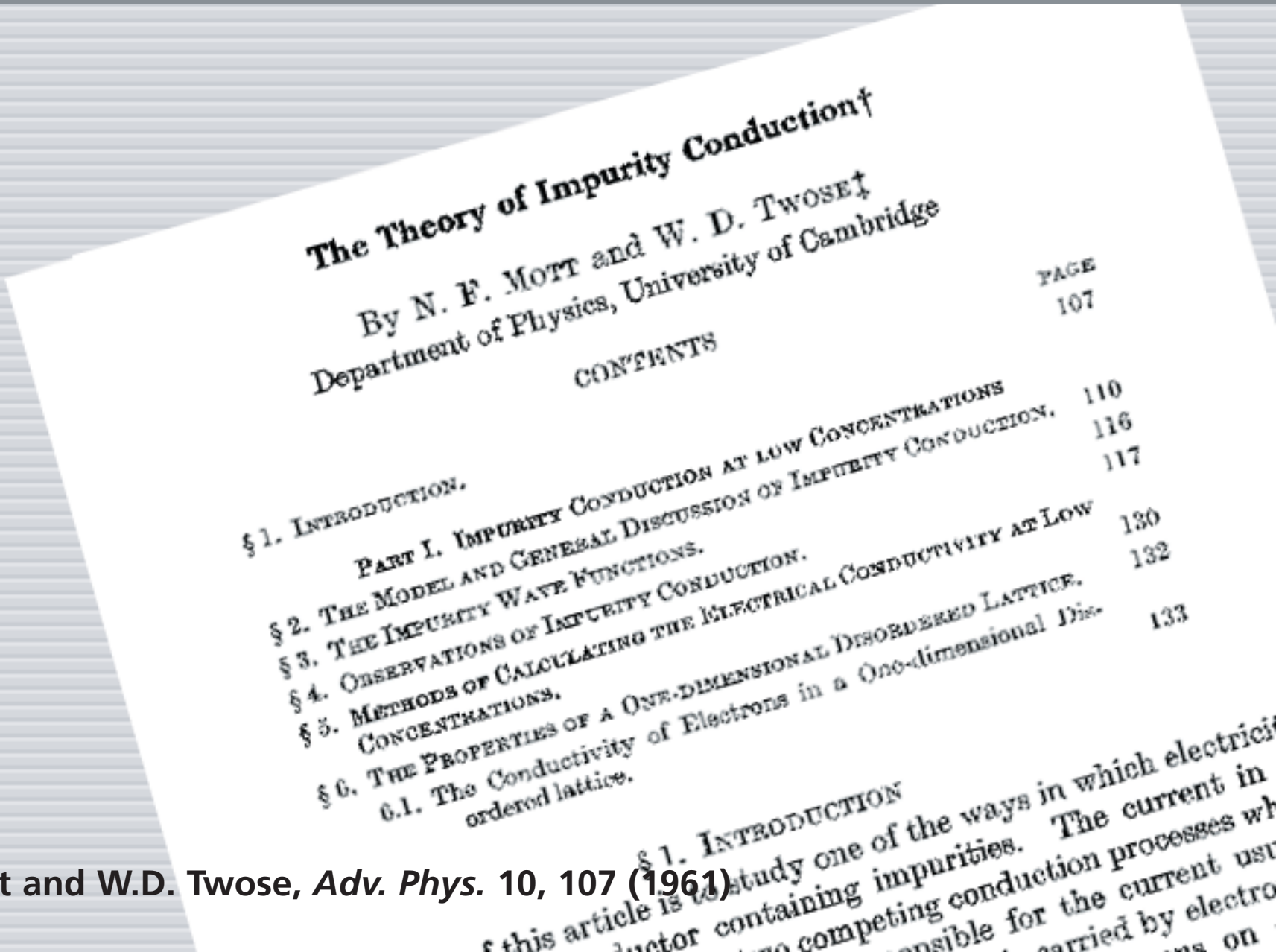
Oscillating between semiconductor and metal: moving ions faster than electron wave functions can spread

Chris Roeser
Maria Kandyla
Albert Kim
Arantza Mendioroz

SUNY Buffalo
18 April 2003

Introduction



The Theory of Impurity Conduction†

By N. F. Mott and W. D. Twose‡
Department of Physics, University of Cambridge

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6.1. The Conductivity of Electrons in a One-dimensional Disordered lattice.

§ 1. Introduction

f this article is to study one of the ways in which electrici
ductor containing impurities. The current in
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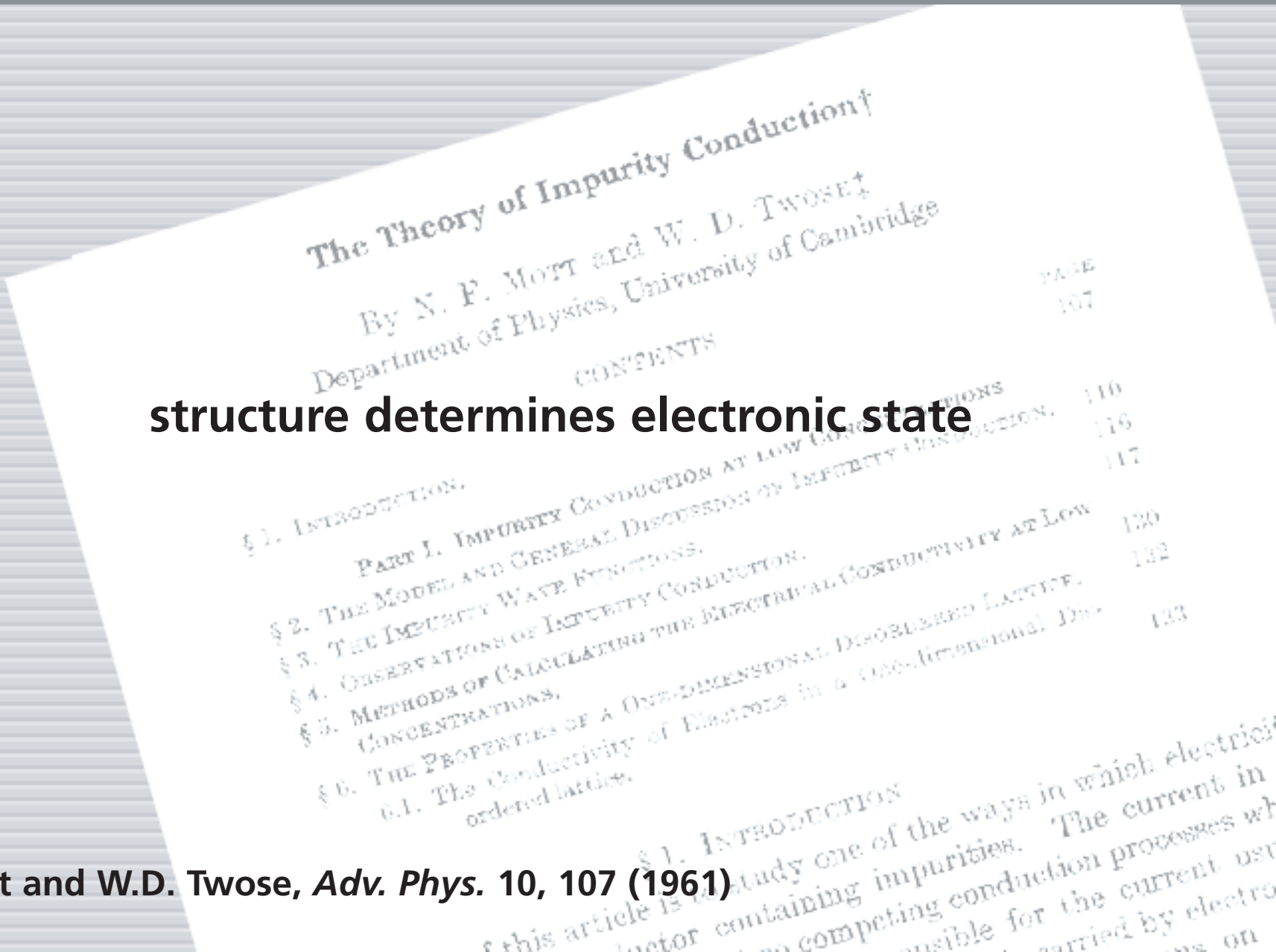
nsible for the current usu
carried by electro
ons on

N. Mott and W.D. Twose, *Adv. Phys.* 10, 107 (1961)

Introduction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6.1. The Conductivity of Electrons in a One-dimensional Disordered lattice.

§ 1. Introduction

structure determines electronic state

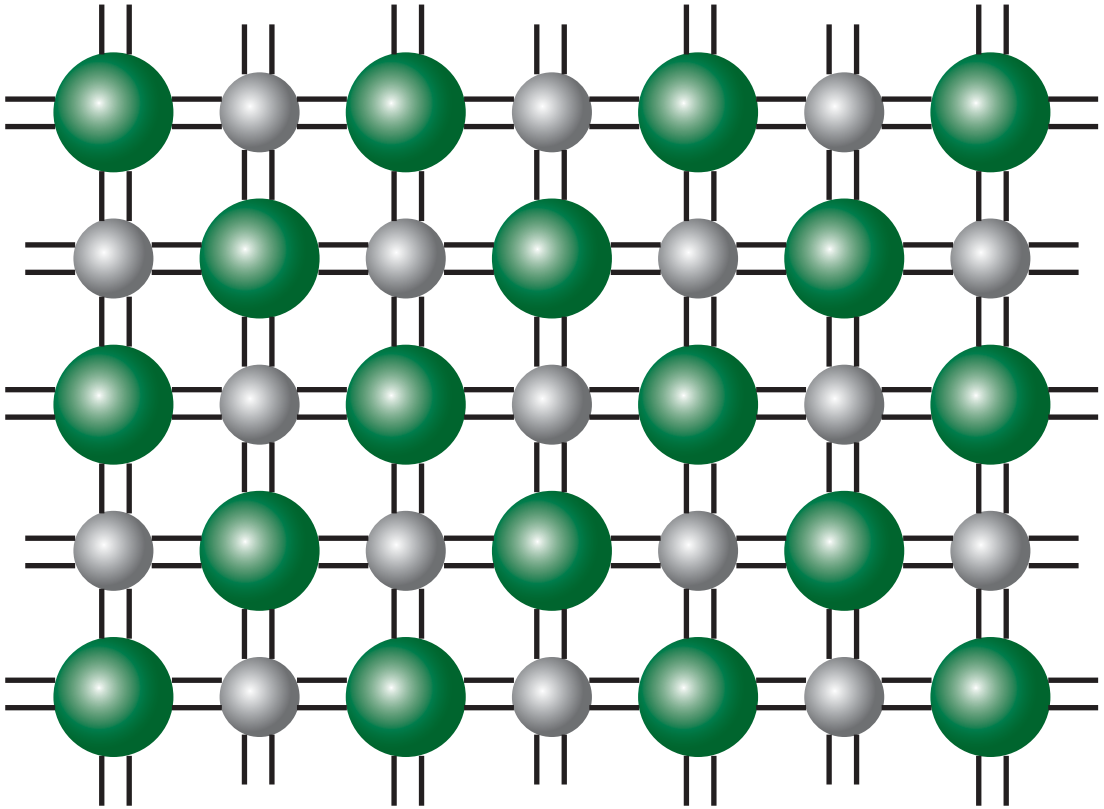
N. Mott and W.D. Twose, *Adv. Phys.* 10, 107 (1961)

Introduction

short laser pulses can drive structural transitions

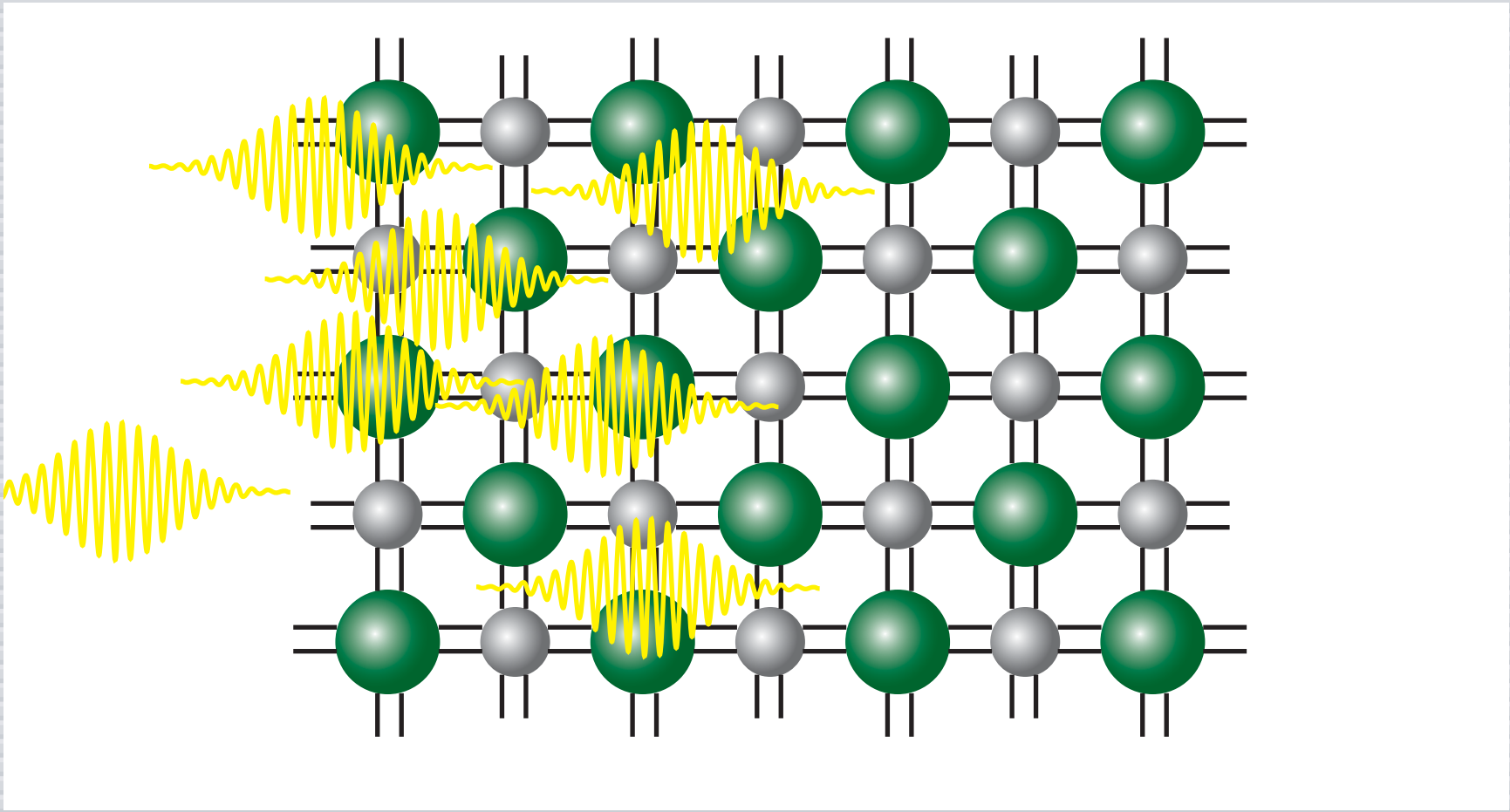
Introduction

how do femtosecond laser pulses alter a solid?

