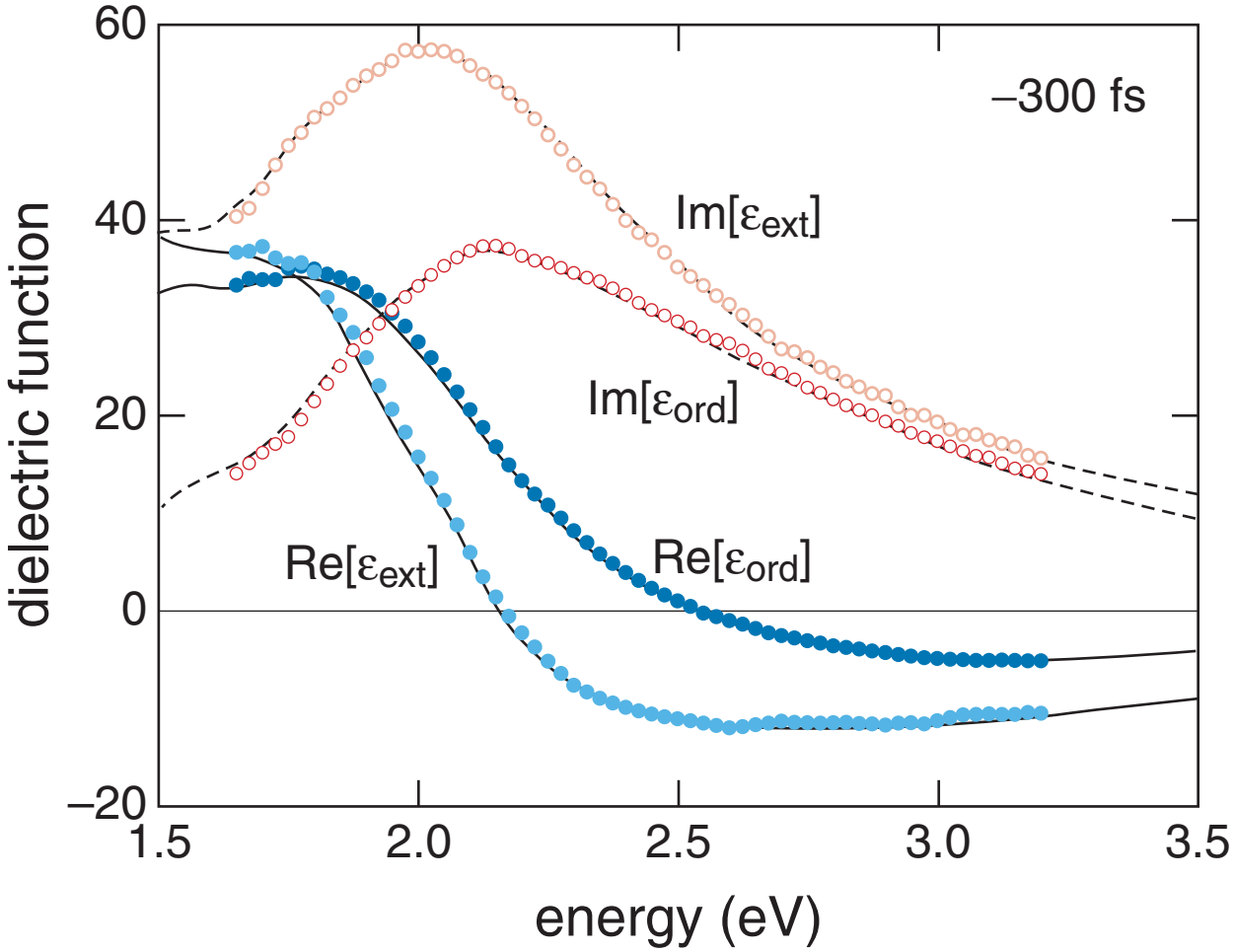
Coherent Phonons in Te

Te is uniaxial, actually



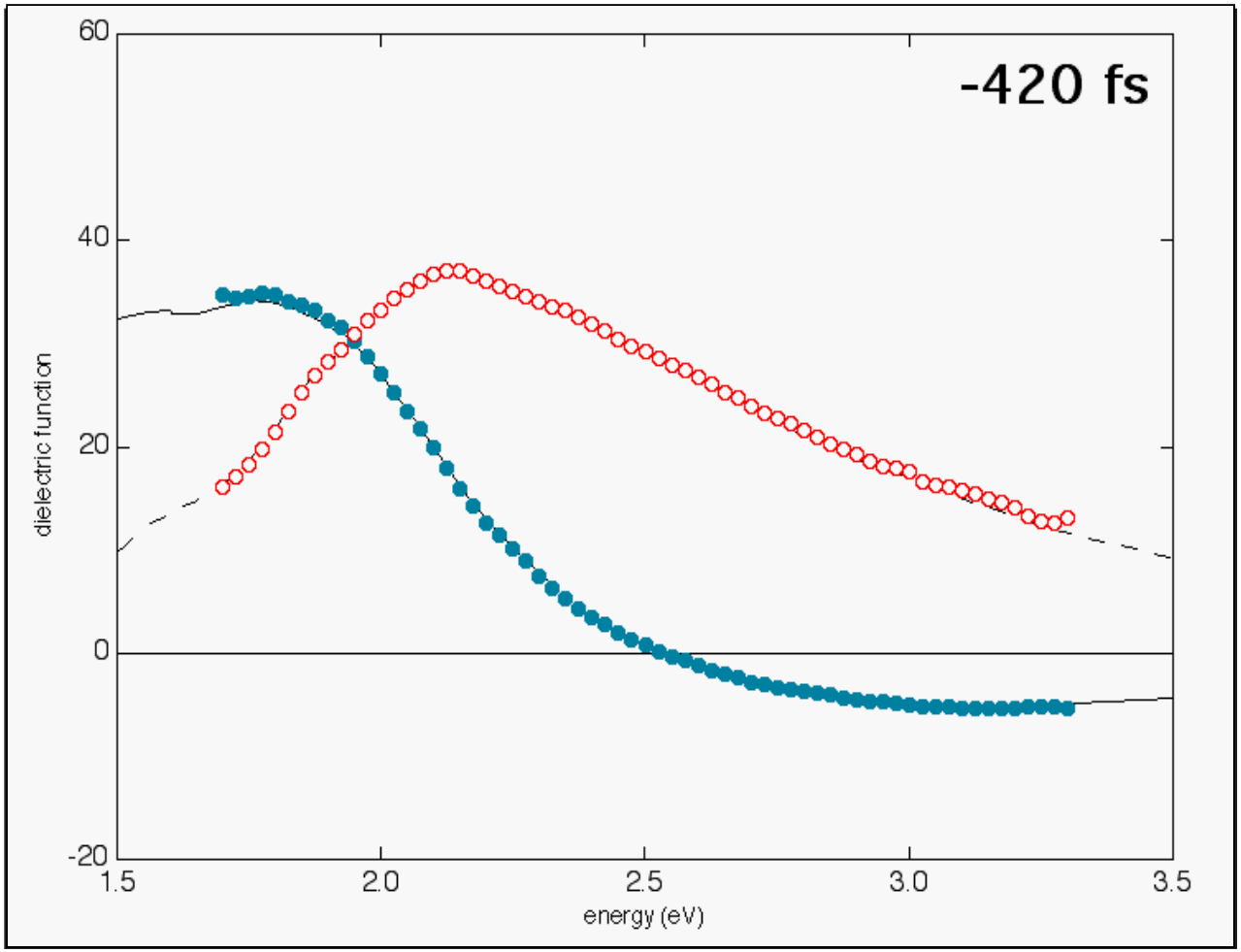
Coherent Phonons in Te

Data agree very well with literature

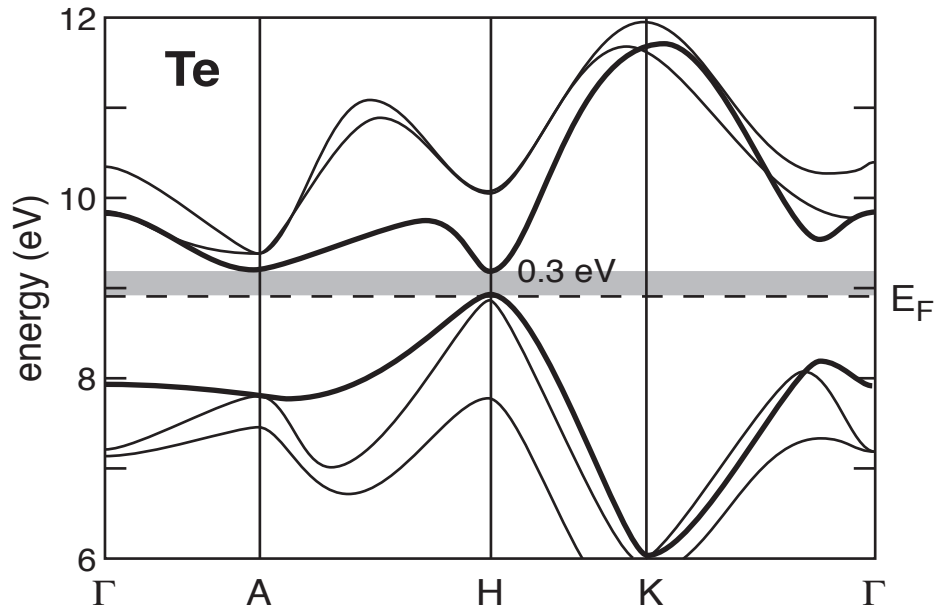


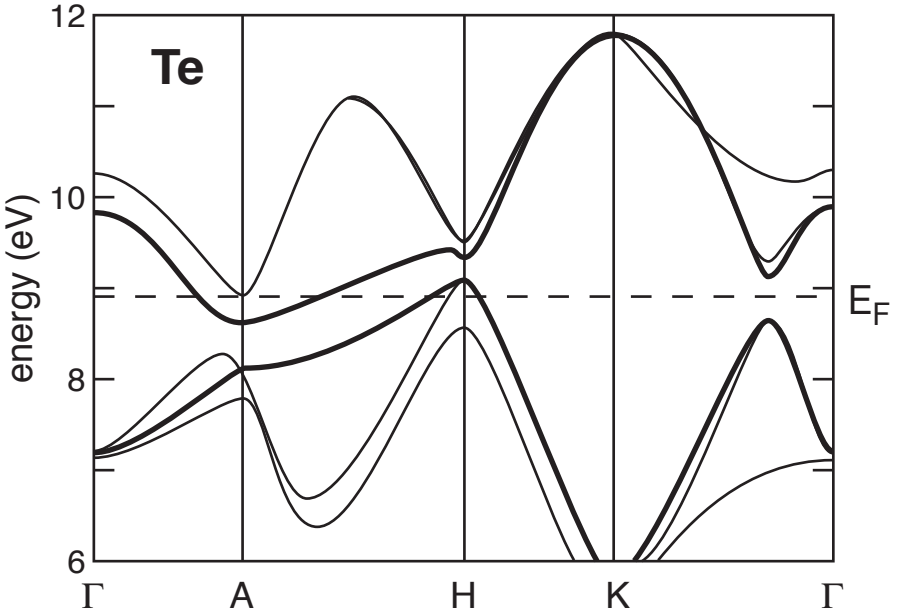
Coherent Phonons in Te

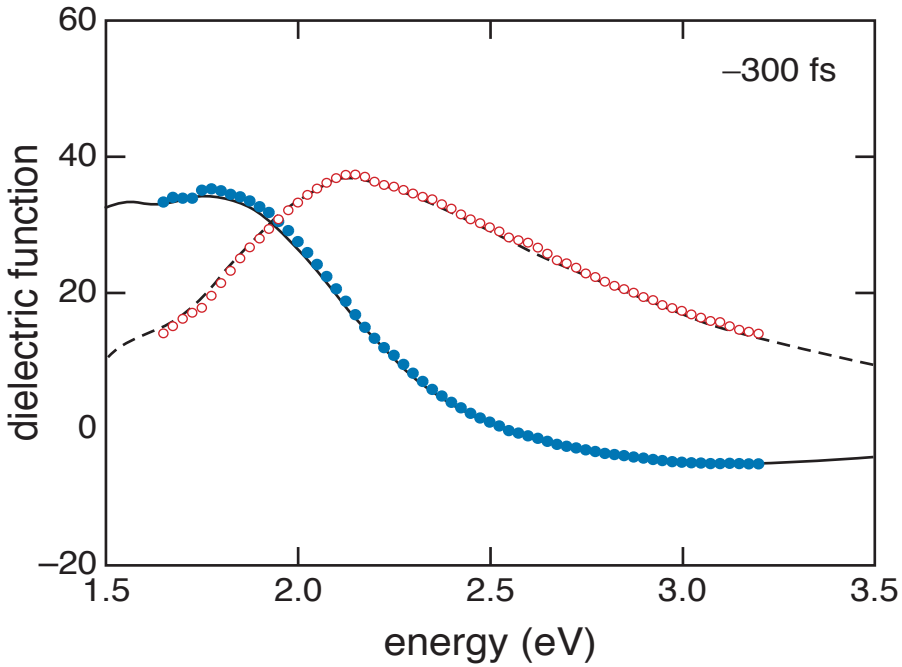
Te under 0.71 F_{th} excitation

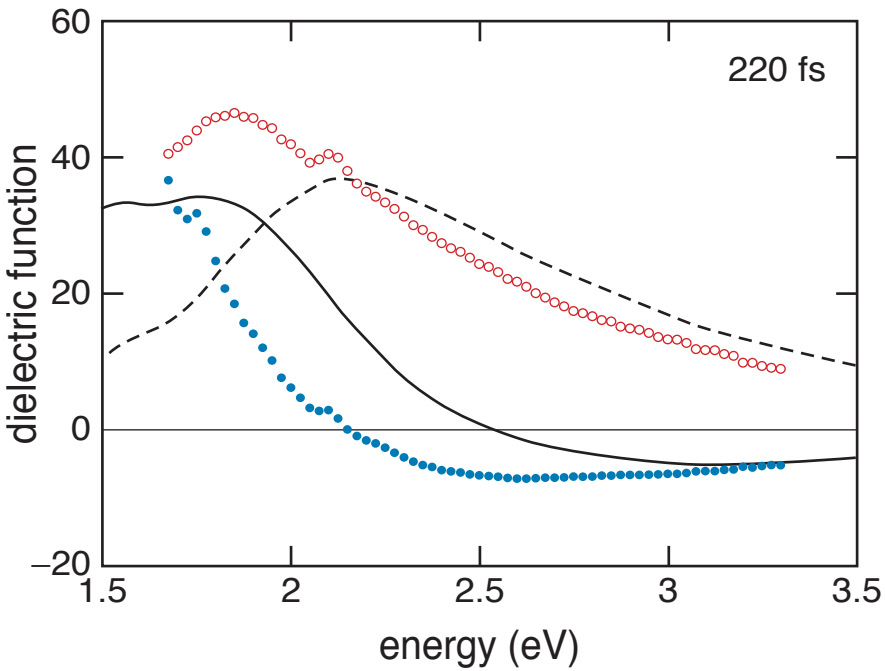


Coherent Phonons in Te

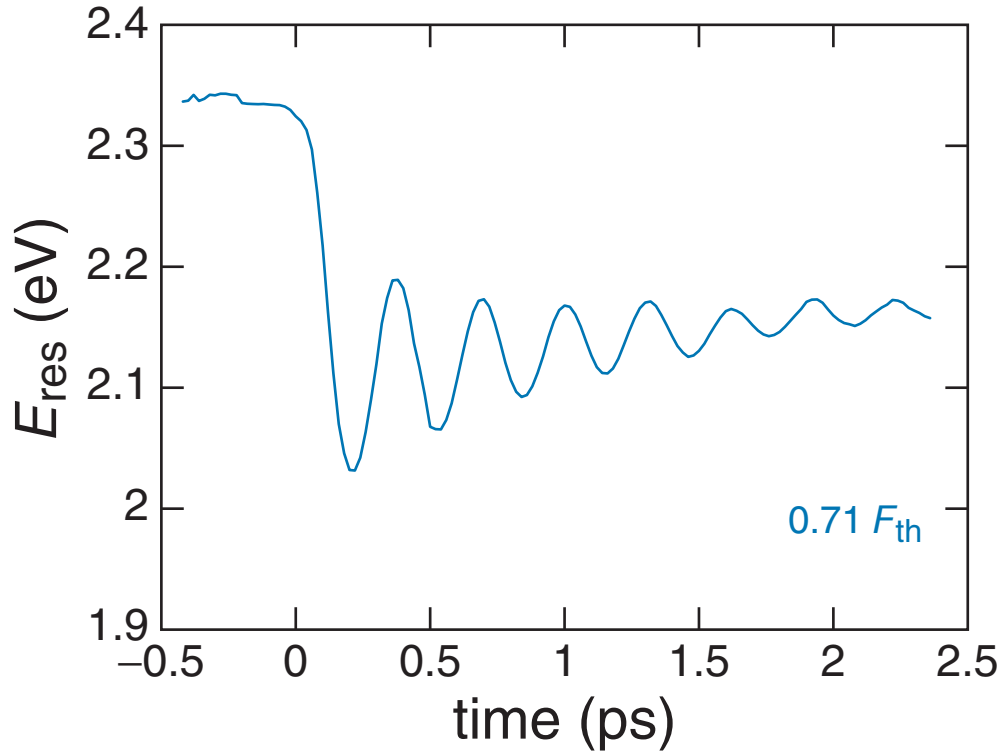






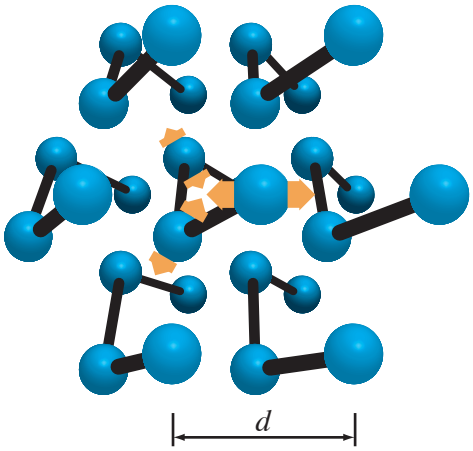