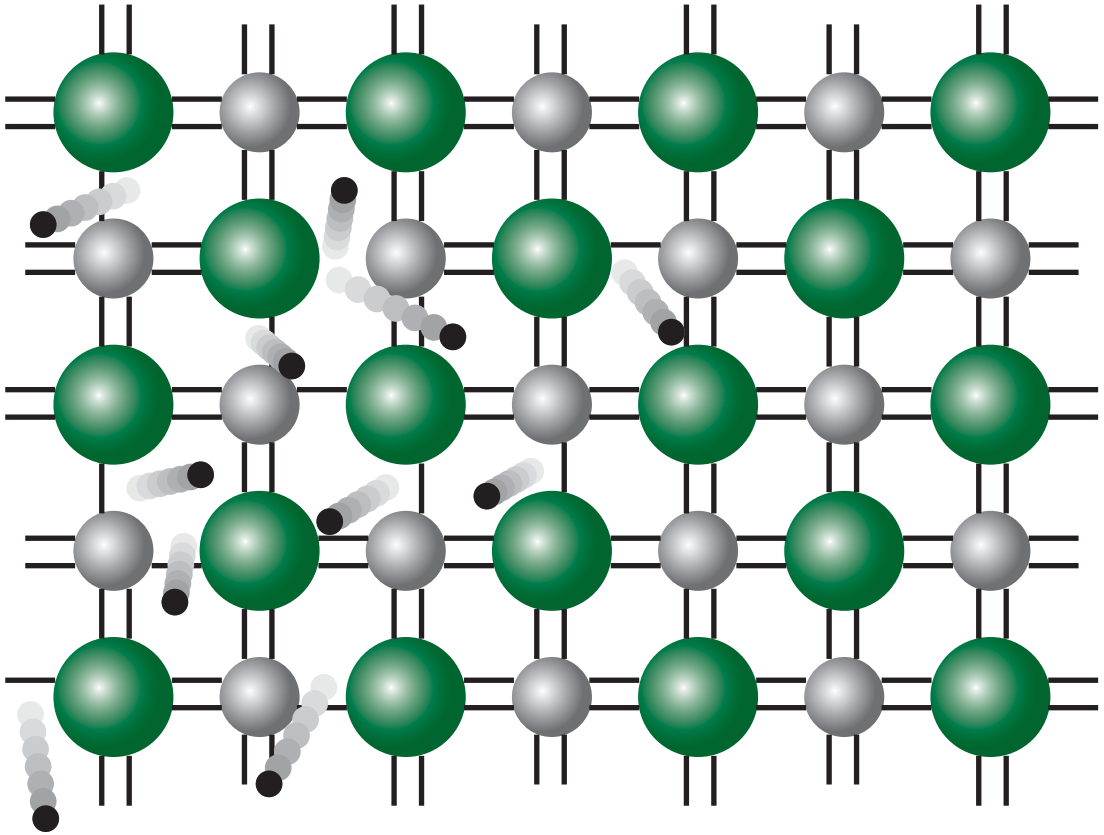


Introduction

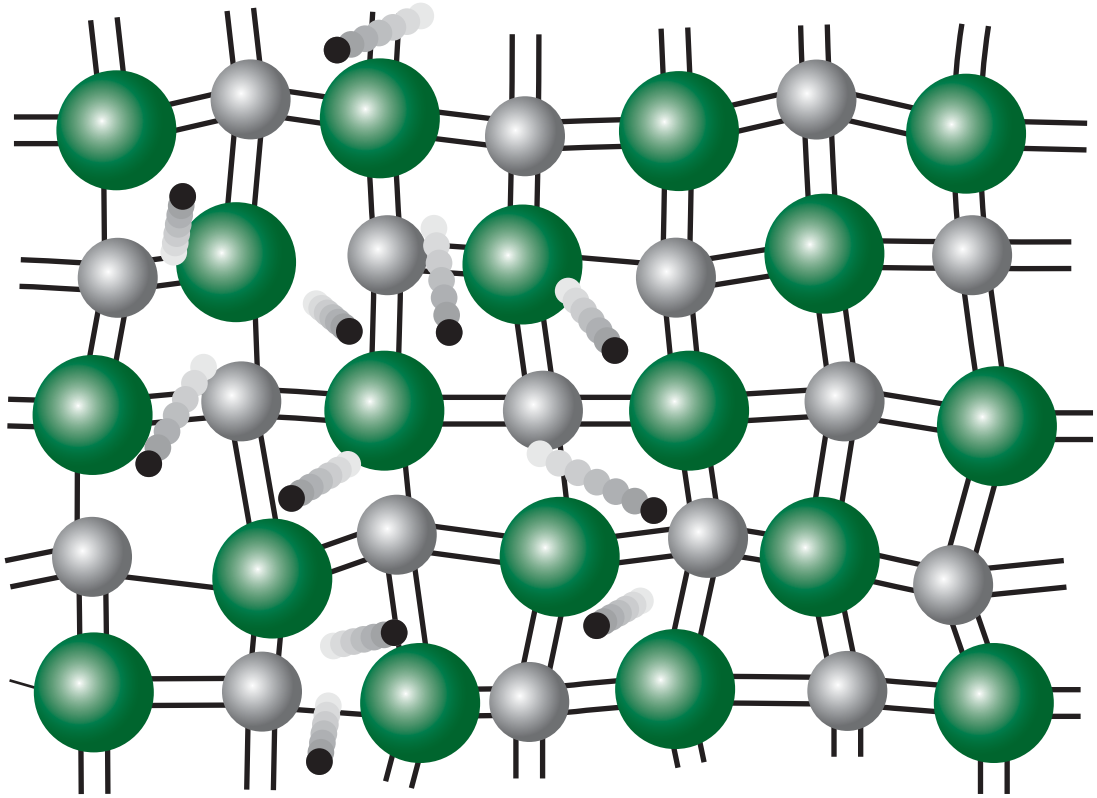
photons excite valence electrons…



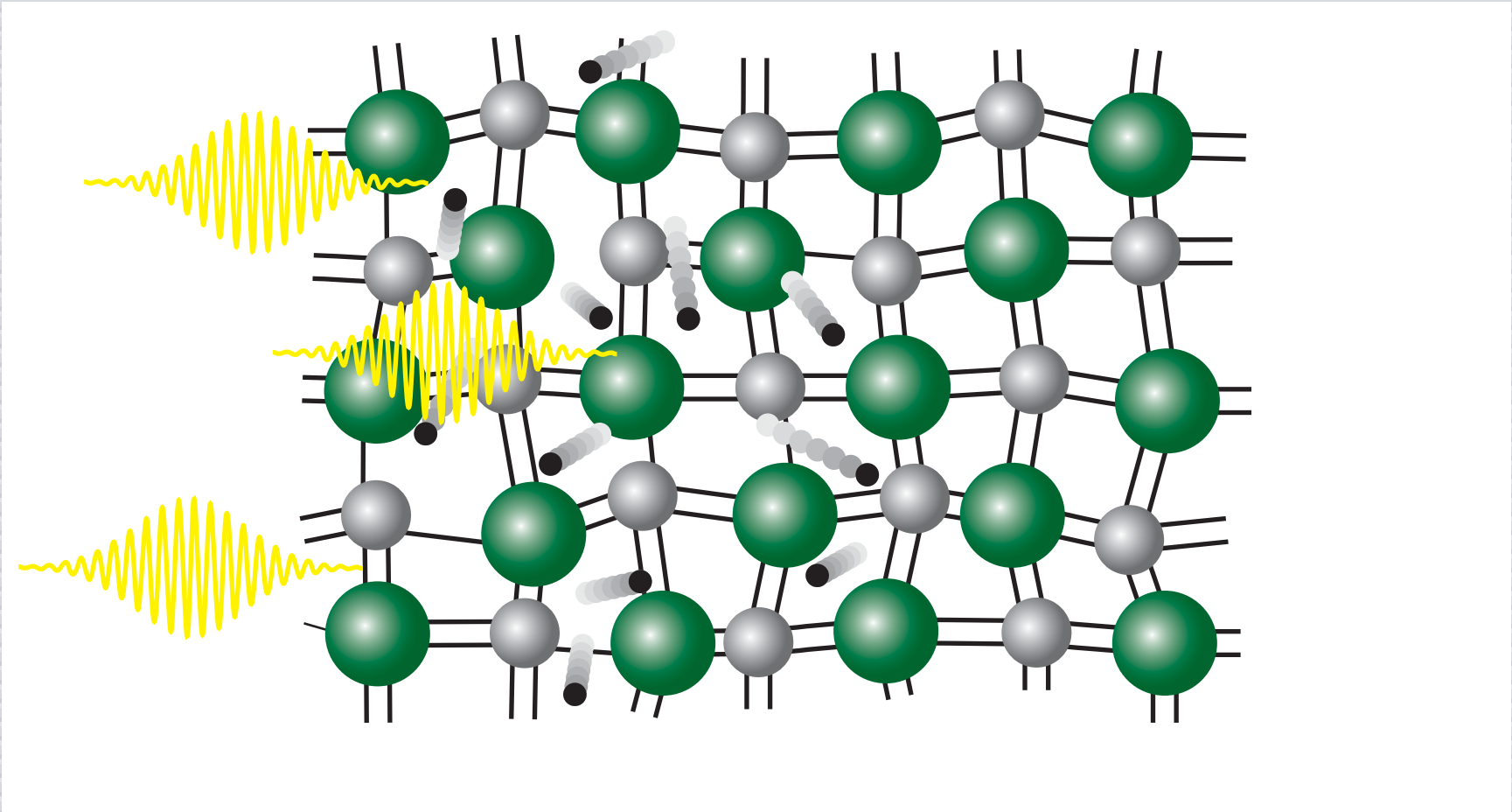
… and create free electrons…



… causing electronic and structural changes…

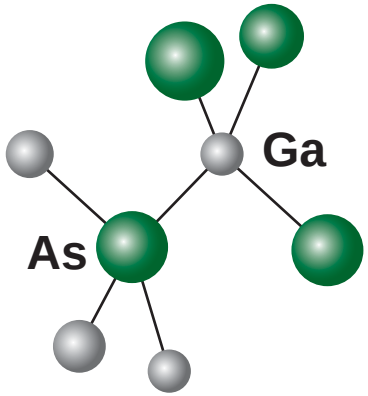


… which we detect with a second laser pulse



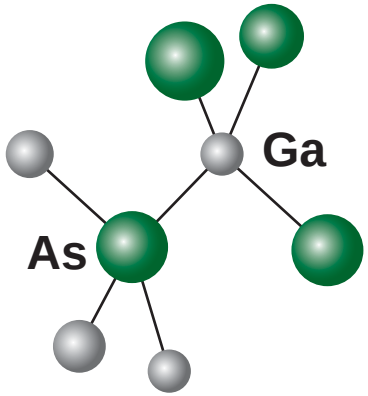
Introduction

structure

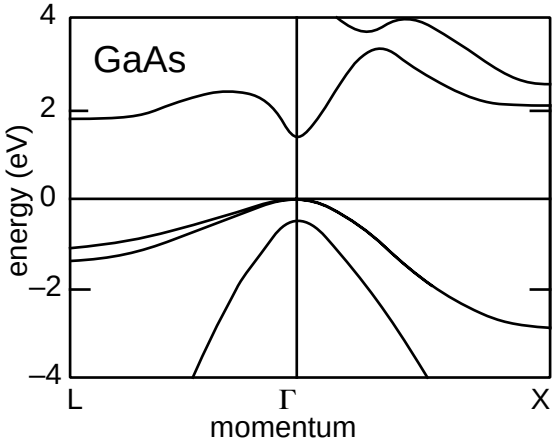


Introduction

structure

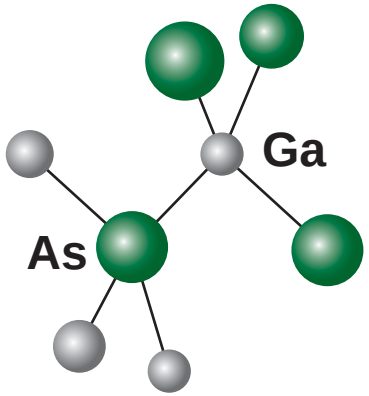


band structure

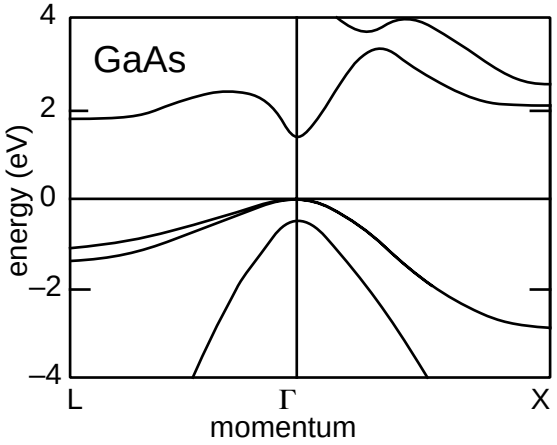


Introduction

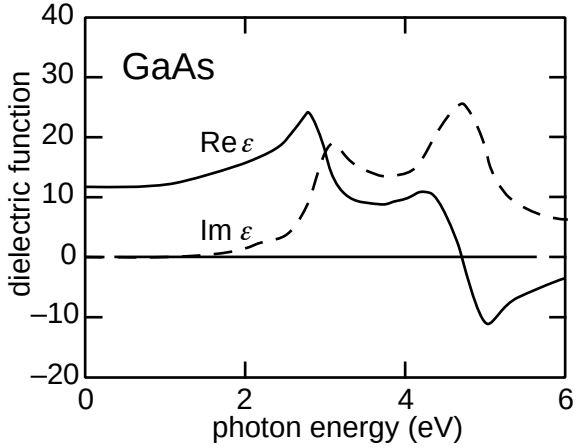
structure



band structure

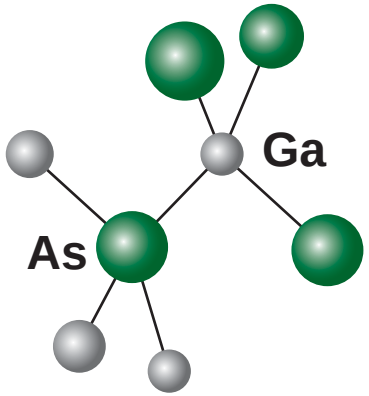


dielectric function



Introduction

structure



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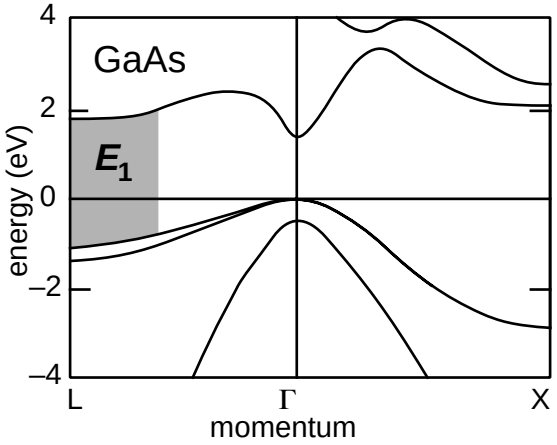
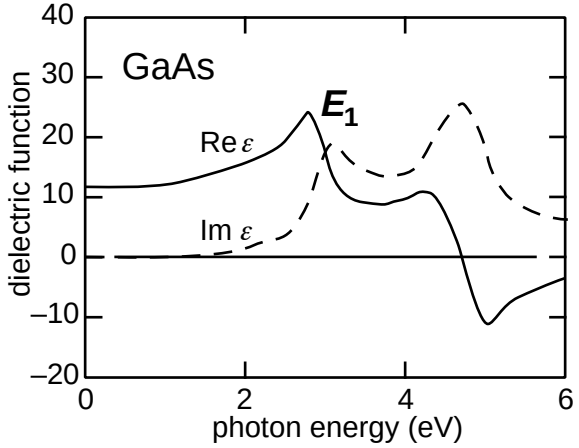
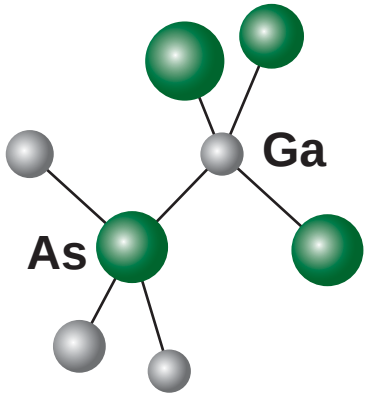


dielectric function

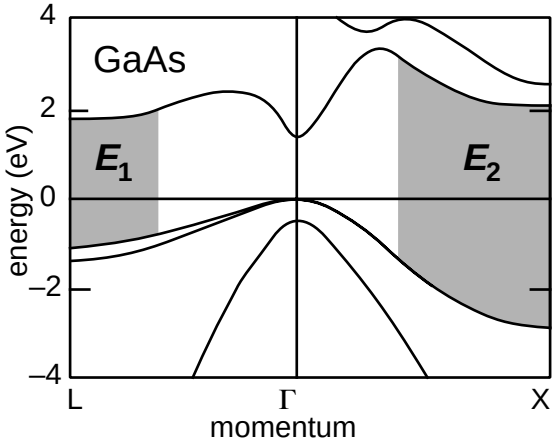


Introduction

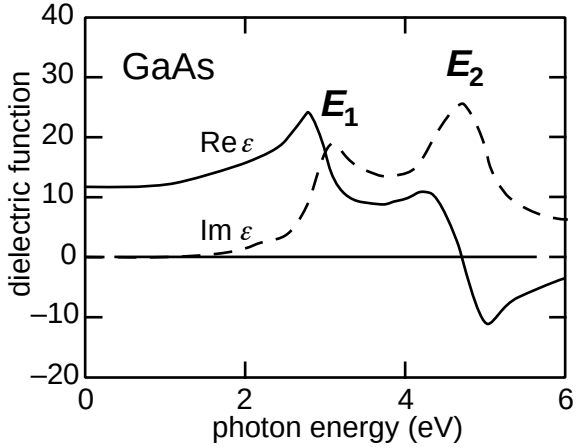
structure



band structure



dielectric function



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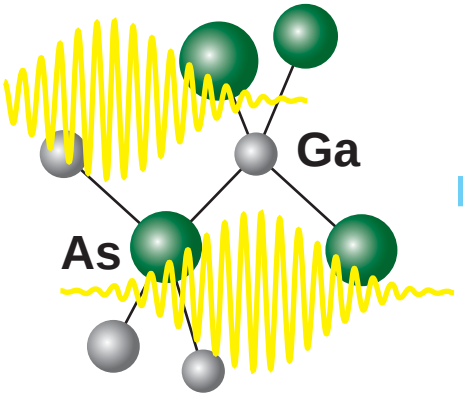
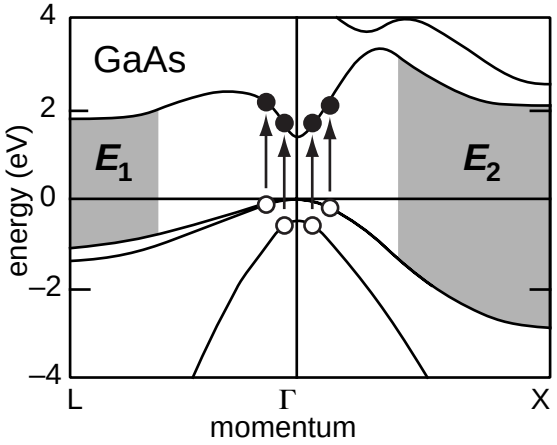
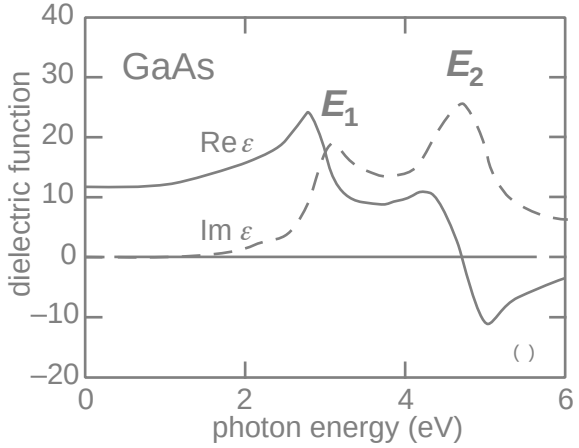
structure



band structure



dielectric function



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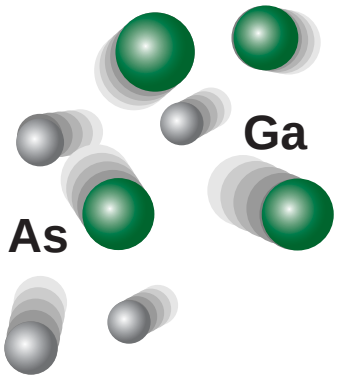
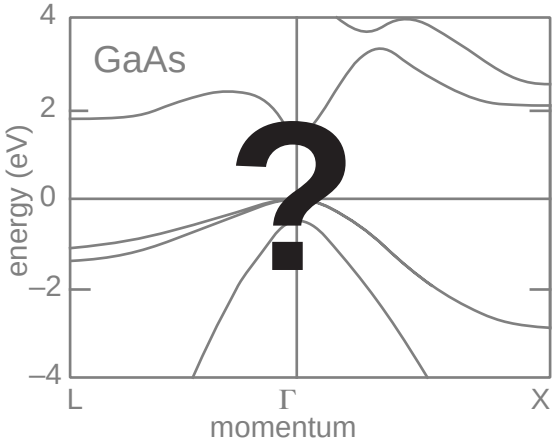
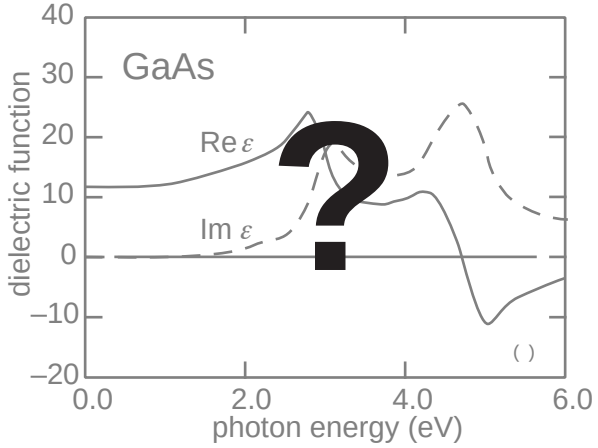
structure



band structure

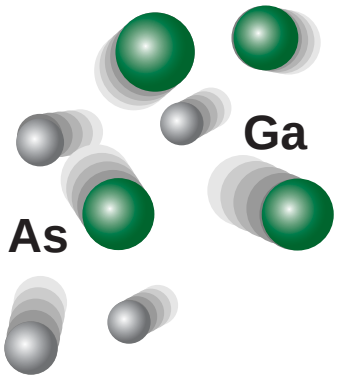


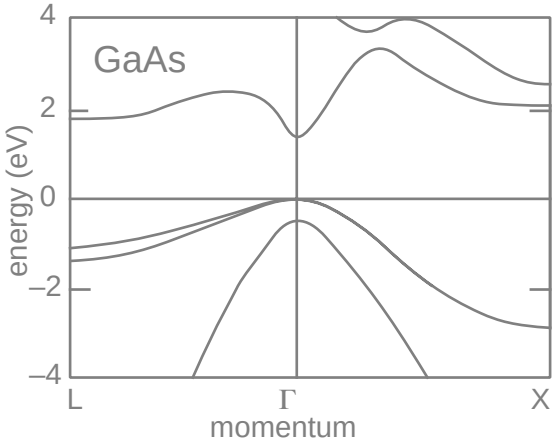
dielectric function

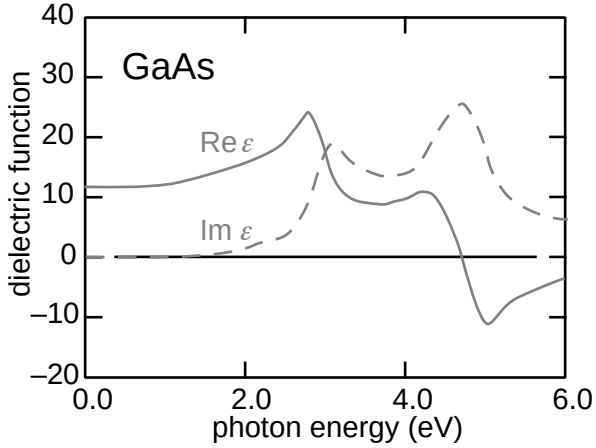


Introduction

structure | **band structure** | **dielectric function**

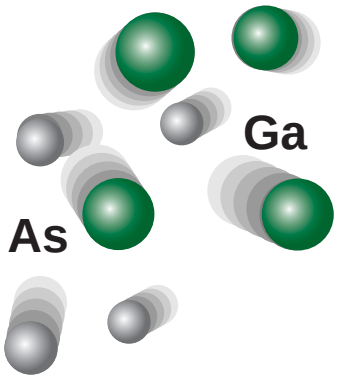






Introduction

structure



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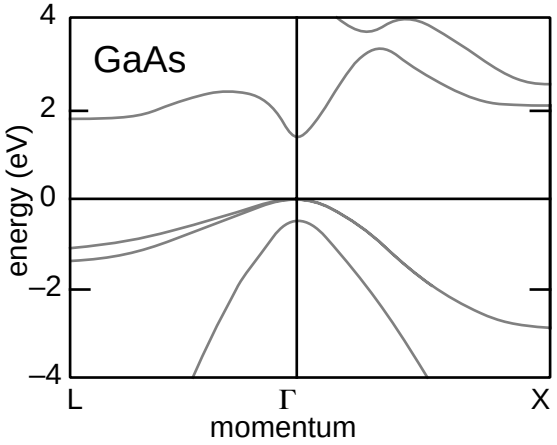

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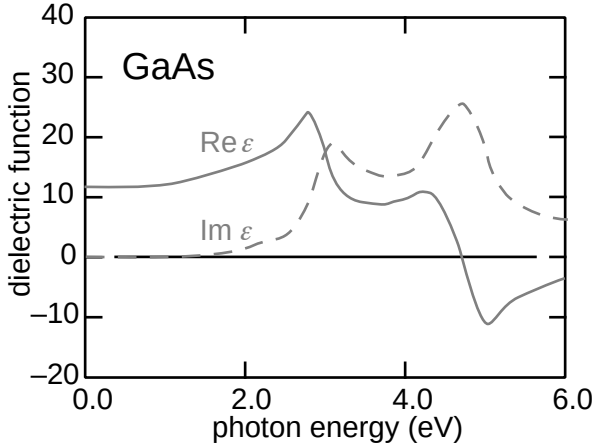

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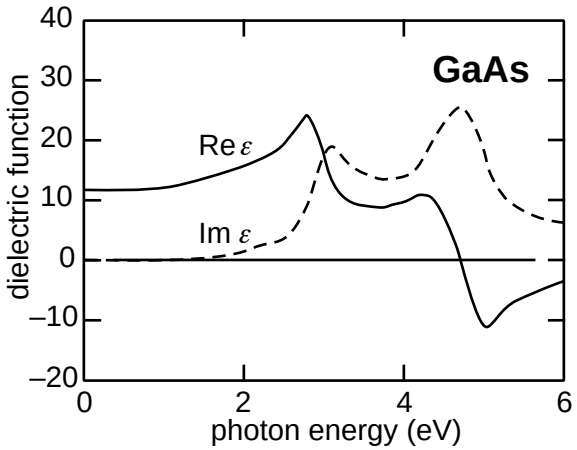

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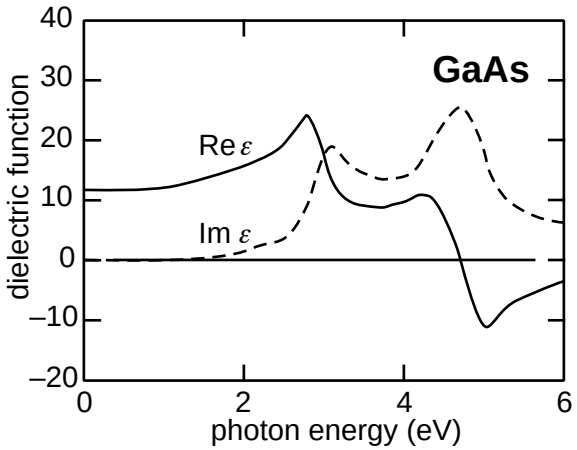

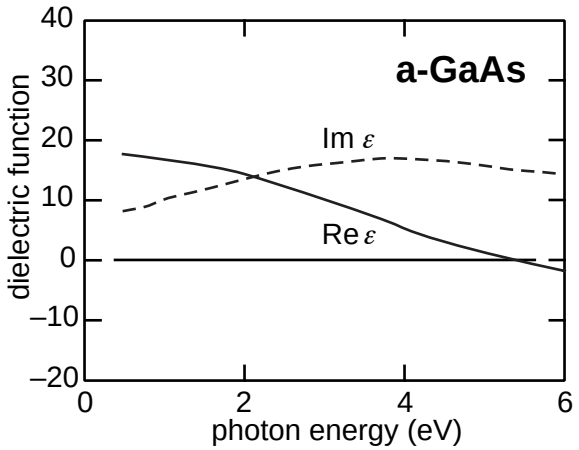

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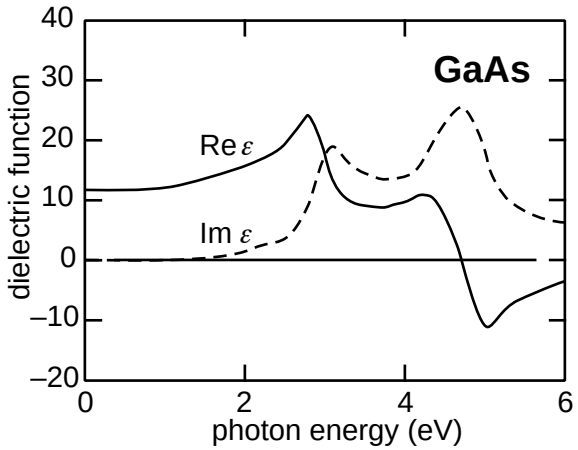