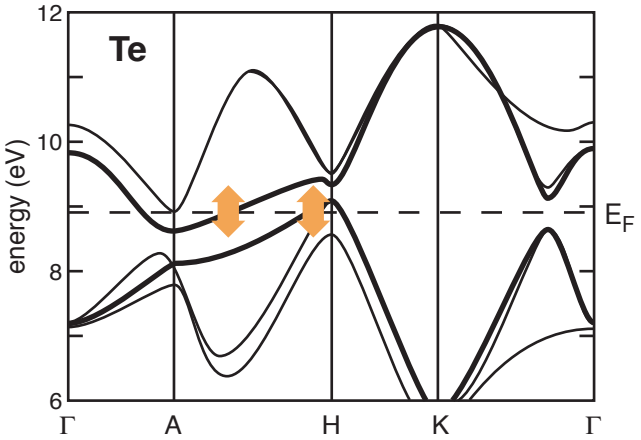
Resonance as a function of time

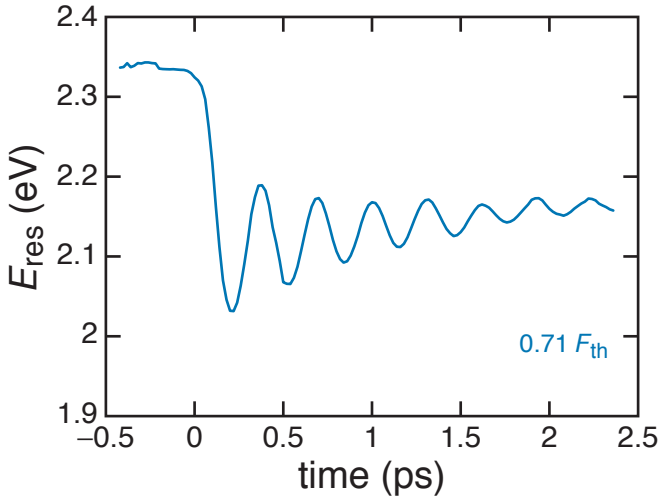


Coherent Phonons in Te

Phonon excitation modifies x, modifies bandstructure, modifies ε



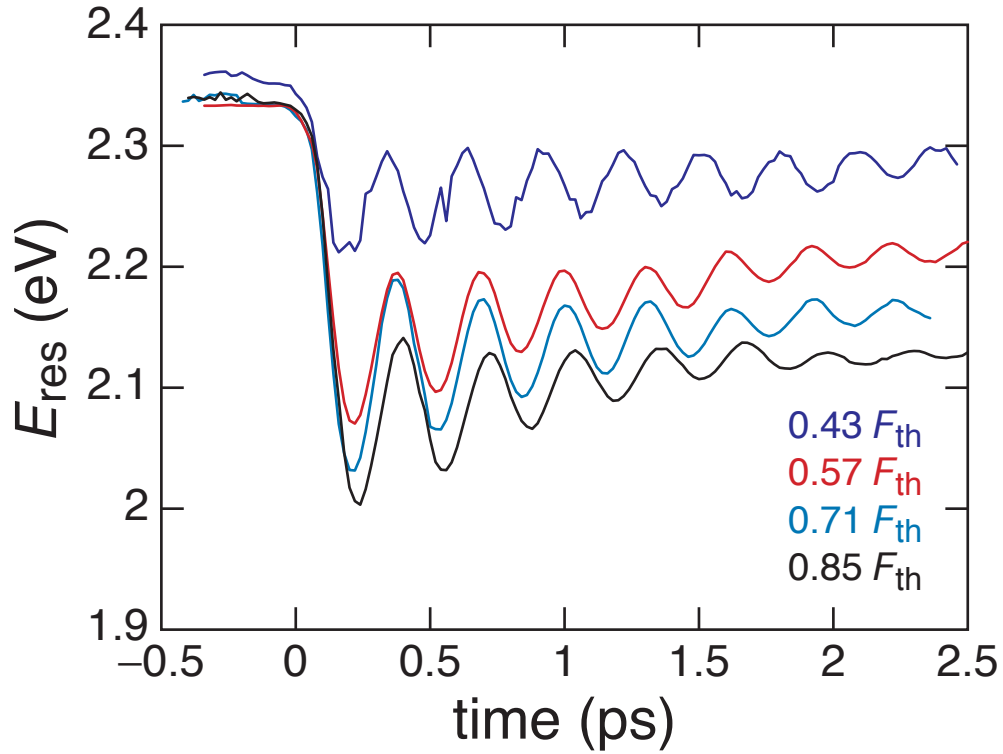




Coherent Phonons in Te

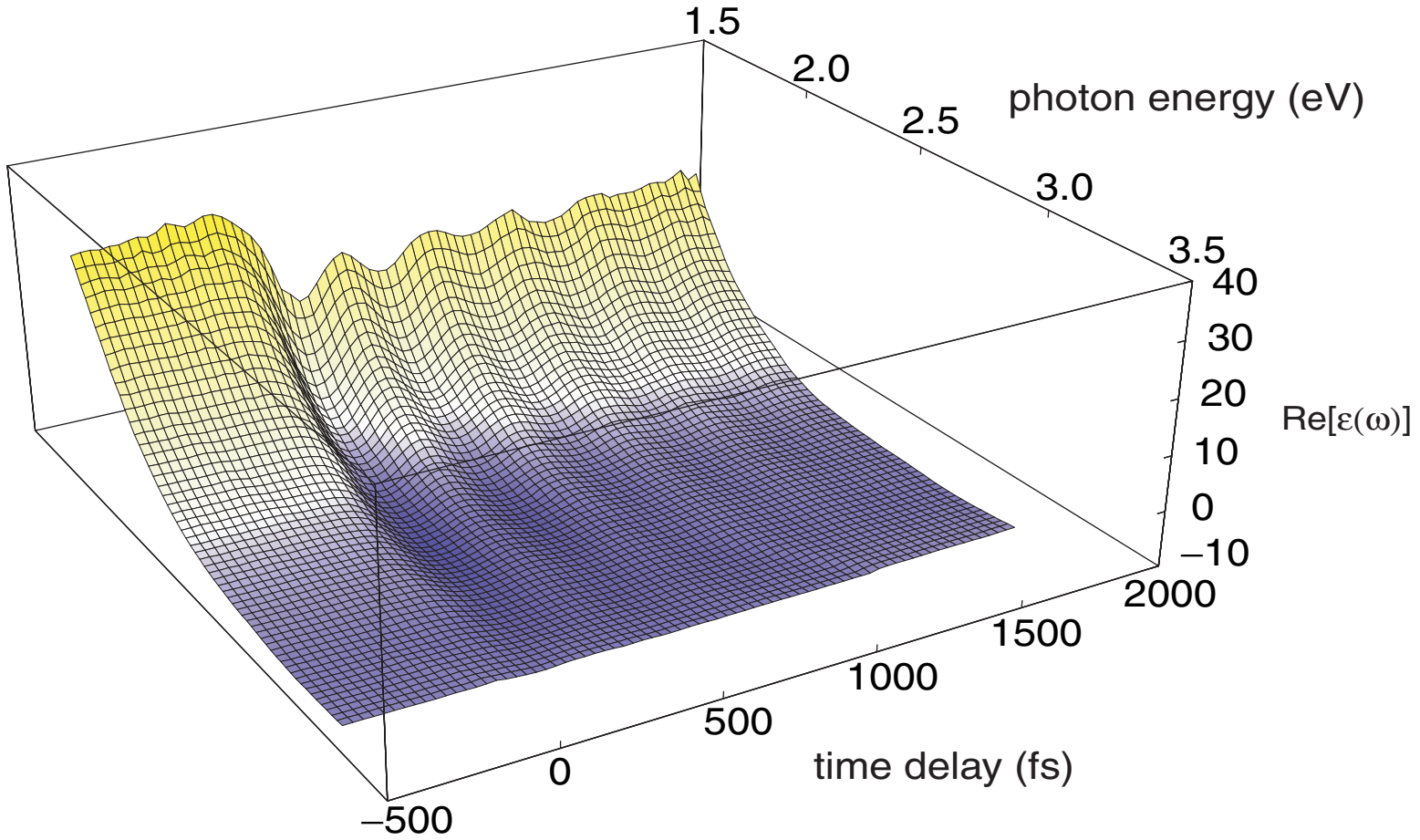
Resonance as a function of time

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



Coherent Phonons in Te

Experimental data on Te



Outline

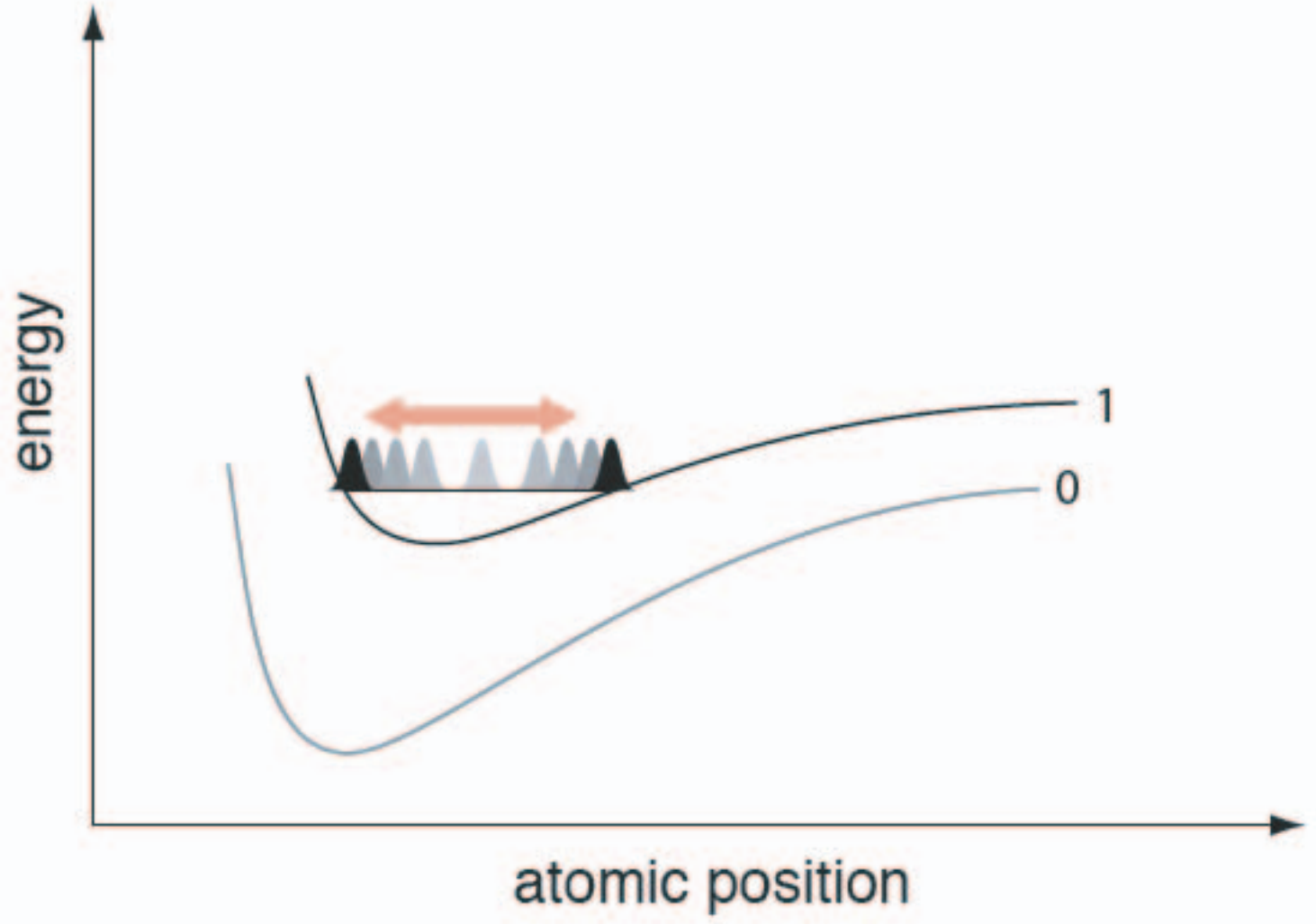