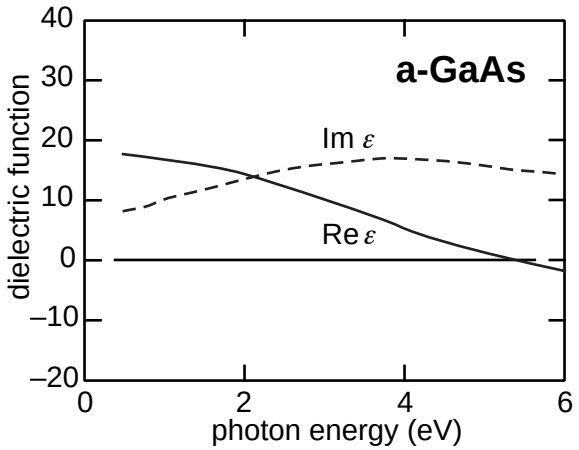

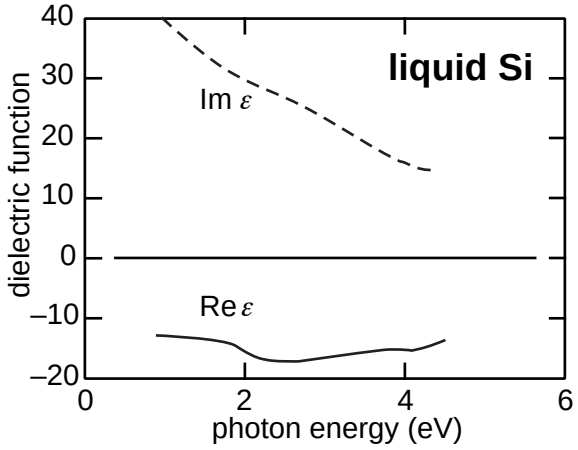

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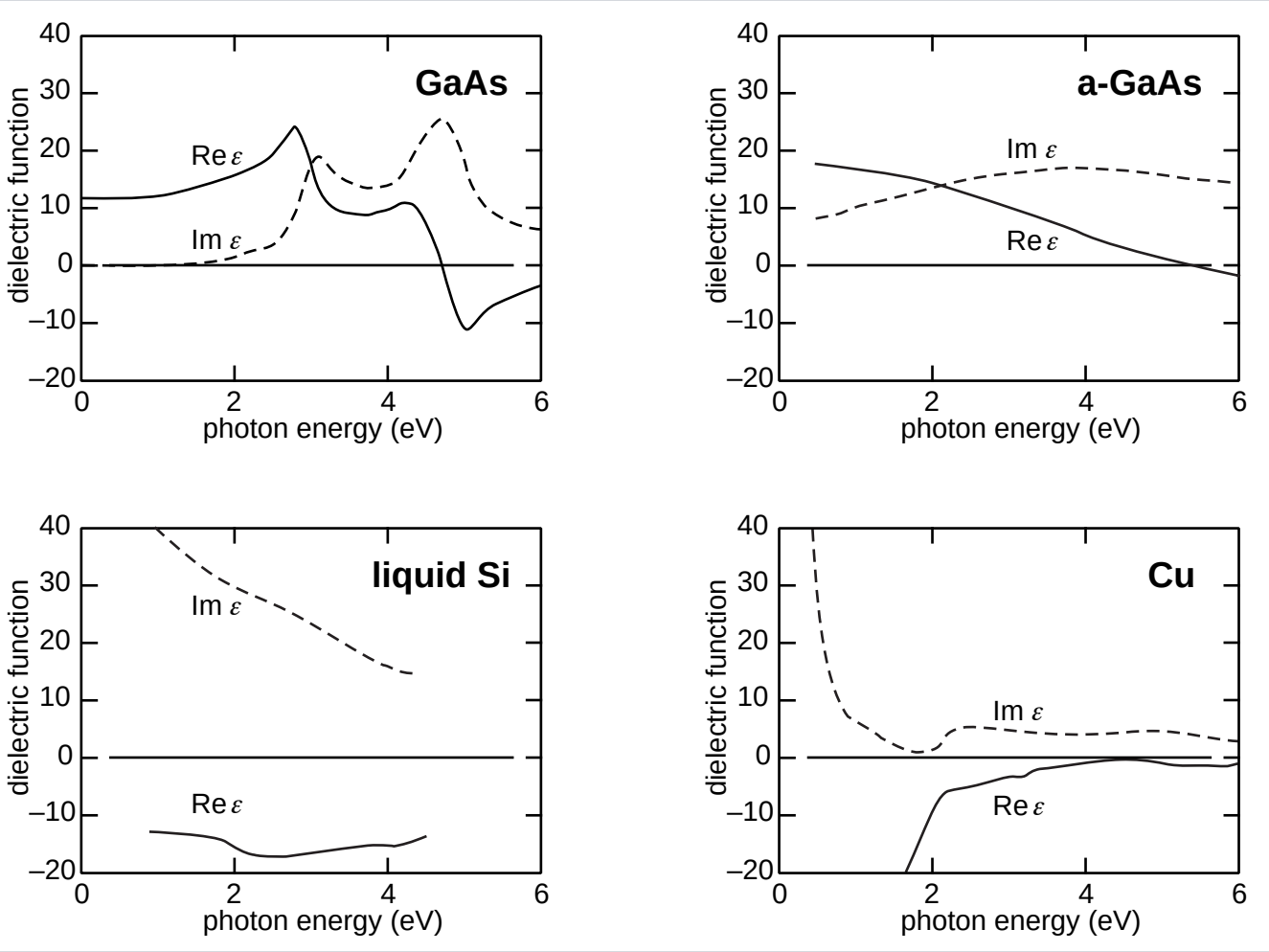


Introduction

- dielectric function: ‘fingerprint’ of state
- light can induce structural transitions

Introduction

- optically induce electronic transitions *without* disordering lattice?
- (coherently) control state of solid?

Introduction

Mechanism for displacive excitation of coherent phonons in Sb, Bi, Te, and Ti_2O_3

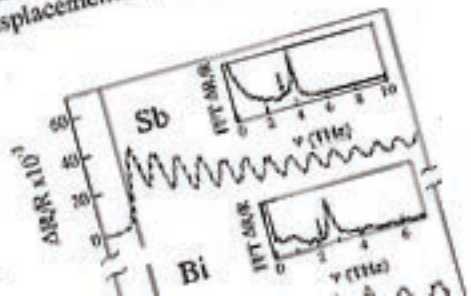
T. K. Cheng, J. Vidal, H. J. Zeiger, G. Dresselhaus, M. S. Dresselhaus, and E. P. Ippen
Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

(Received 1 July 1991; accepted for publication 9 August 1991)

Coherent phonons in Sb, Bi, Te, and Ti_2O_3 can be generated impulsively, and detected in the time domain through reflectivity modulation using 60 fs pulses of laser light at 2 eV. Experimental data for these opaque solids suggest that a direct Raman excitation mechanism is not responsible for coherent phonon generation. Rather, the excitation is attributed to an electronically induced displacement of the ion equilibrium coordinates.

In recent years, there have been numerous reports of coherent molecular[1-4] and lattice[4-9] vibrations in time-resolved optical pump-probe measurements. In this letter, we propose a model to explain our observations of the excitation of very large coherent phonon amplitudes with only A_1 symmetry in opaque single-crystal samples of Sb, Bi, Te, and Ti_2O_3. The experimental data for these opaque solids suggest that the coherent phonon excitation involves an electronic resonance but is not directly driven by an impulsive stimulated Raman scattering mechanism (ISRS),[2-5] as is often the case in the experiments for large [illegible]. Our conclusion follows from observation of the coherent phonon [illegible] selectivity and careful measurement of the coherent phonon [illegible] in each material. [illegible] excitation and detection of coherent phonons were [illegible] the output of a dispersion-compensated [illegible] laser[10] (producing 60 fs, [illegible] rate of 100 MHz [illegible] pump-

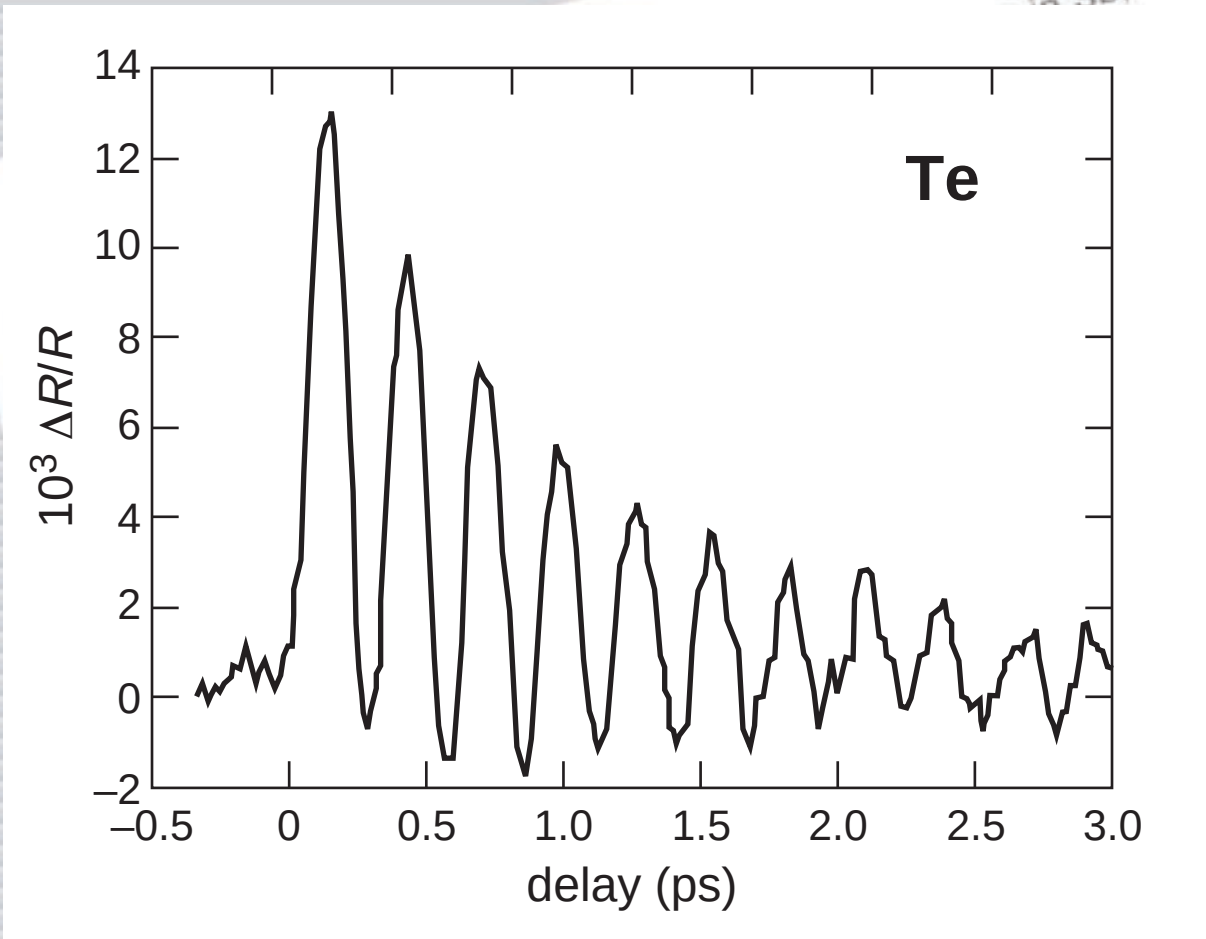
2 to 1 ratio for A_{1g} to E_g mode intensities in Sb and Bi). Rather, we believe the coherent phonon generation is due to a displacement of A_1 symmetry of the ion quasi-equilibrium coordinates produced by electronic excitation, which we call displacive excitation of coherent phonons (DECP). The DECP mechanism is closely related to the displaced ion equilibrium model for molecular systems. The ions cannot respond on the time scale of the electronic quasi-equilibration time and are therefore set into oscillation in an A_1 mode of vibration about the displaced quasi-equilibrium A_1 ion coordinate Q_0. Any other quasi-equilibrium displacements (displacements of E_g symmetry, for exam-



Cheng *et al.*, *Appl. Phys. Lett.* 59, 1923 (1991)

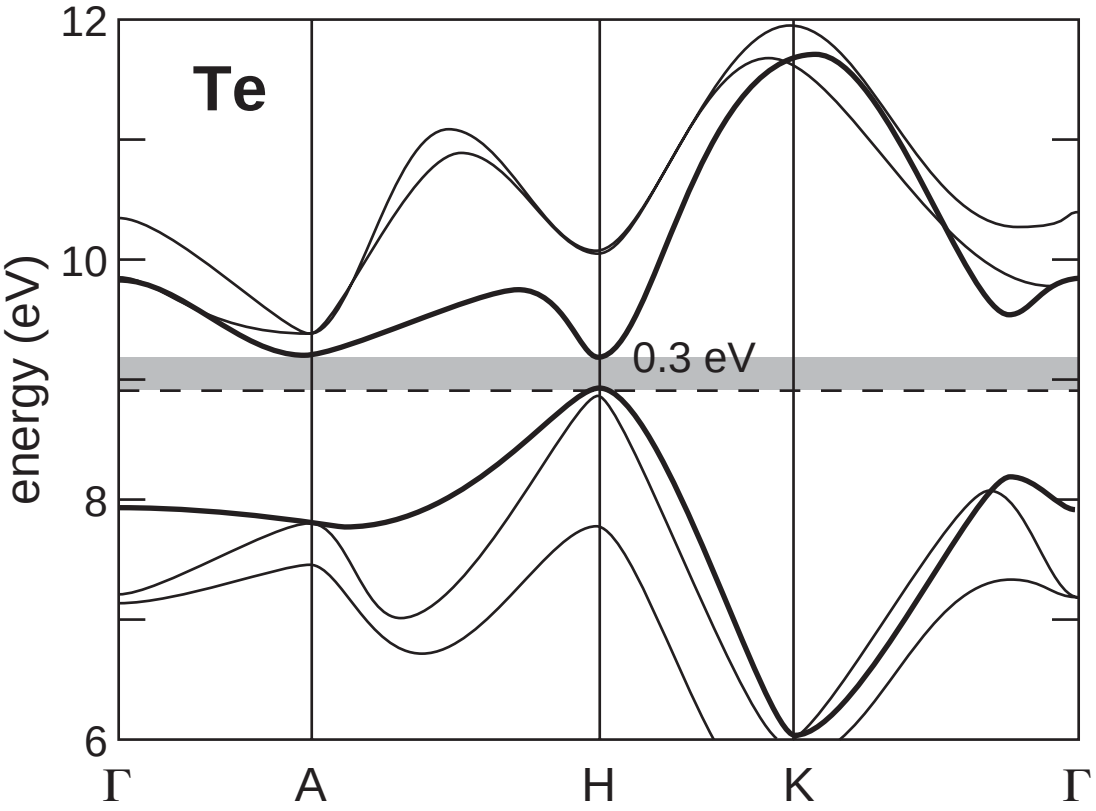
Introduction

optically induce large amplitude phonons



Cheng *et al.*, *Appl. Phys. Lett.* 59, 1923 (1991)

Tellurium: close to transition (0.3 eV gap)



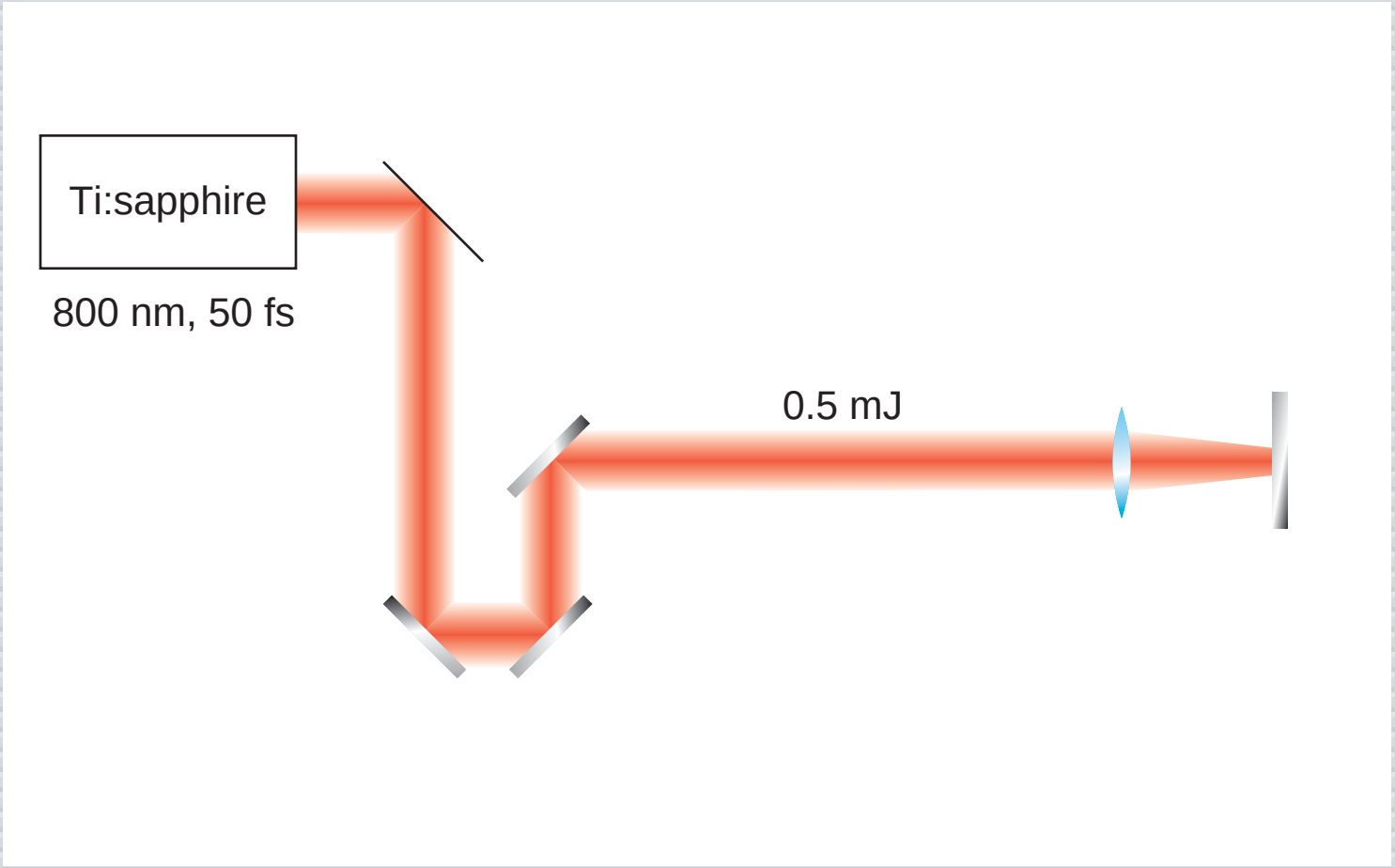
P. Tangney (Princeton) and S. Fahey (Cork), *private communication*