1. Introduction - Interaction of light with solids

2. Experimental process and technique

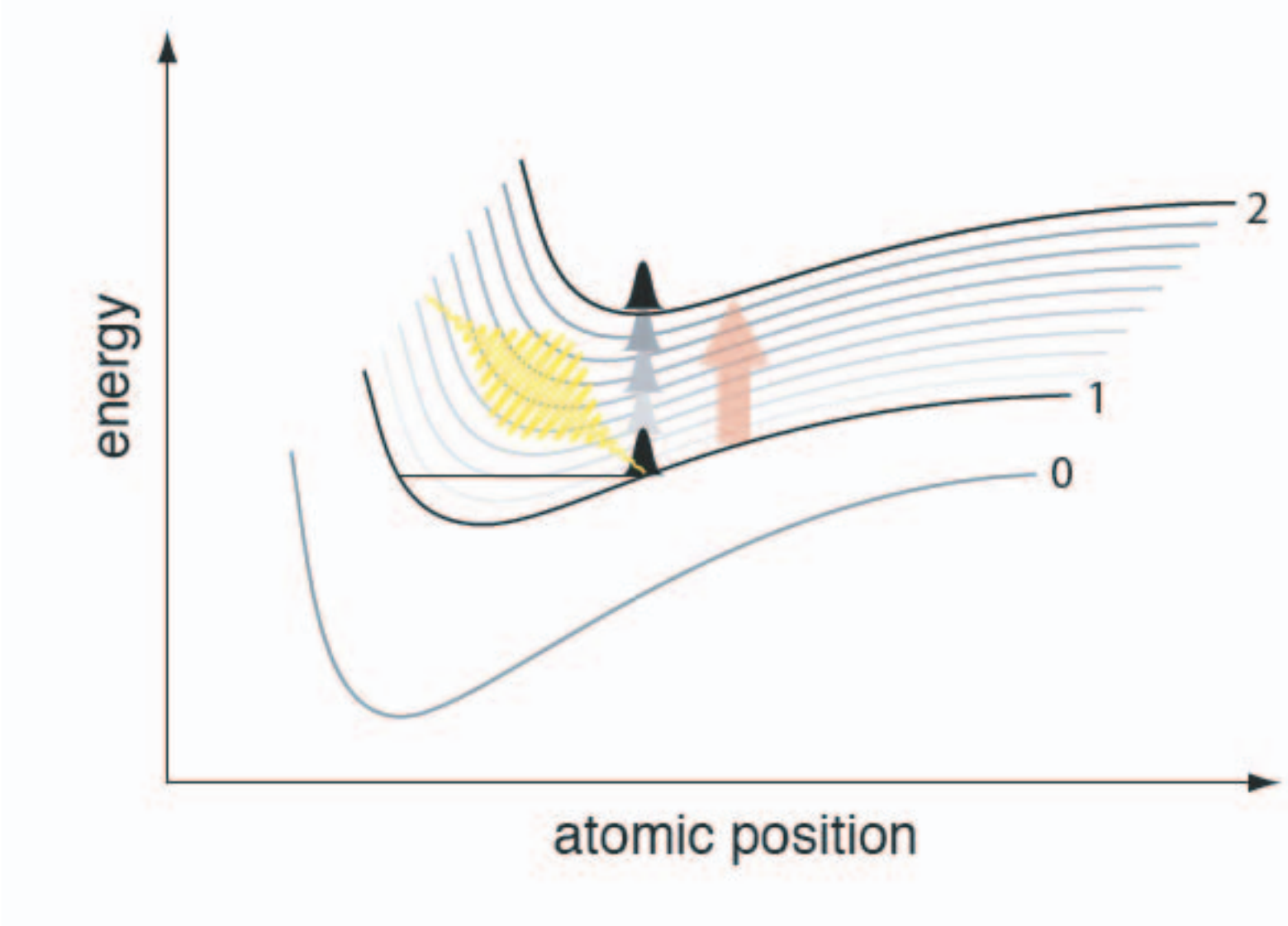
3. Excitation of high amplitude coherent phonons

4. **Optical control of coherent phonons**

Nuclear wavepacket oscillates around new equilibrium position

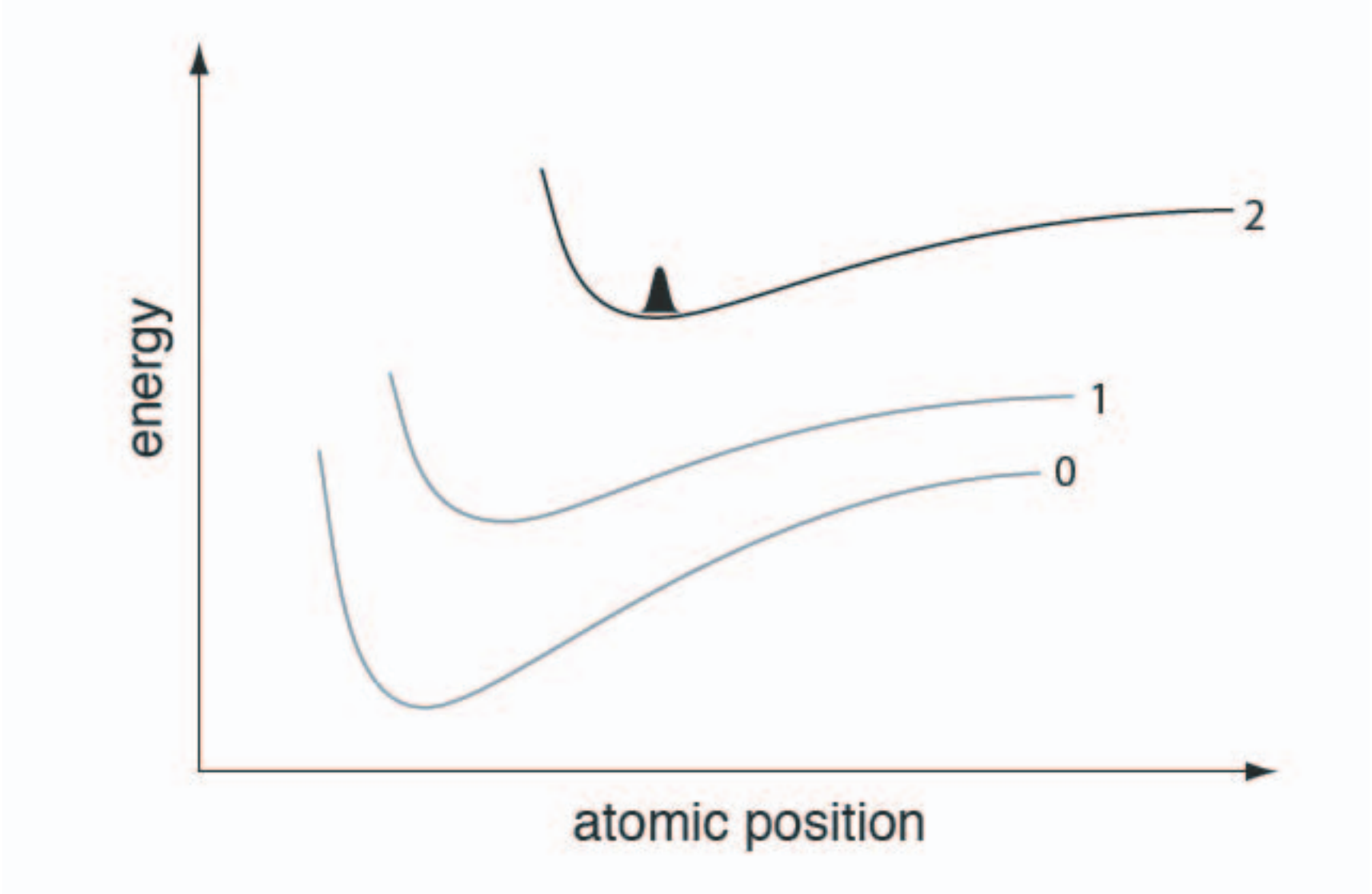


How about changing the equilibrium position once more?



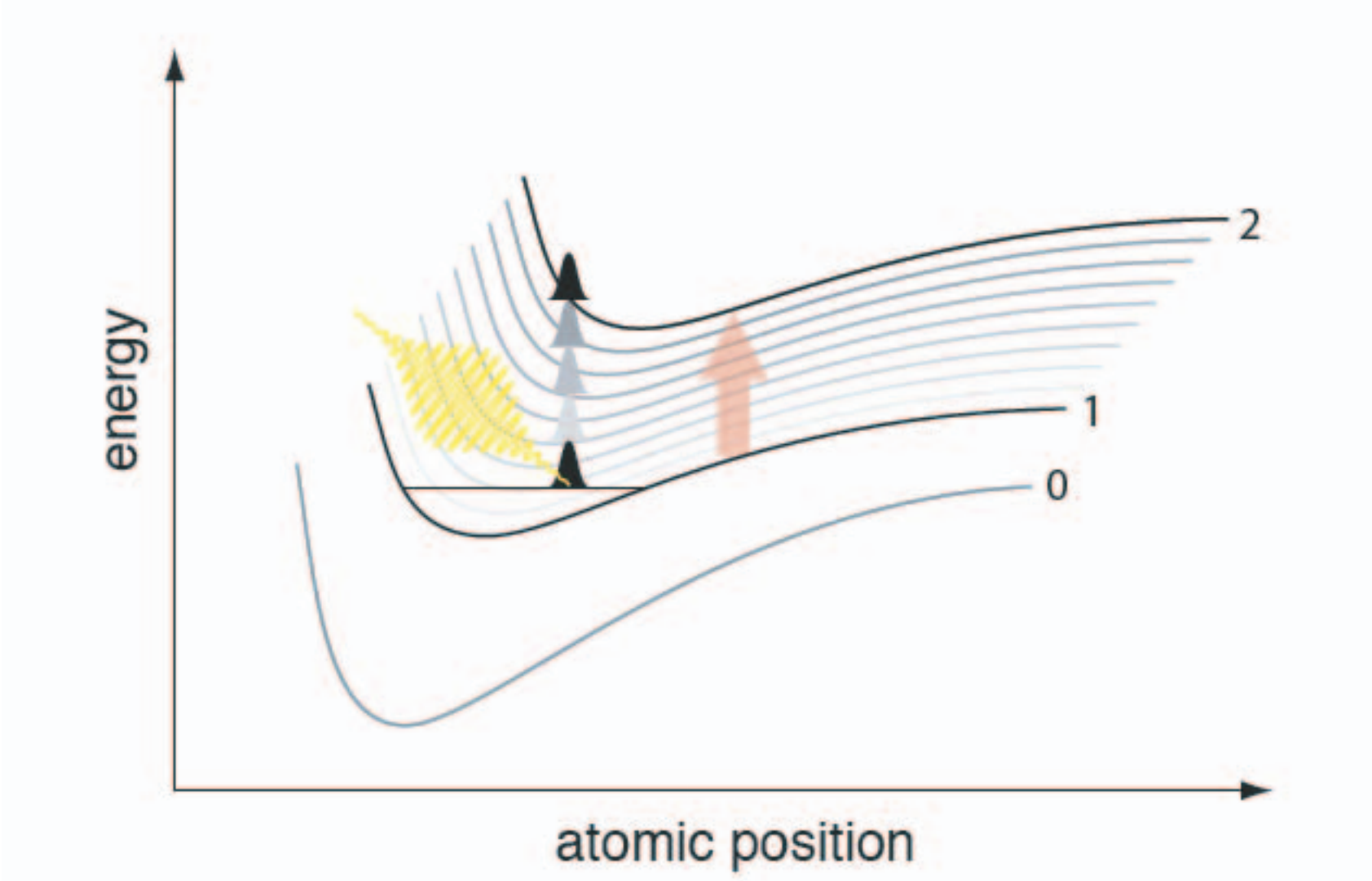
Optical control

Oscillations should stop if equilibrium position is now where outer turning point was before