Outline

- technique
- results
- discussion

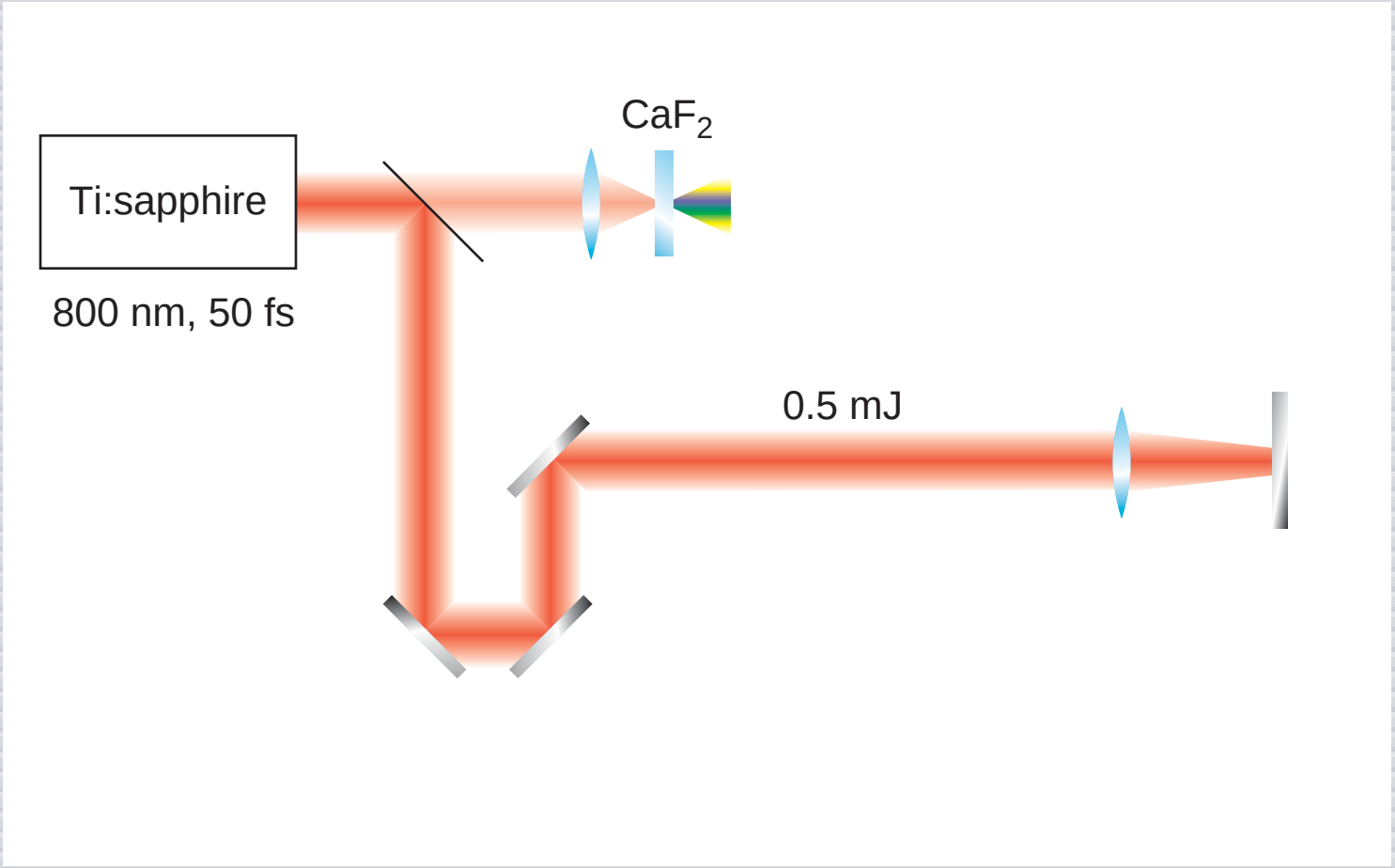
Technique

broadband time-resolved ellipsometry

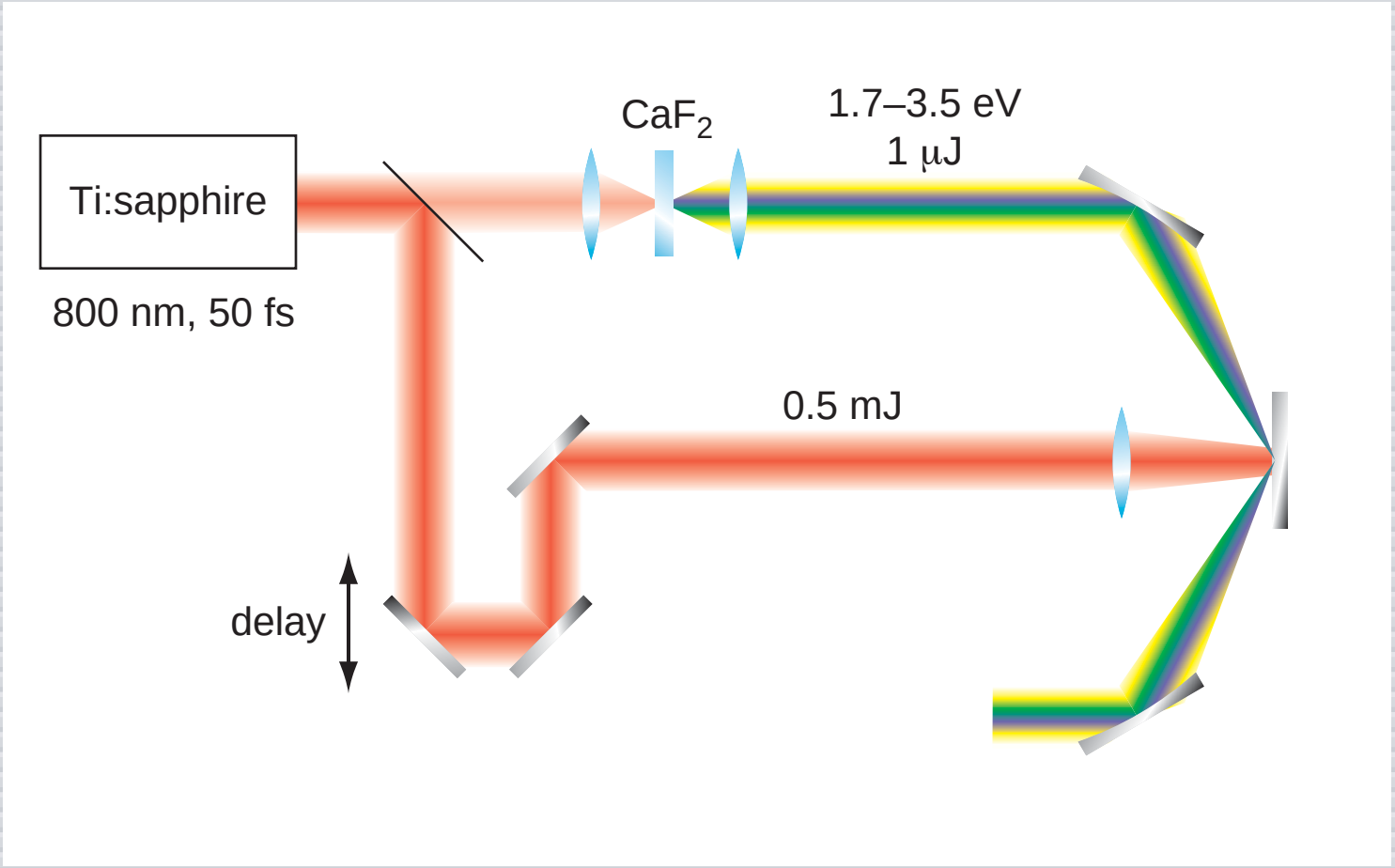


Technique

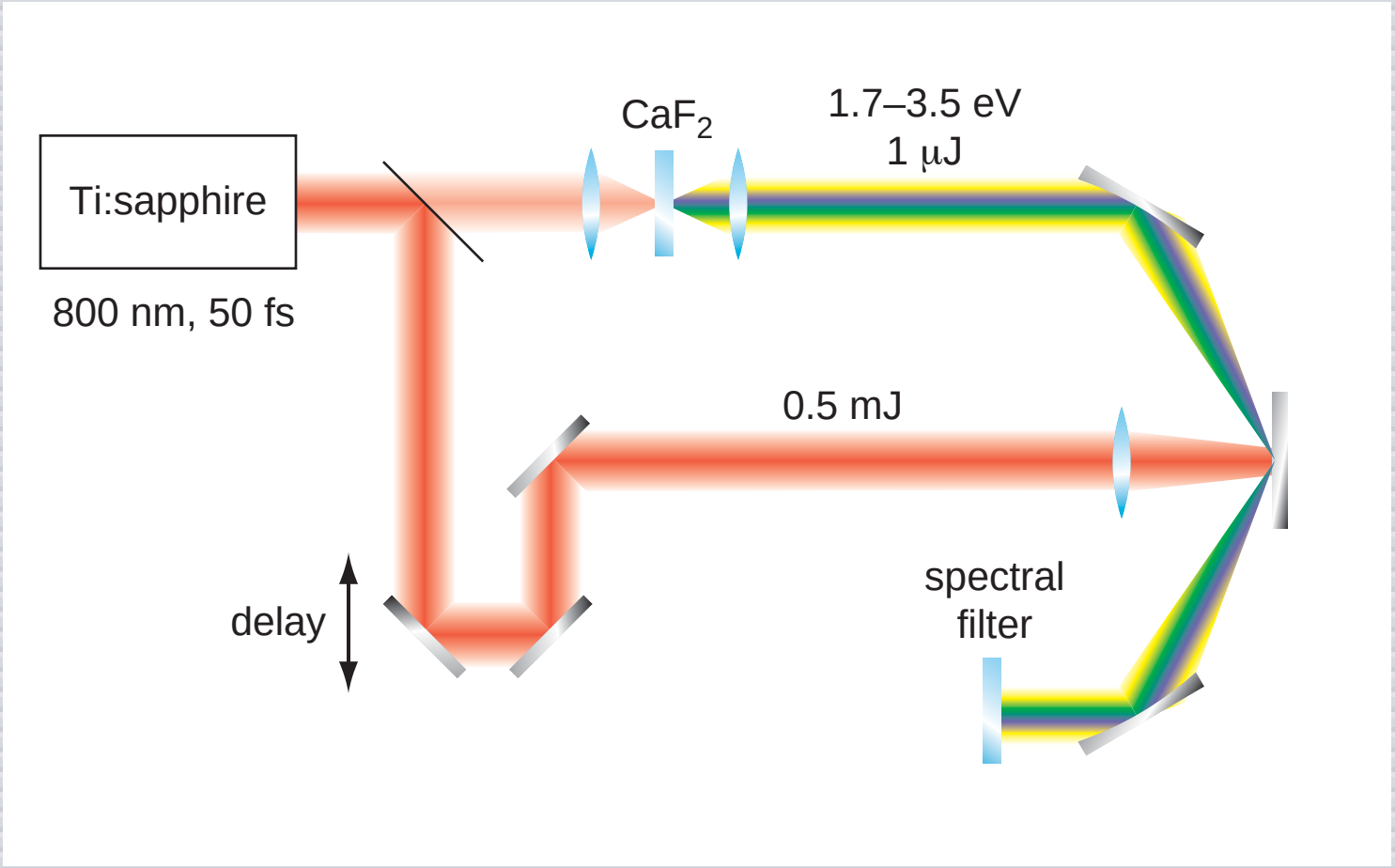
broadband time-resolved ellipsometry



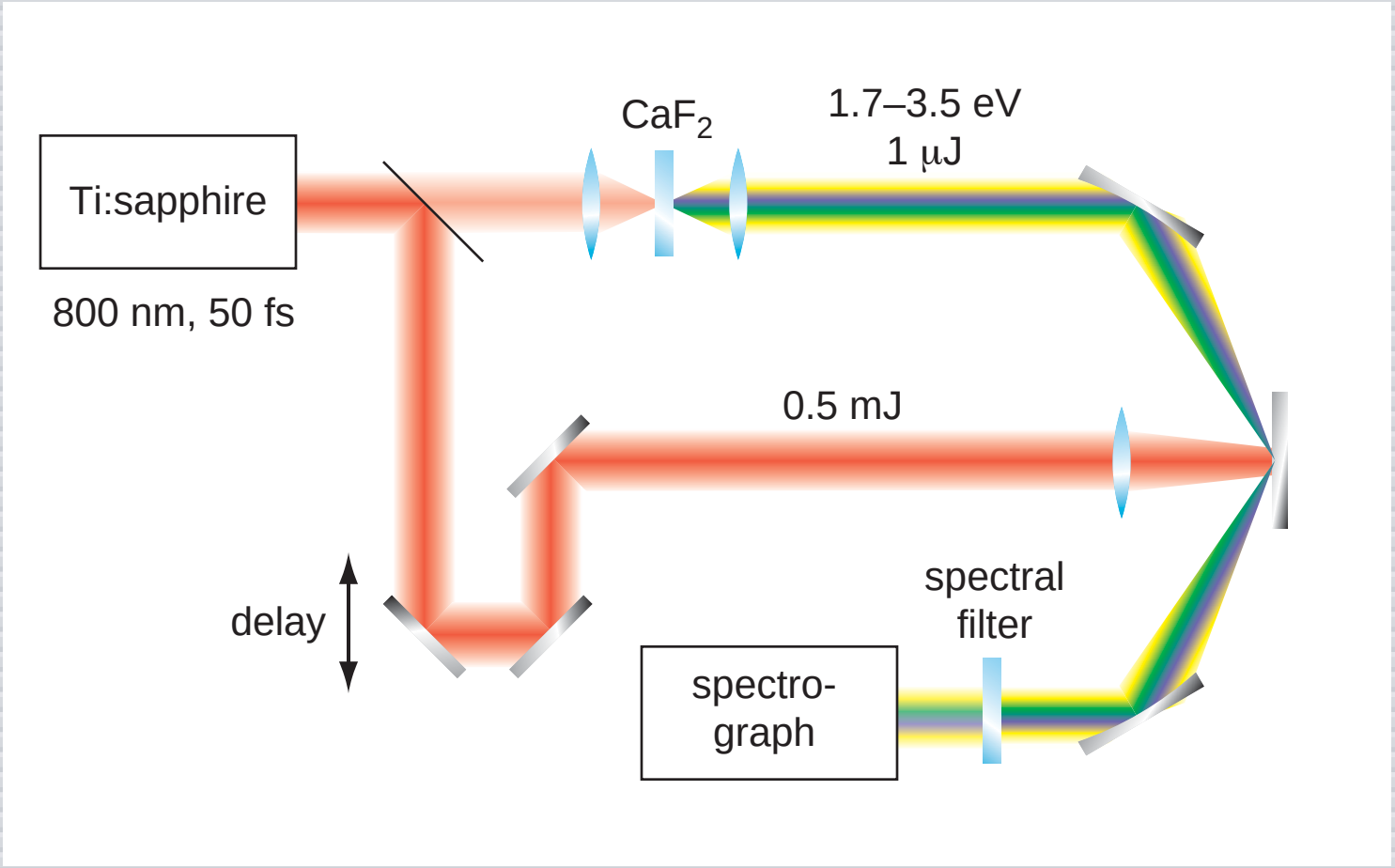
broadband time-resolved ellipsometry



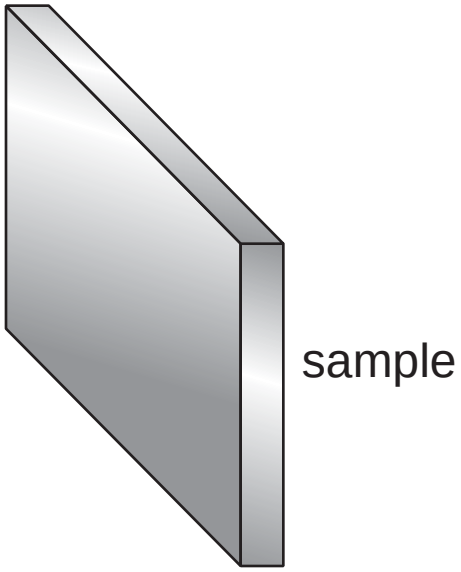
broadband time-resolved ellipsometry



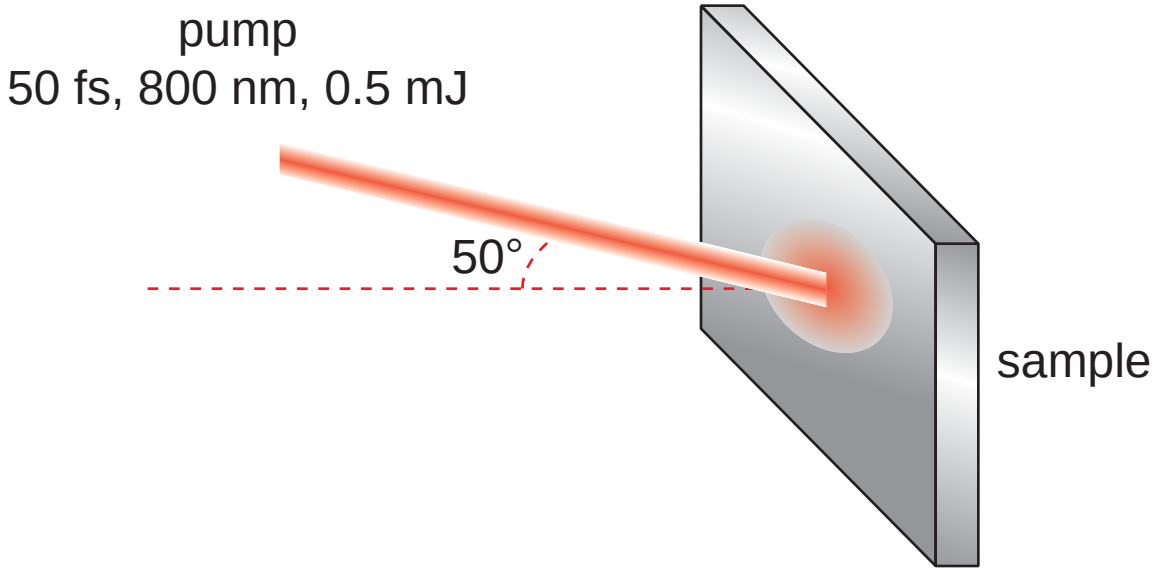
broadband time-resolved ellipsometry



broadband time-resolved ellipsometry

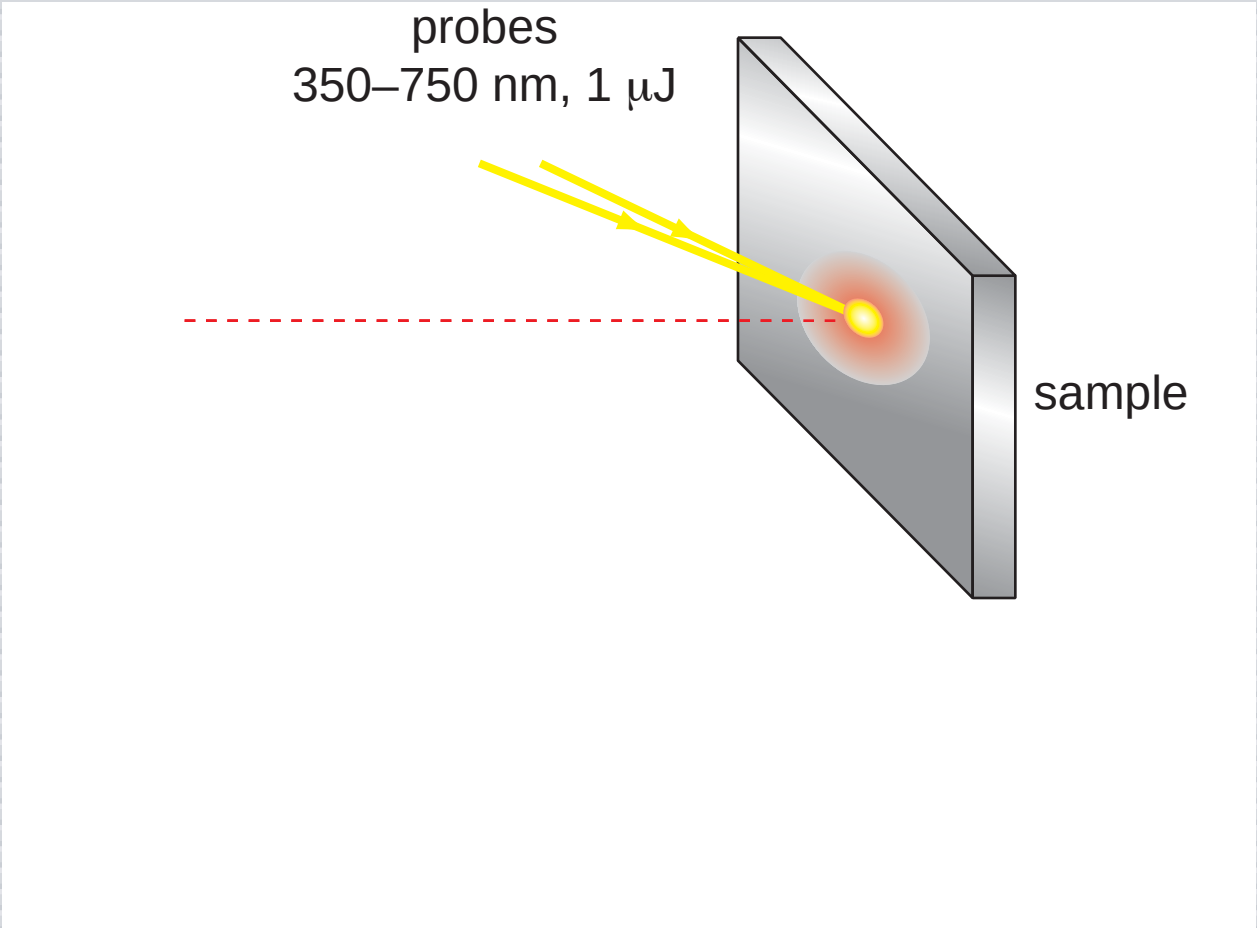