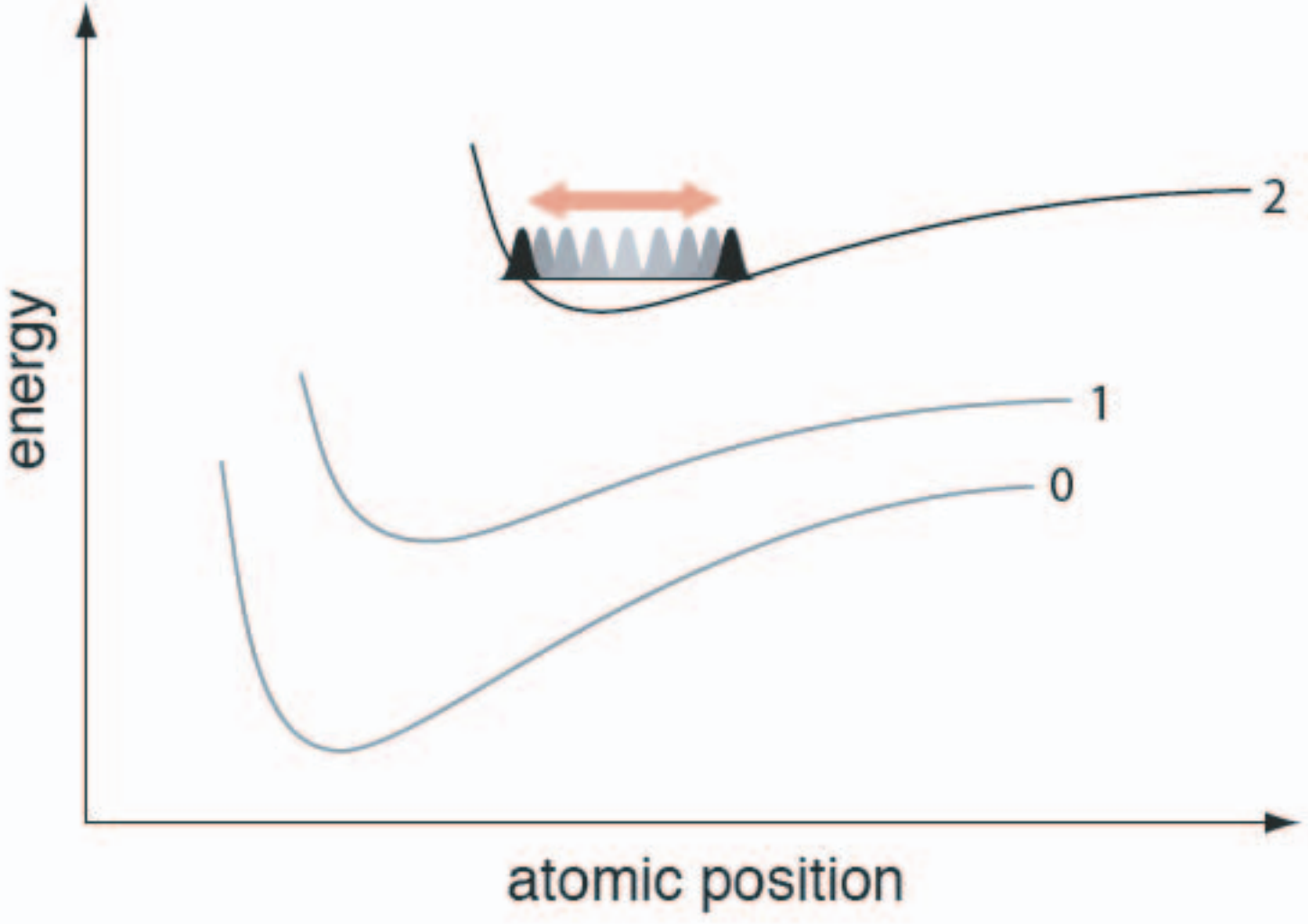


Optical control

If timing wrong...

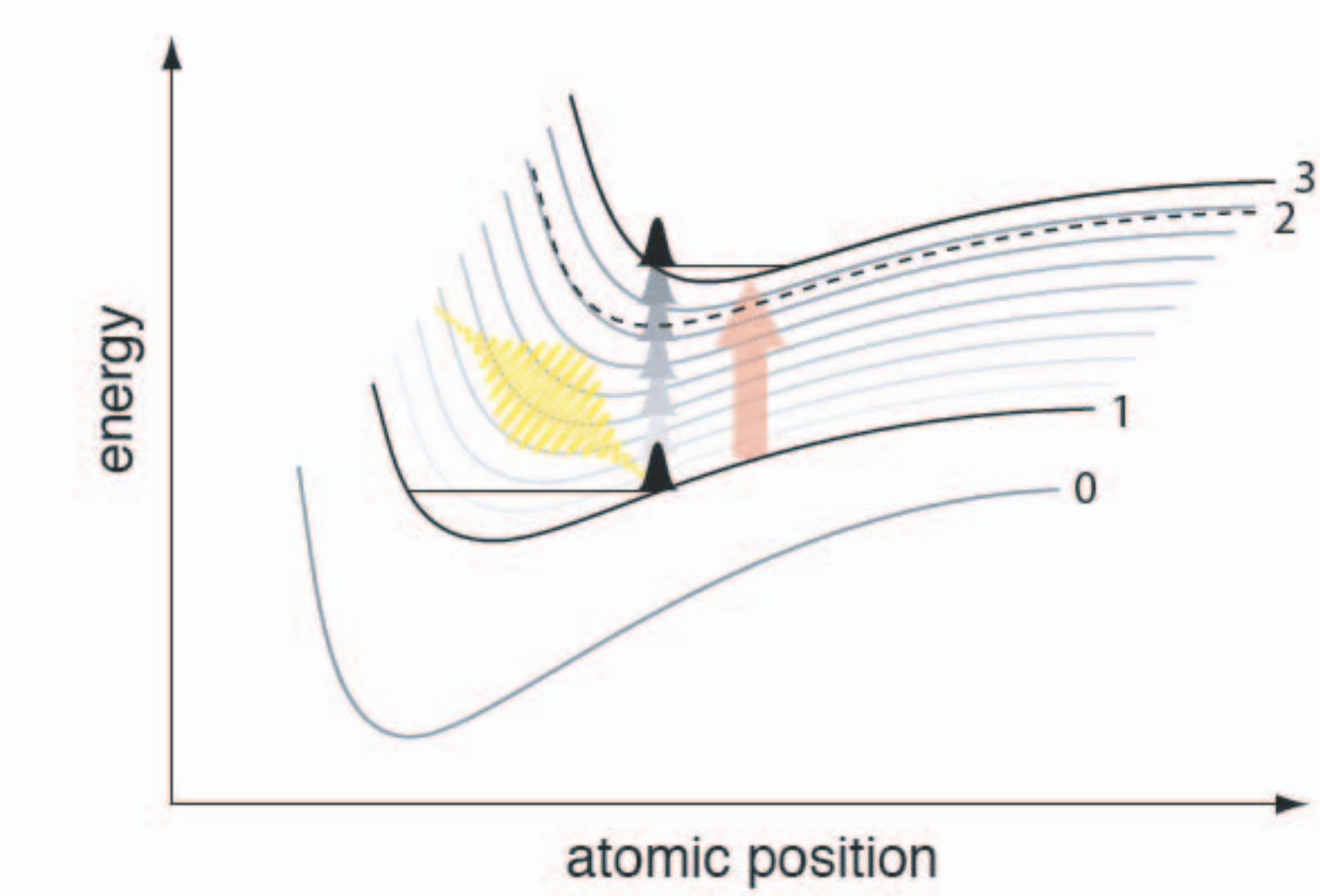


...oscillations continue



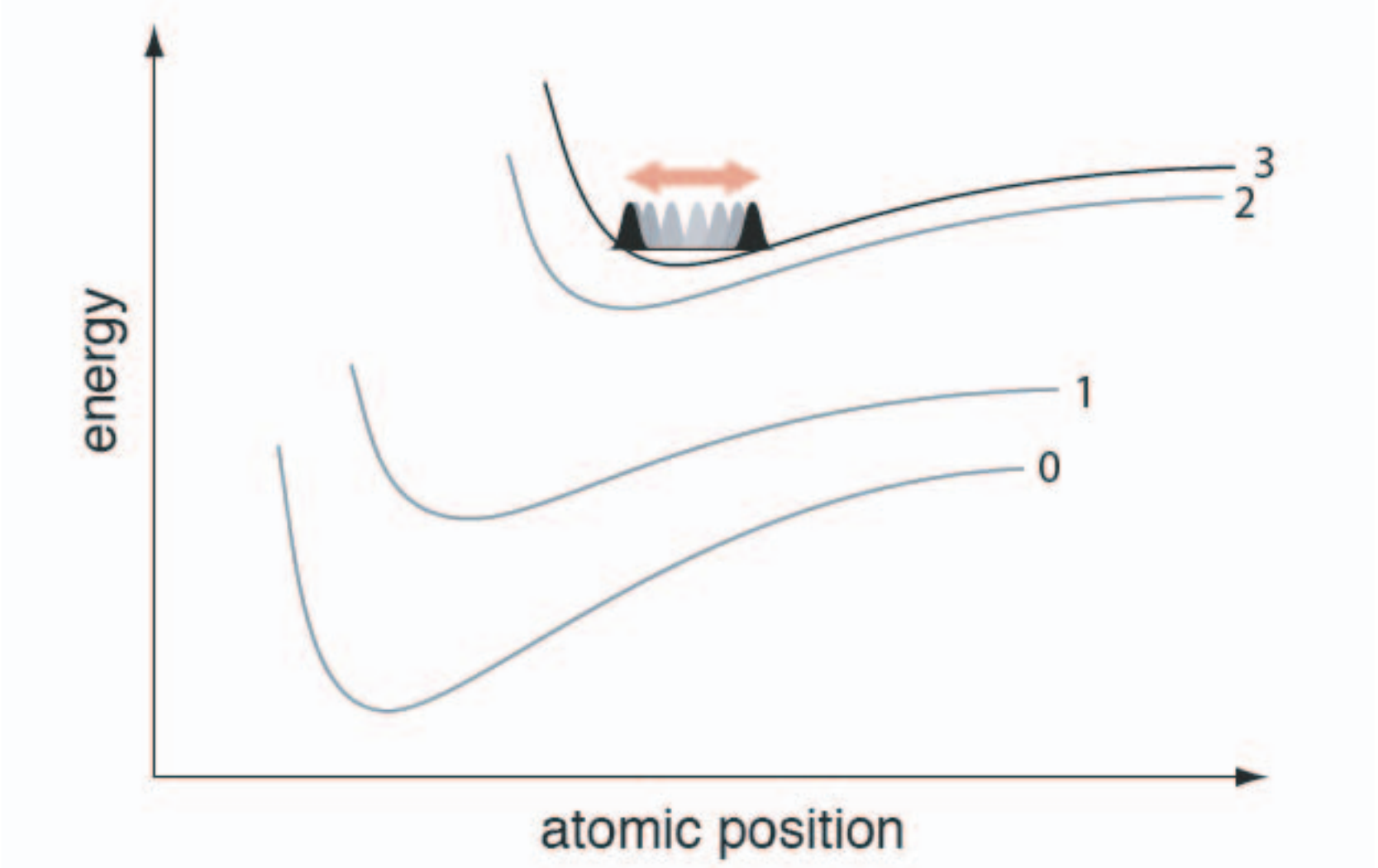
Optical control

If fluence wrong we access wrong potential surface...



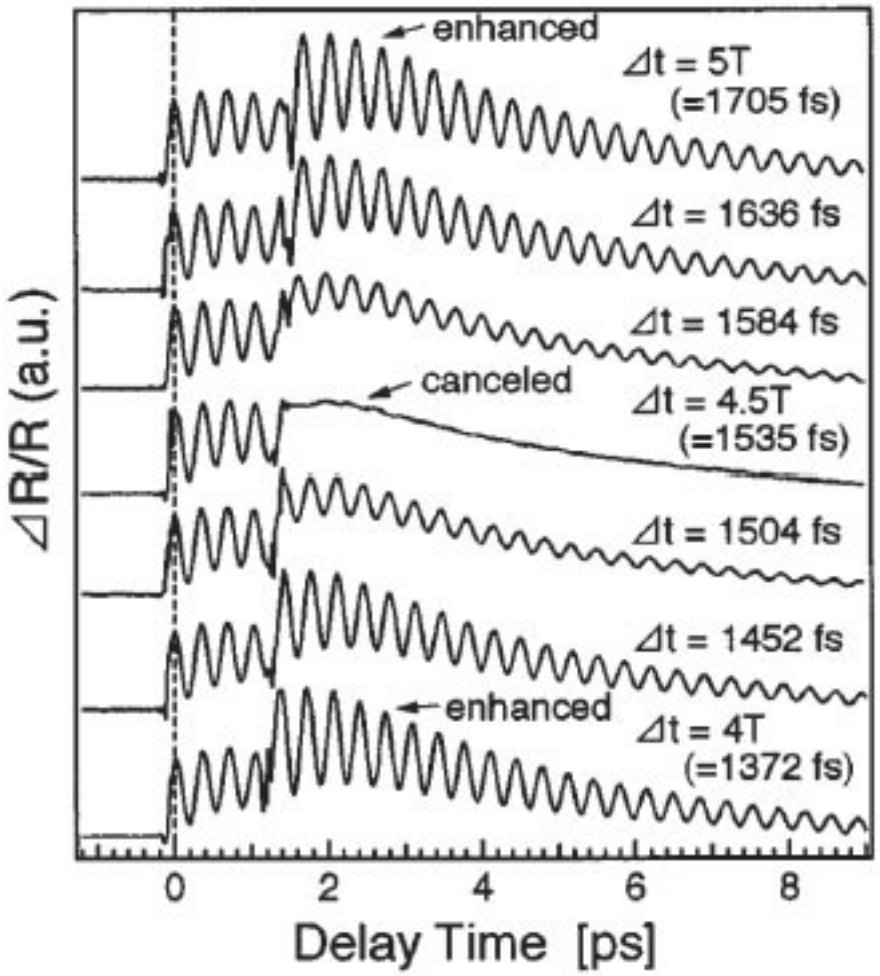
Optical control

...and oscillations continue



Optical control

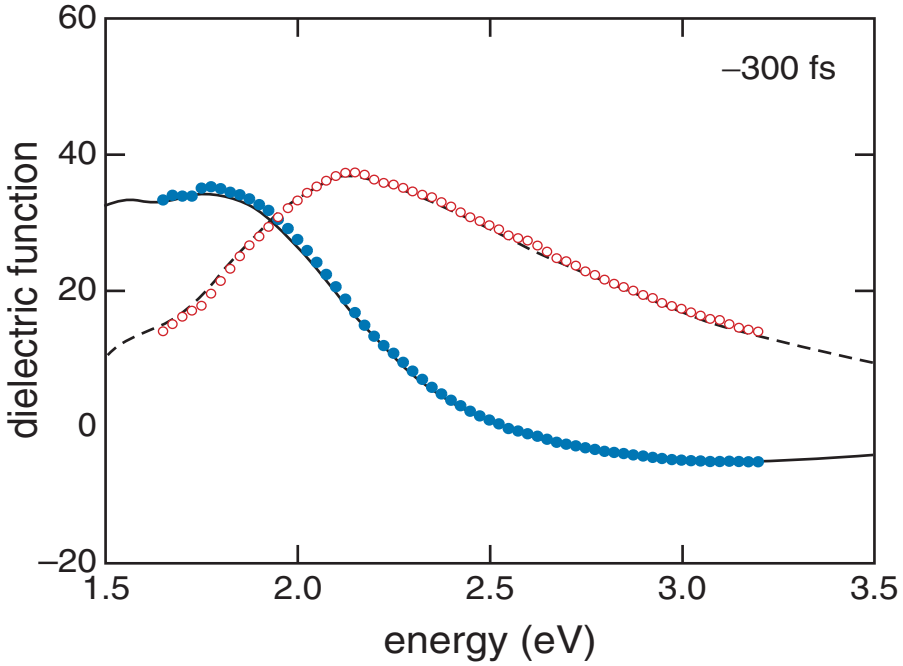
Low fluence experiment: bismuth

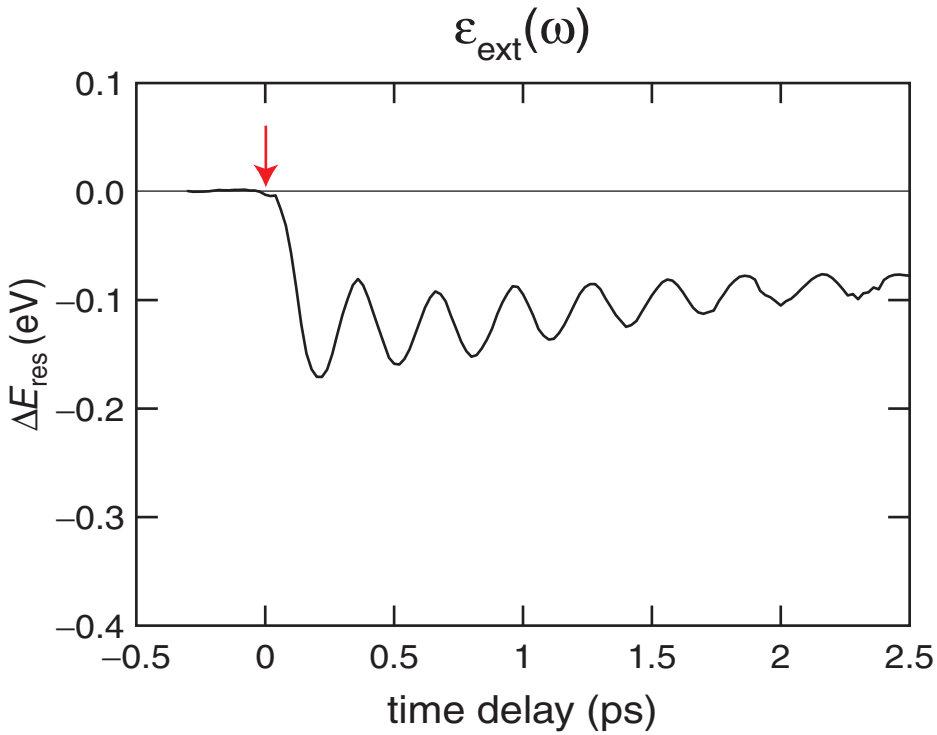


Hase et al., *Appl. Phys. Lett.* **69**, 2474 (1996)