broadband time-resolved ellipsometry



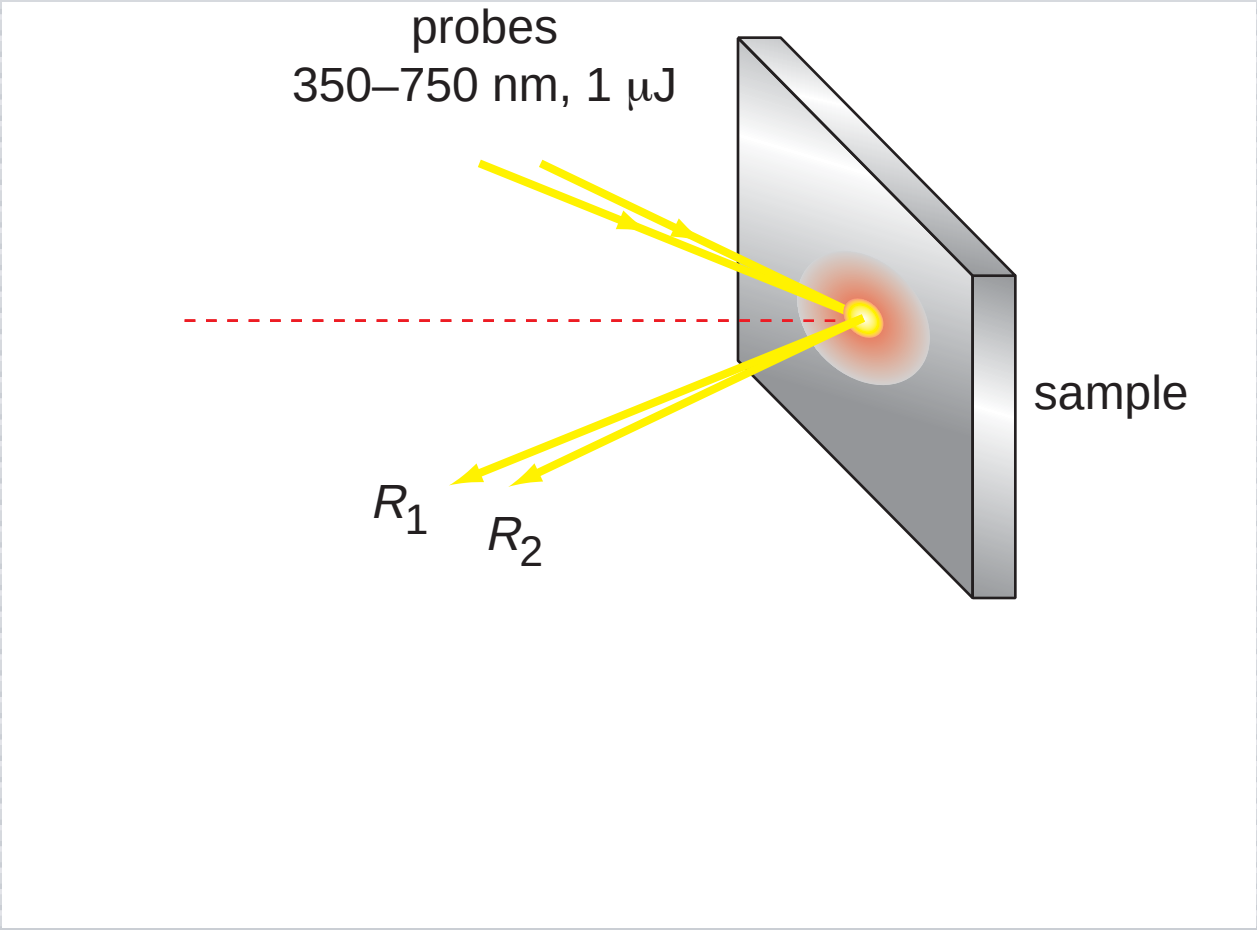
Technique

broadband time-resolved ellipsometry



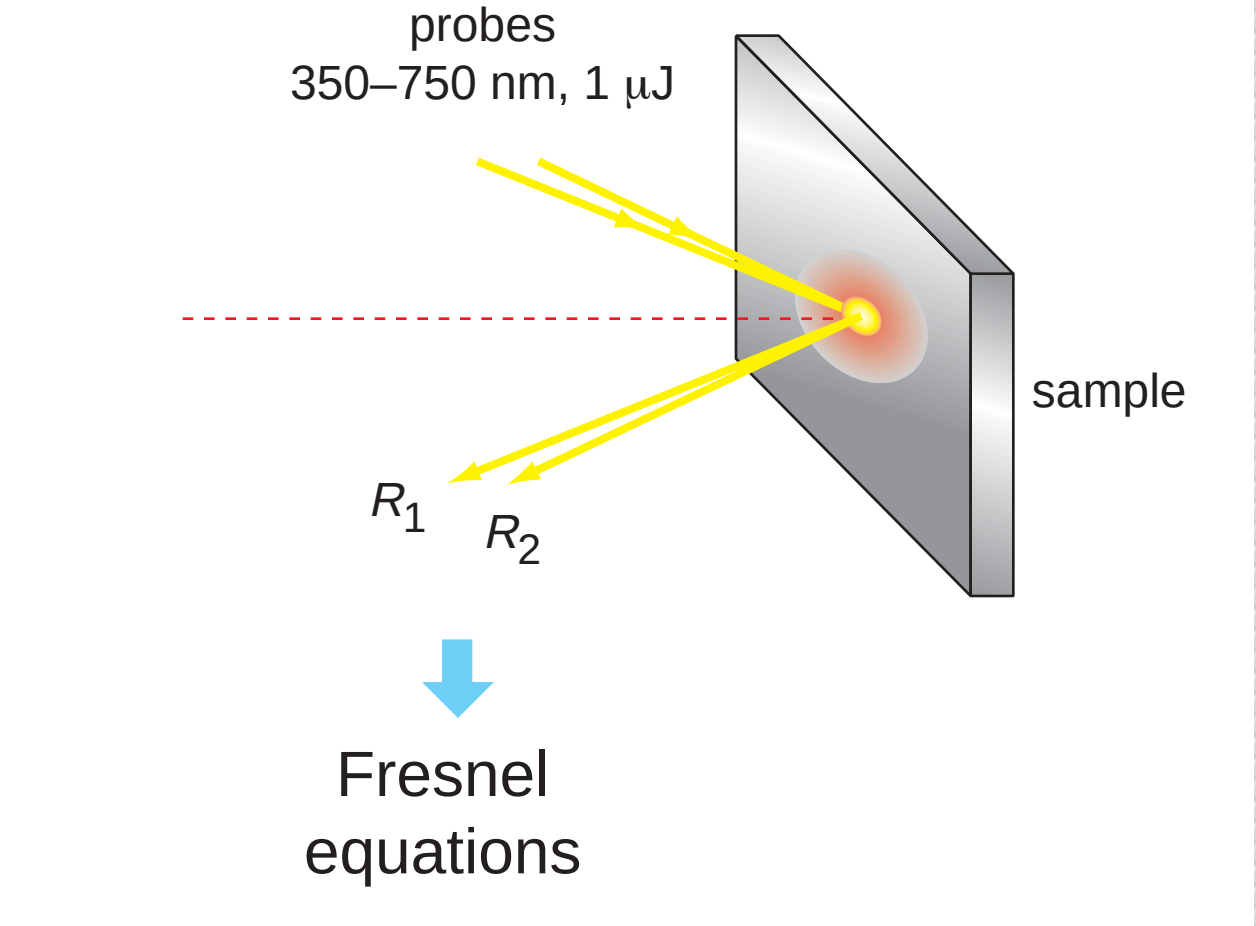
Technique

broadband time-resolved ellipsometry



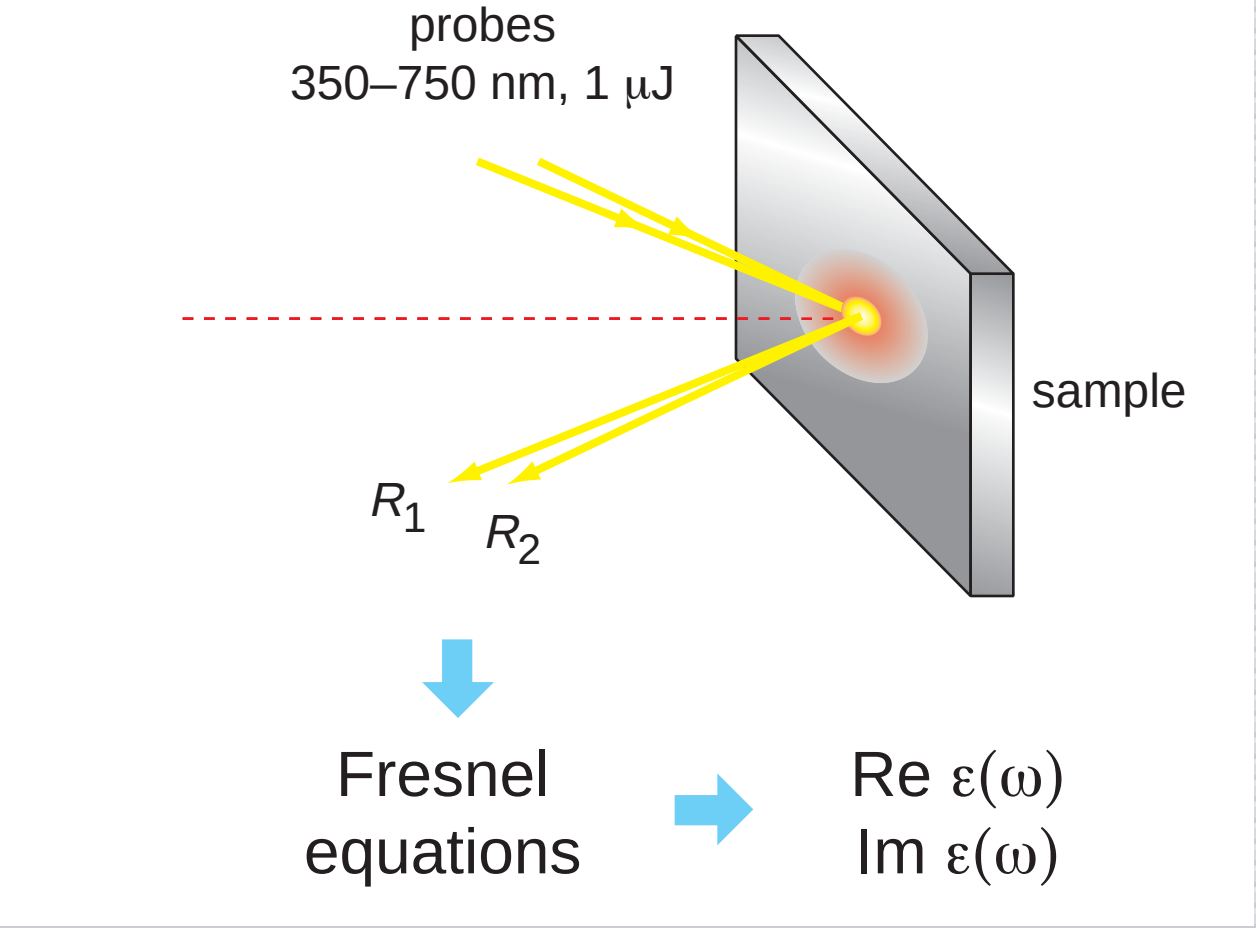
Technique

broadband time-resolved ellipsometry

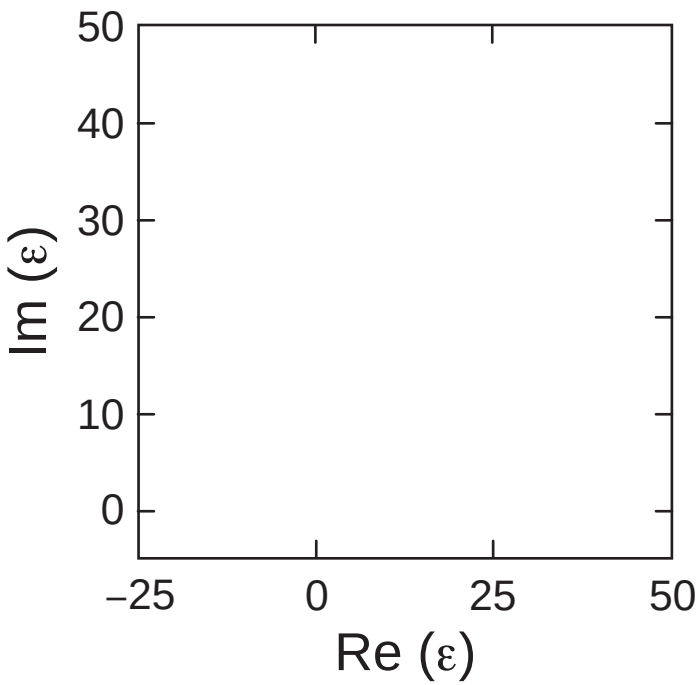


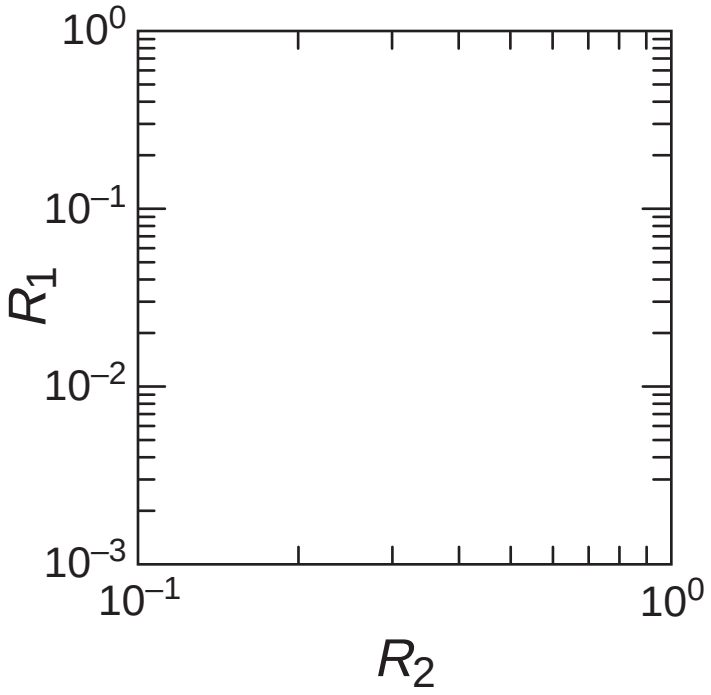
Technique

broadband time-resolved ellipsometry



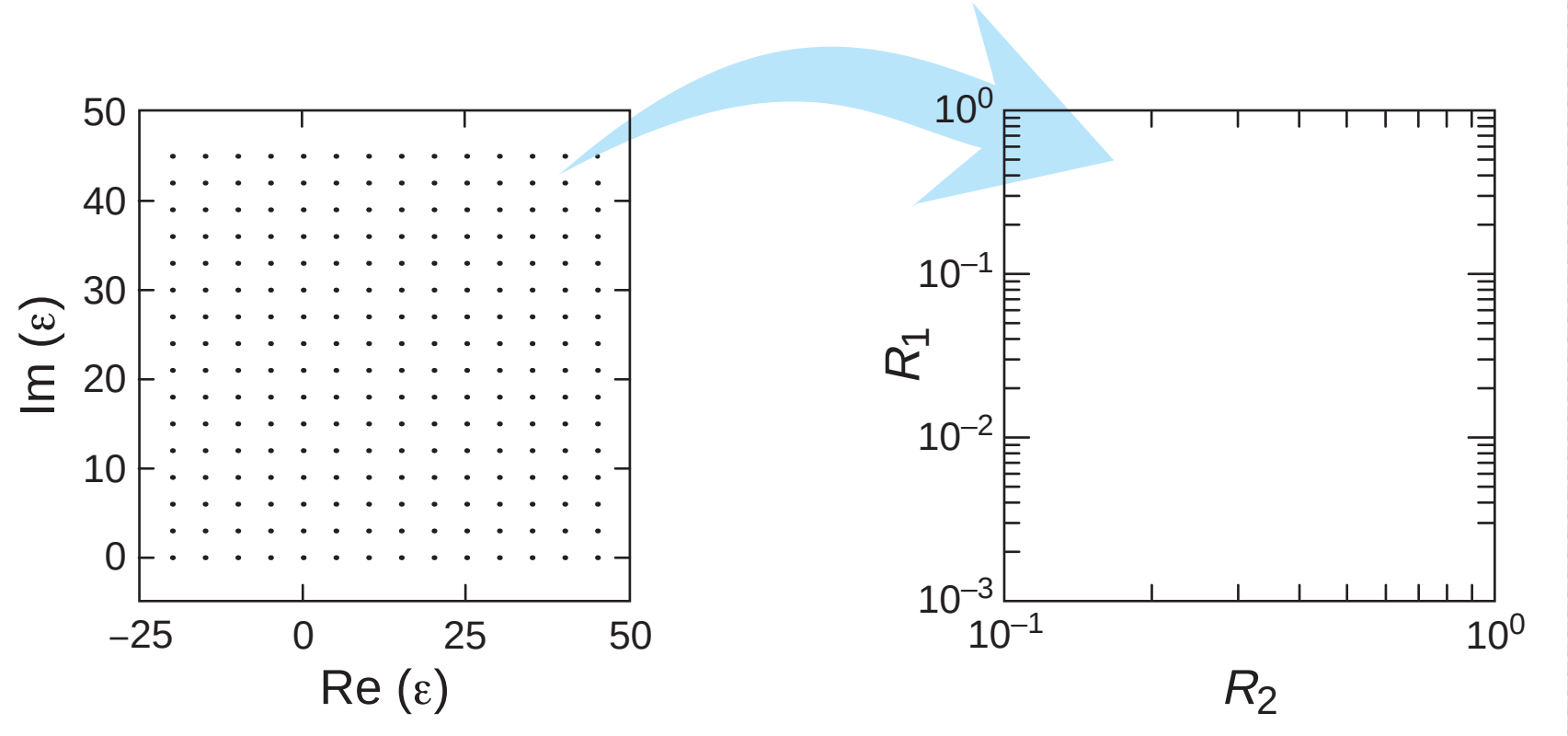
choice of angles





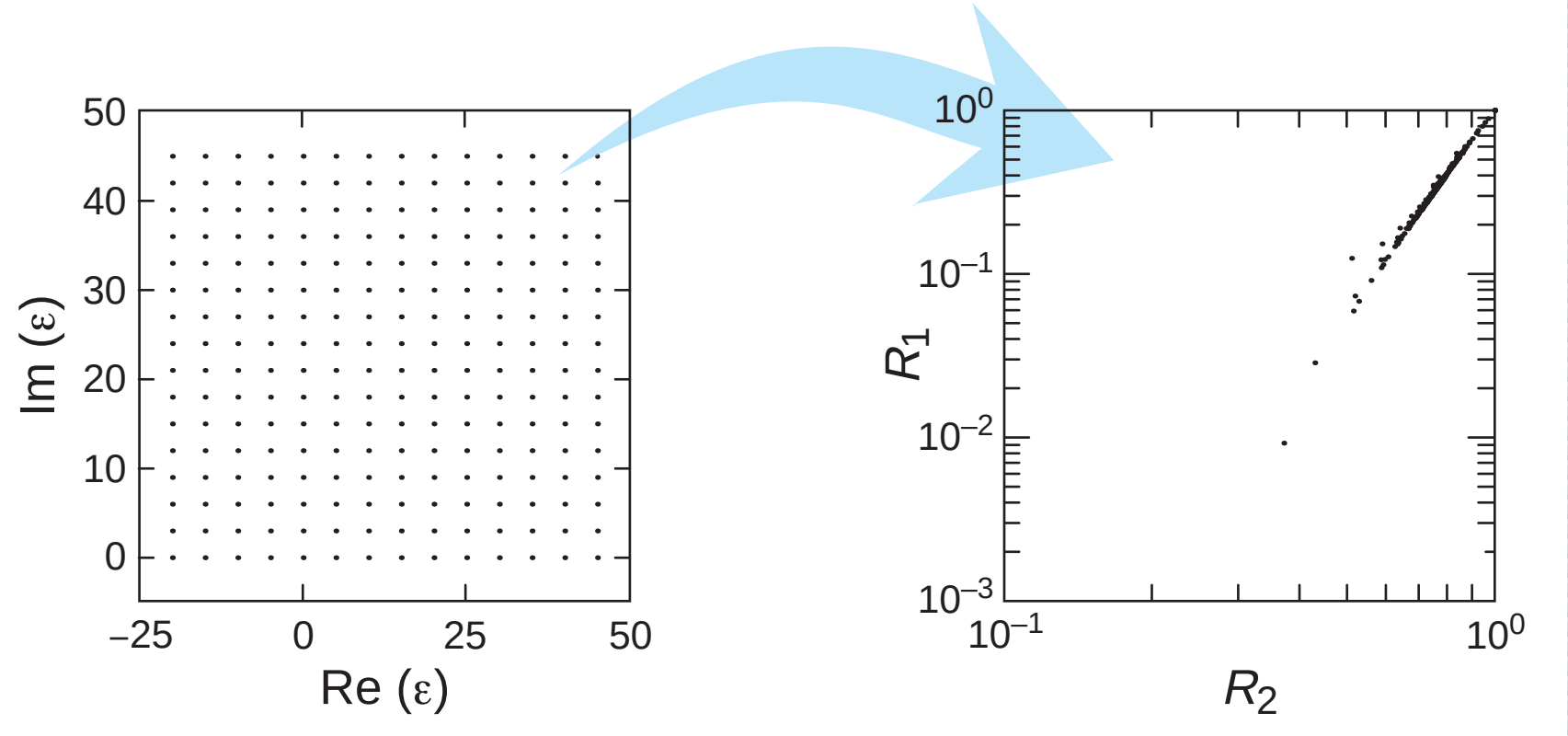
Fresnel equations cannot be inverted analytically