Optical control

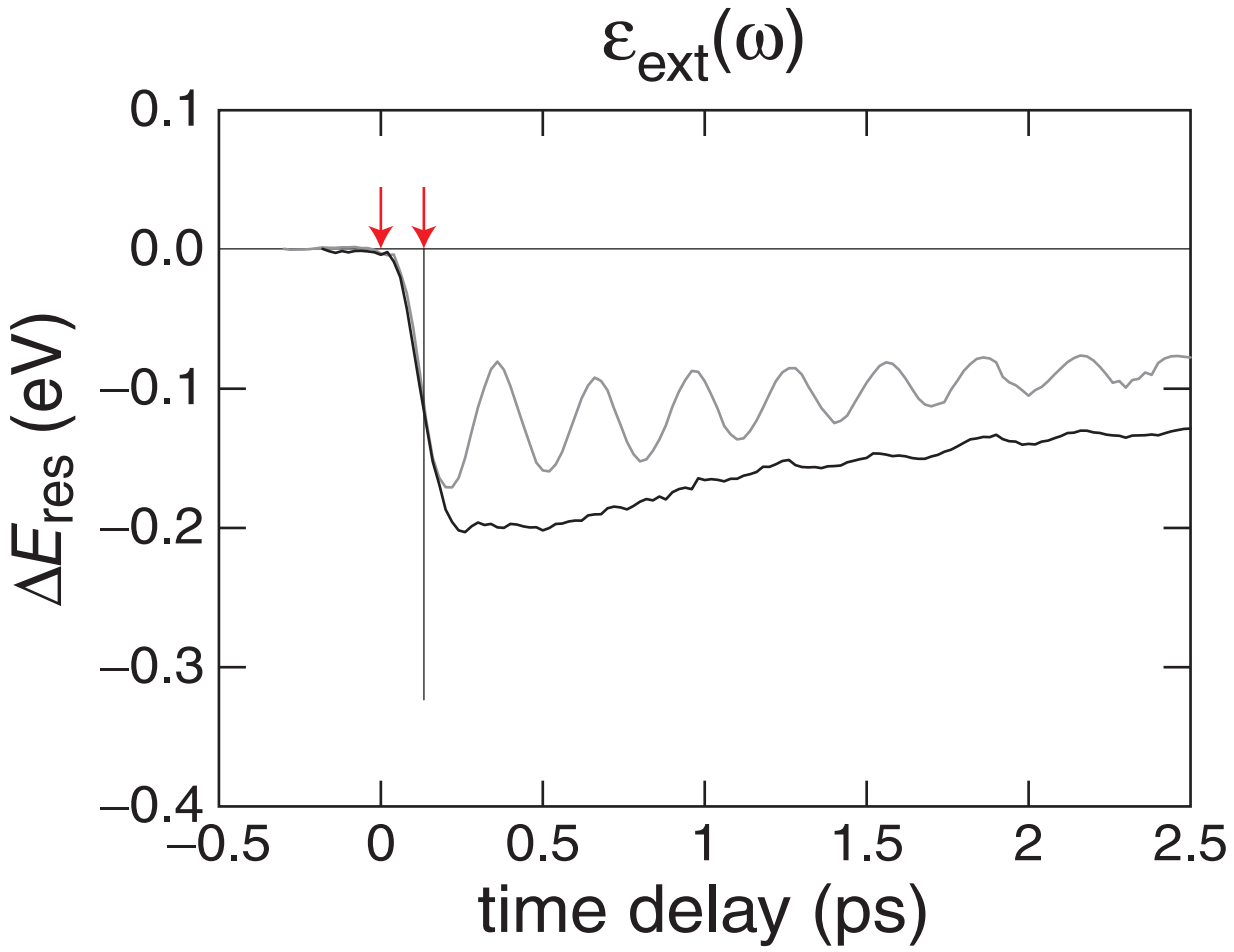
$$F_1 = 0.57\ F_{th}$$





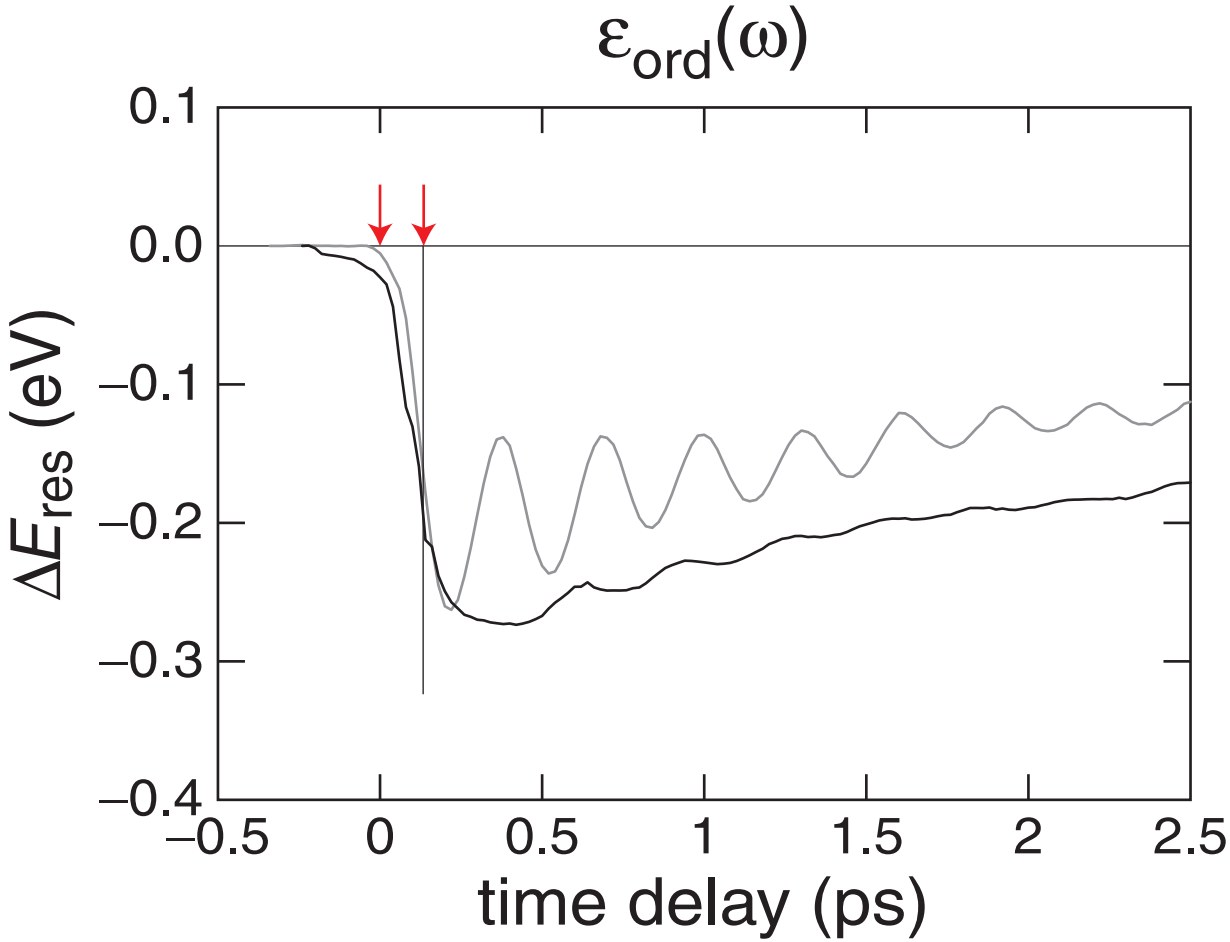
Optical control

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



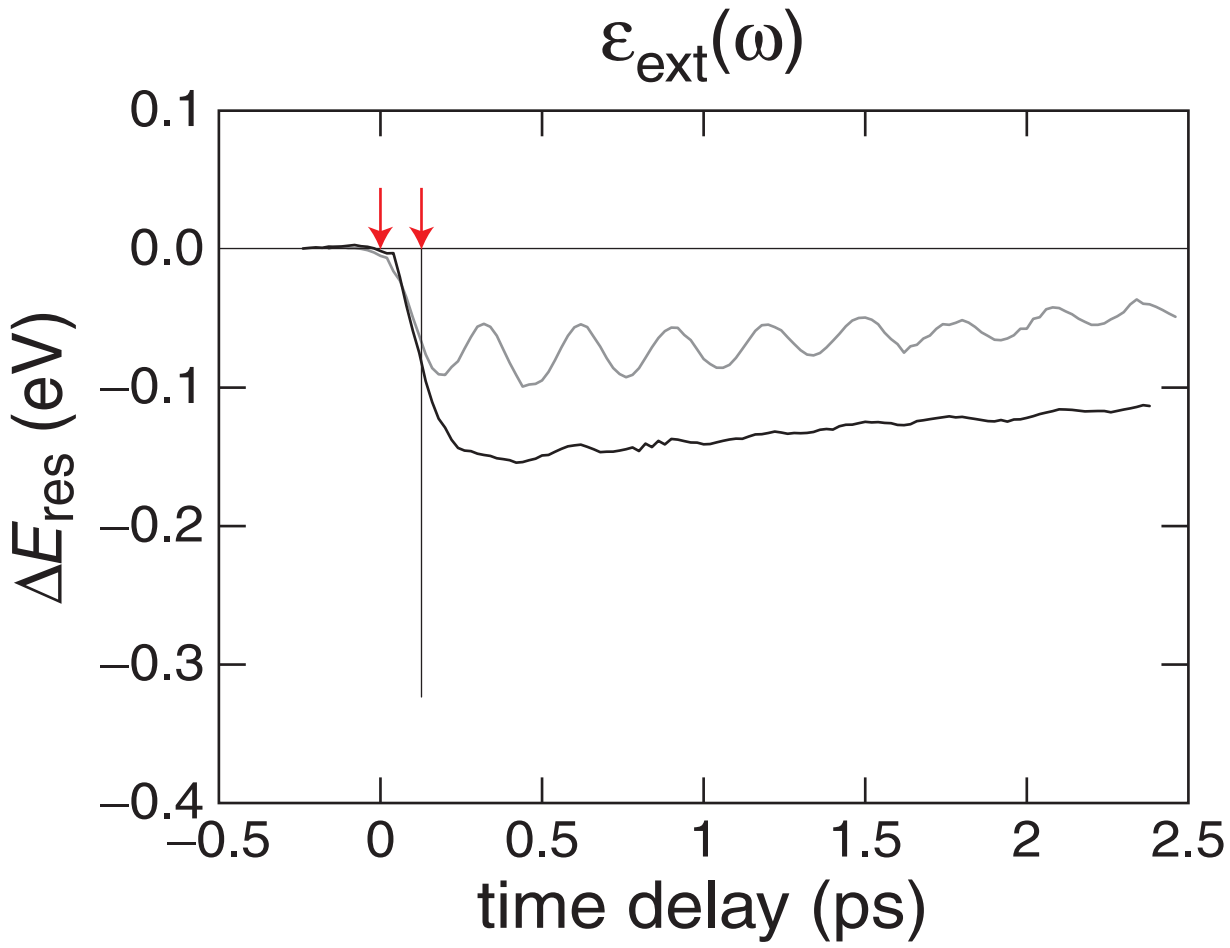
Optical control

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



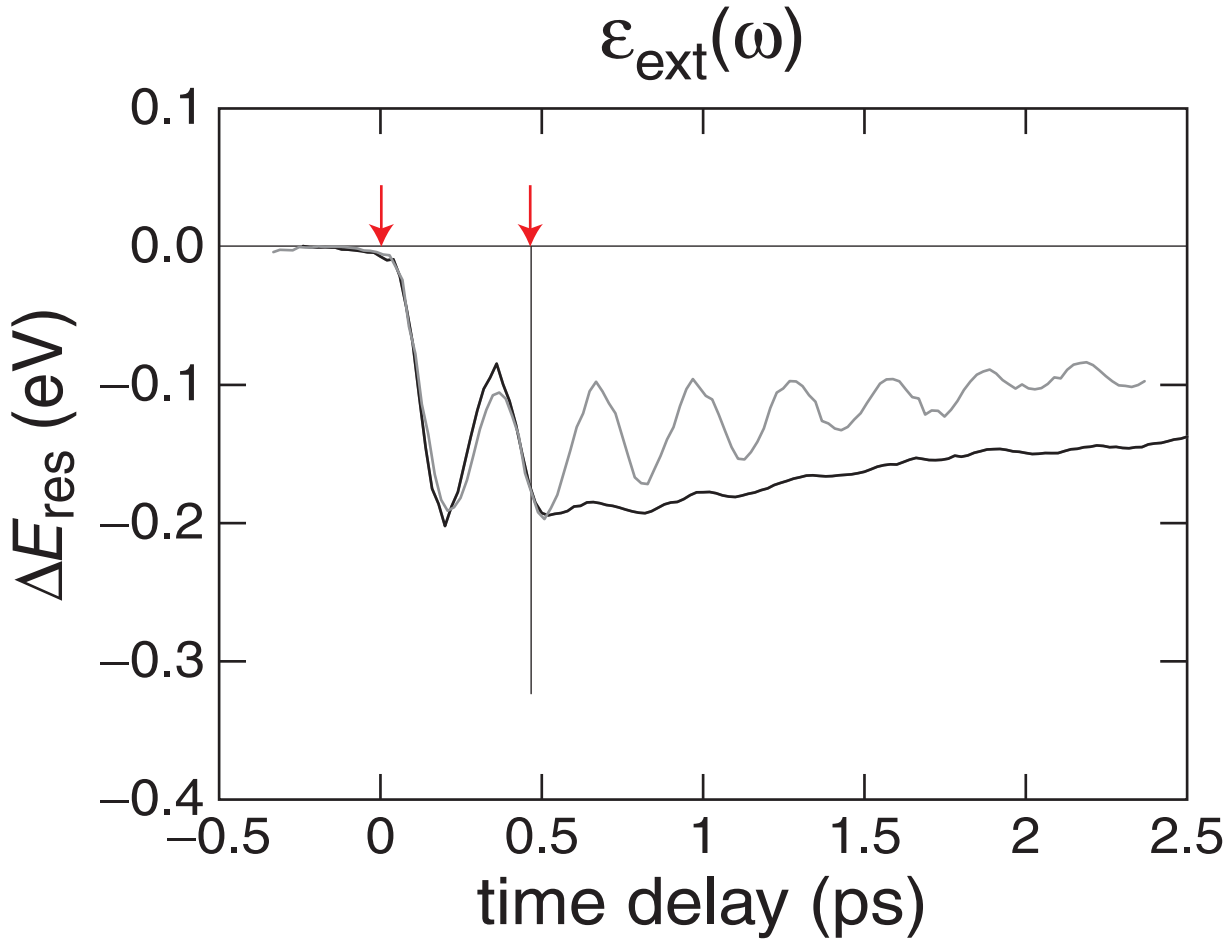
Optical control

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



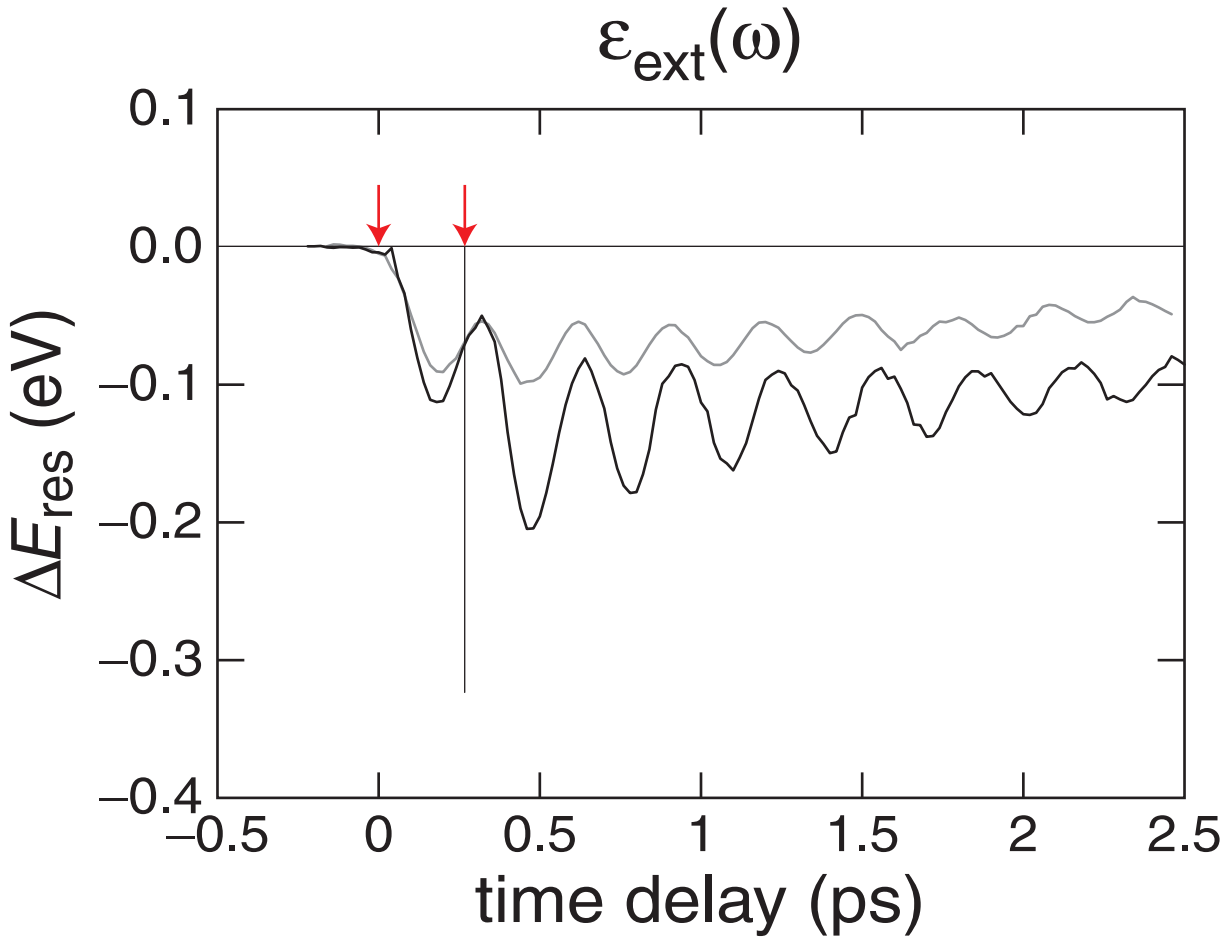
Optical control

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



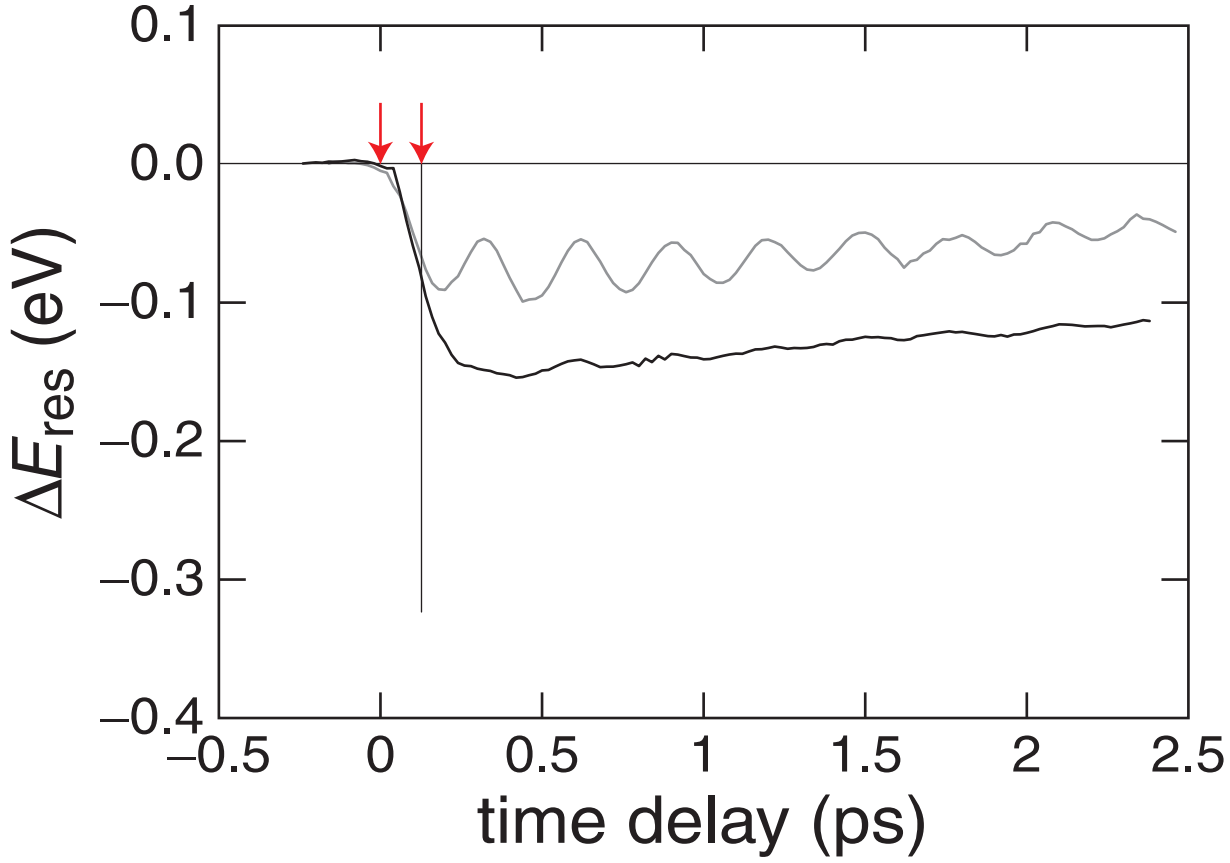
Optical control

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



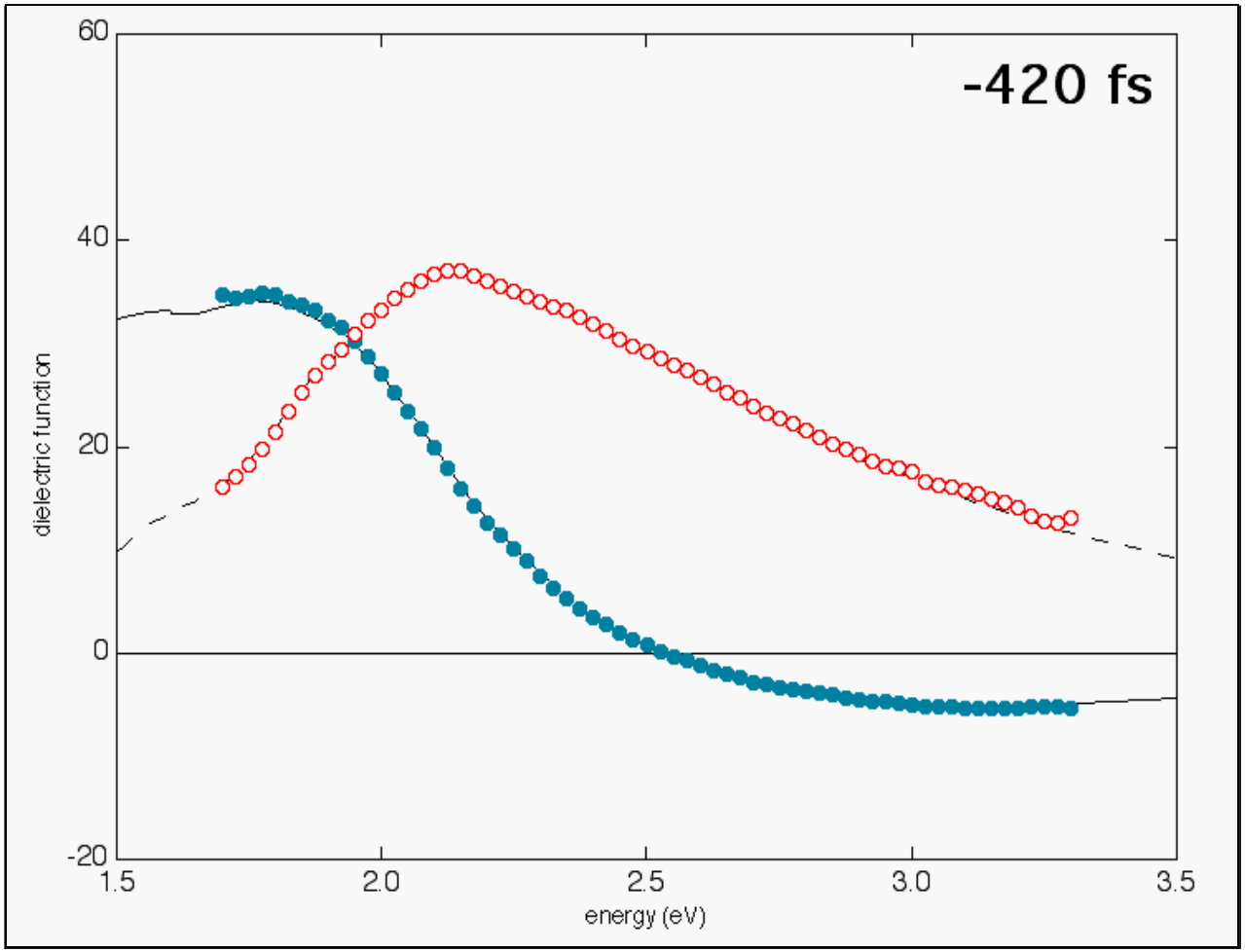
Optical control

Cancellation on first peak



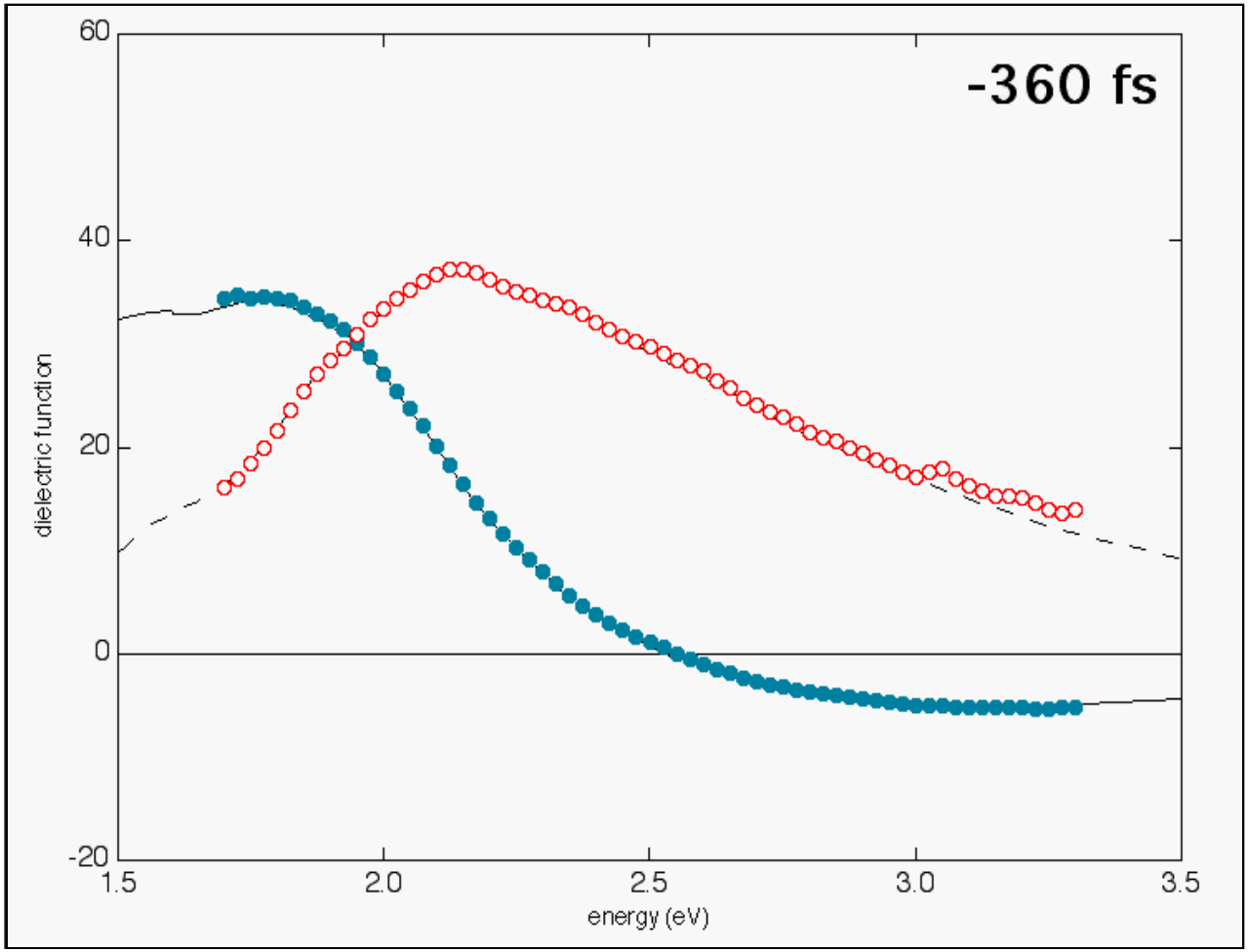
Optical control

Te under 0.71 F_{th} excitation

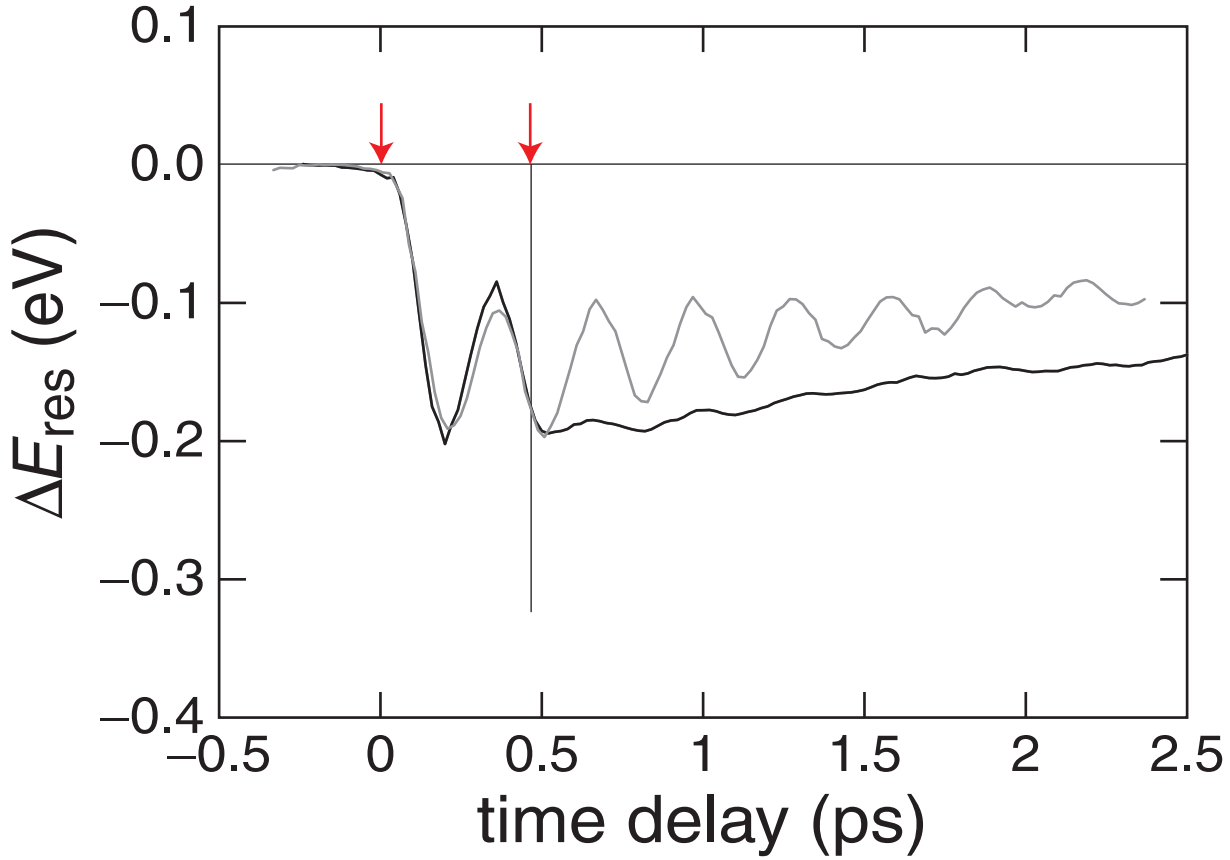


Optical control

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

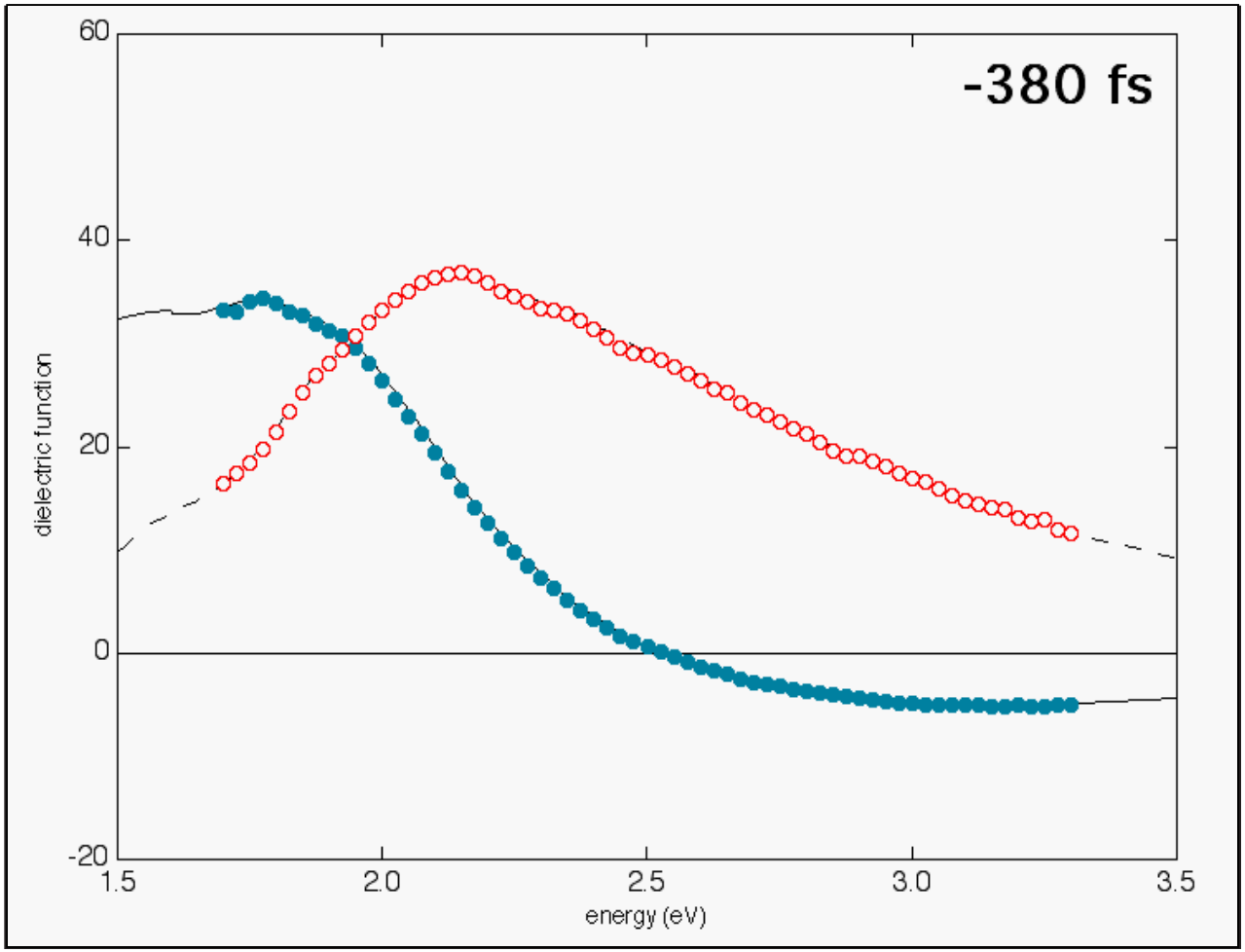


Cancellation on second peak

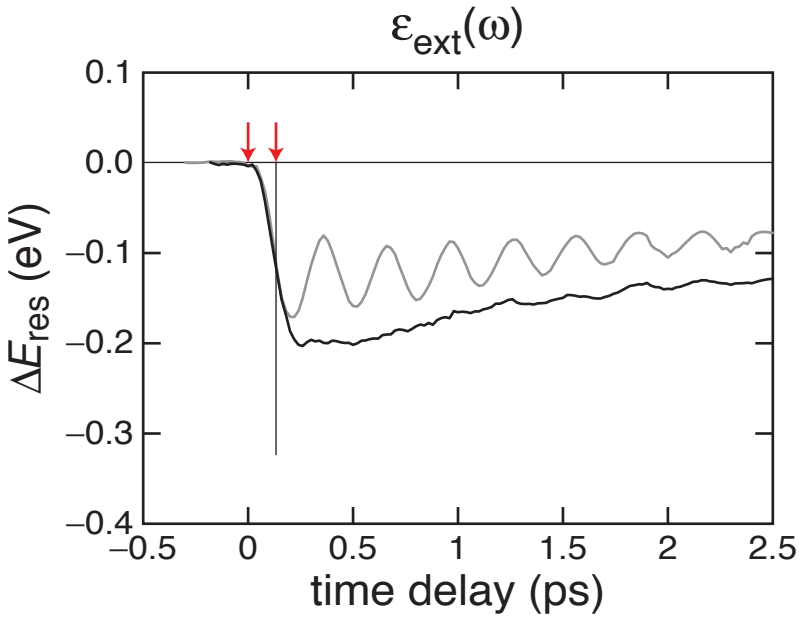


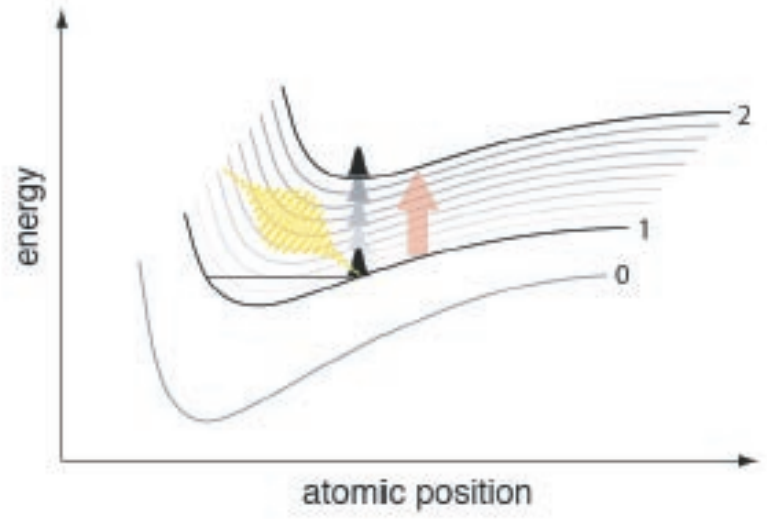
Optical control

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



Optical control



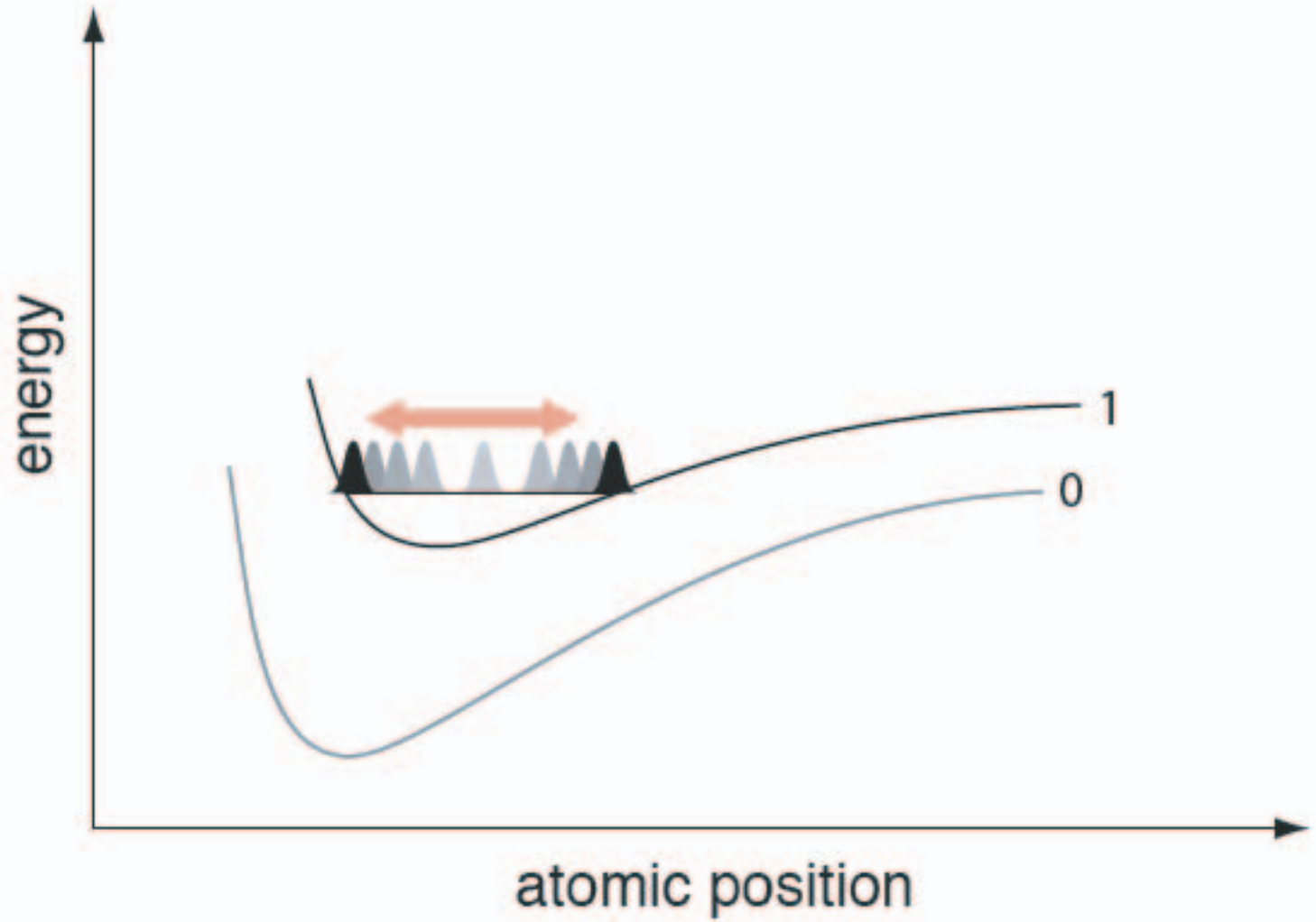


1. Second pulse always earlier than expected

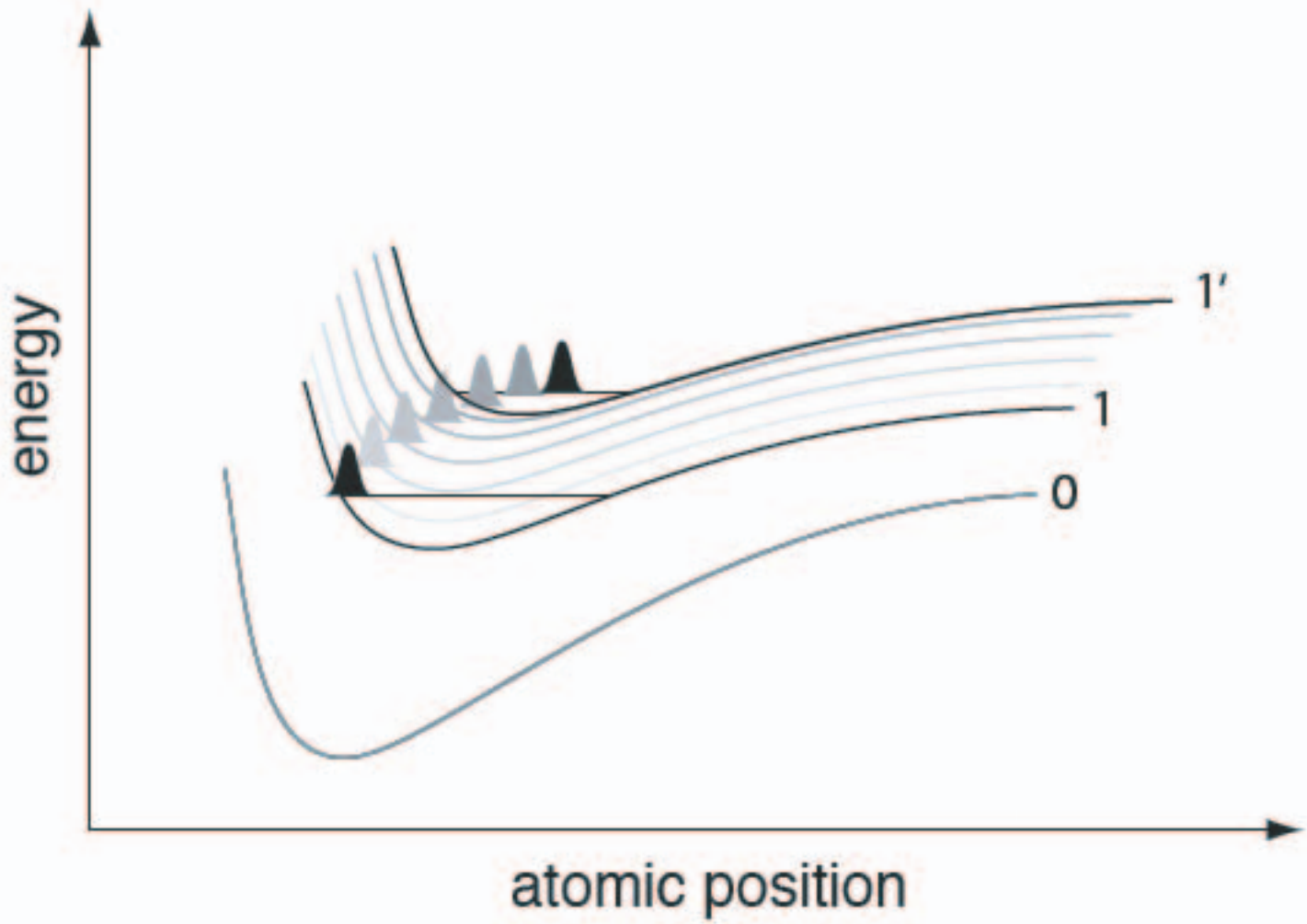
2. Resonance redshifts more than in the single pulse case

Optical control

Assumption: nuclear motion does not affect potential landscape



In reality the potential changes while lattice oscillates



Summary

1. **Our experimental apparatus measures broadband dielectric function with femtosecond time resolution**

2. **High amplitude coherent phonons can be excited and monitored in the time domain**

3. **We can optically control the position of the atoms in the lattice of Te and similar materials**

Thanks to the Mazur group