choice of angles



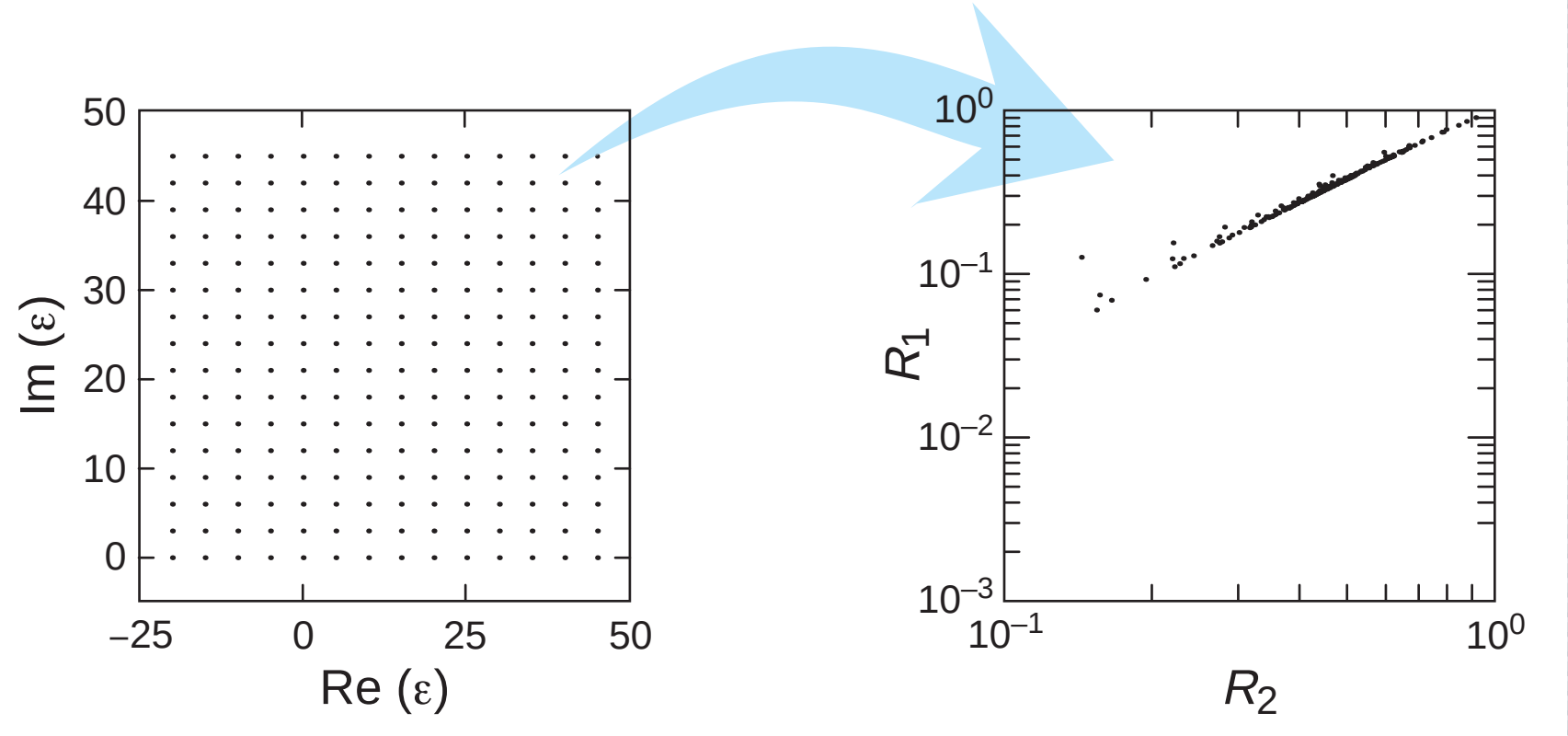
need numerical inversion

choice of angles



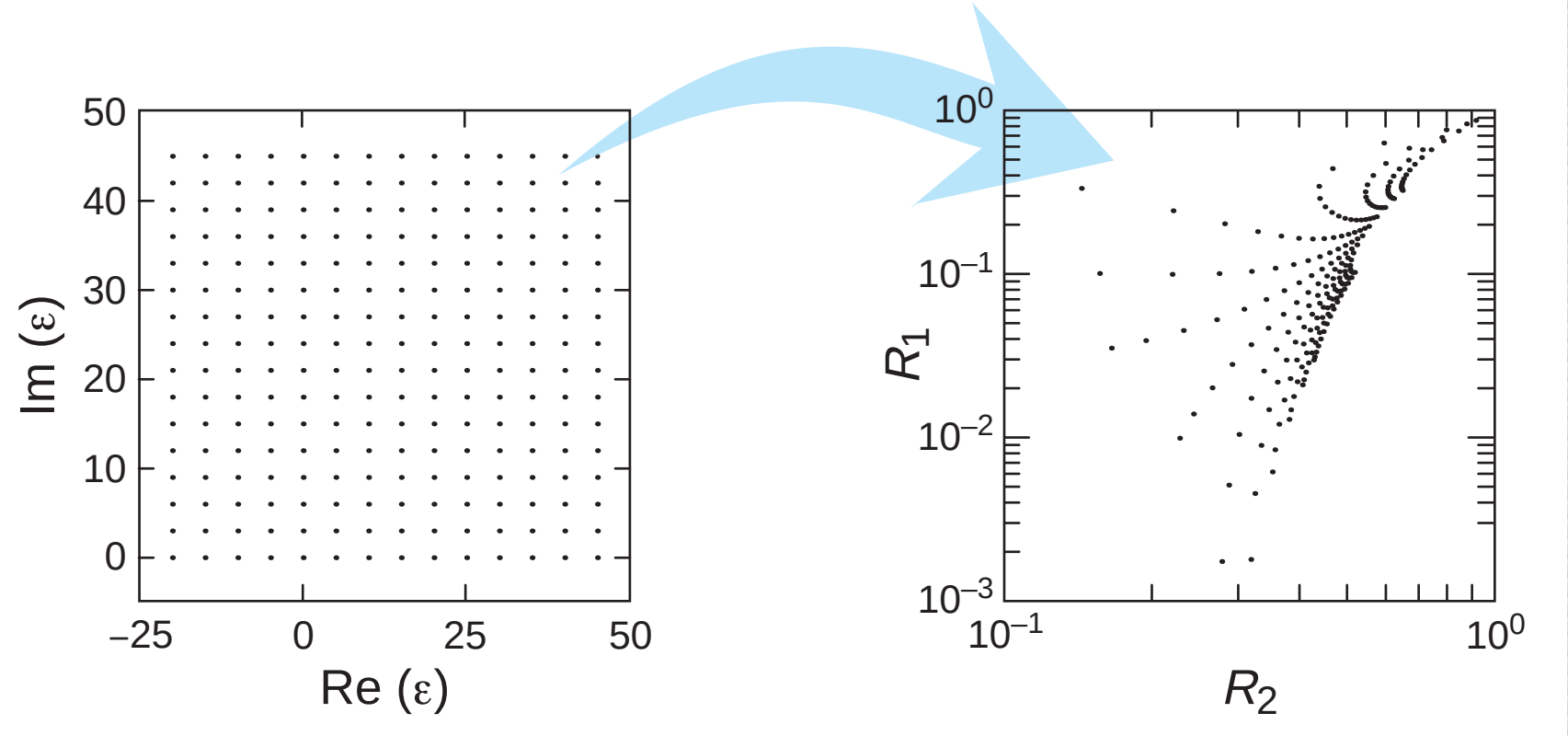
R_1 = 45° *p*-pol, R_2 = 45° *s*-pol

choice of angles



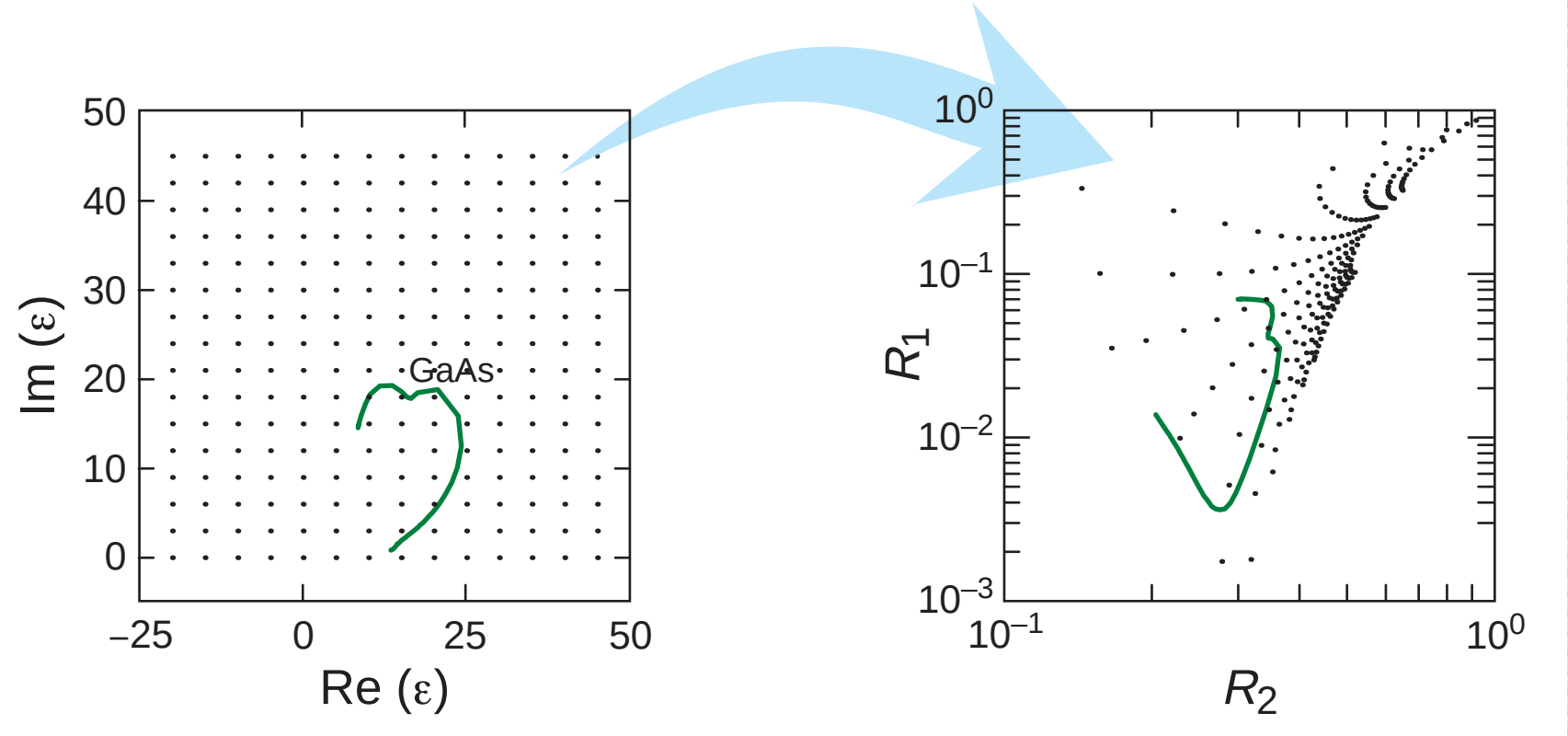
R_1 = 60° *p*-pol, R_2 = 45° *p*-pol

choice of angles



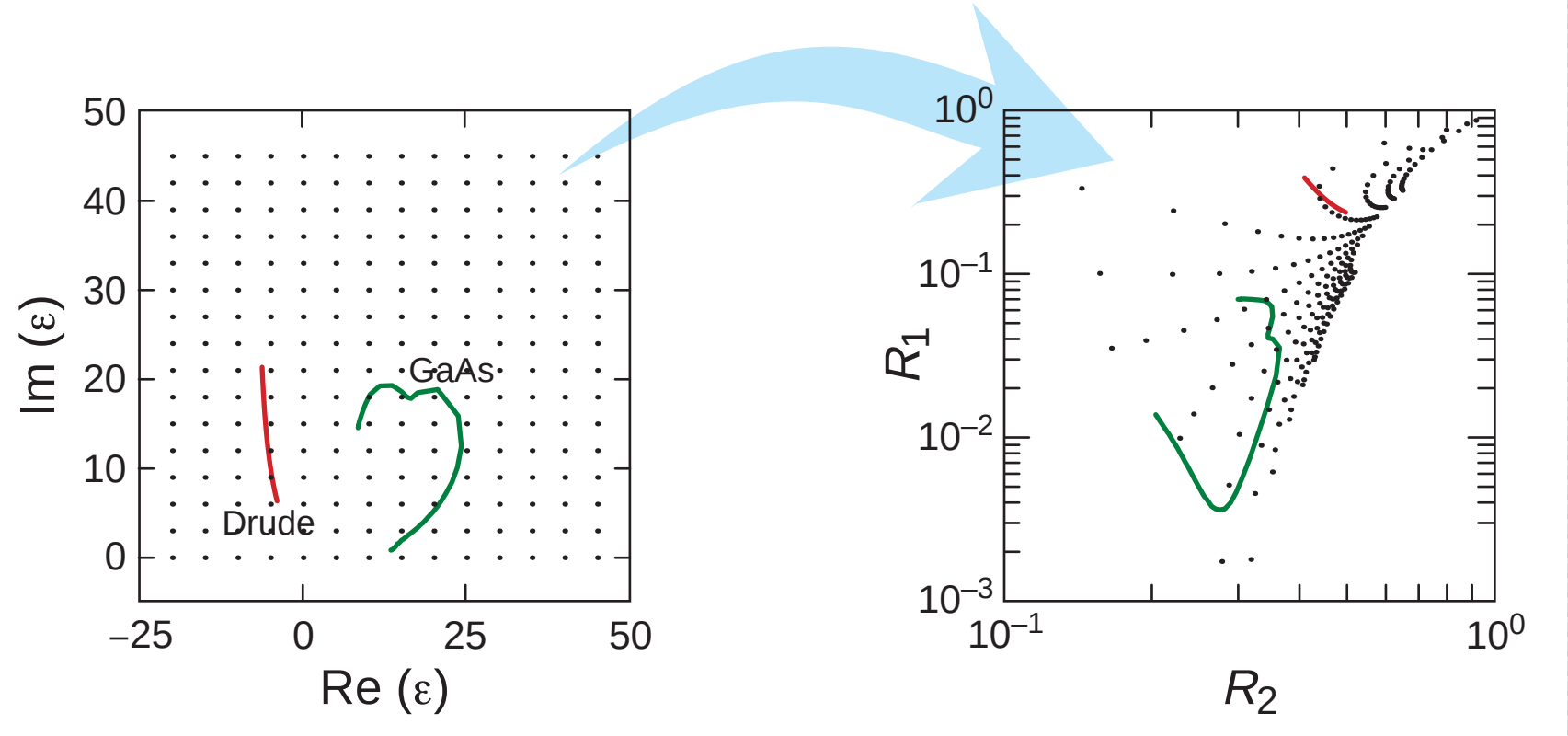
R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

choice of angles



R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

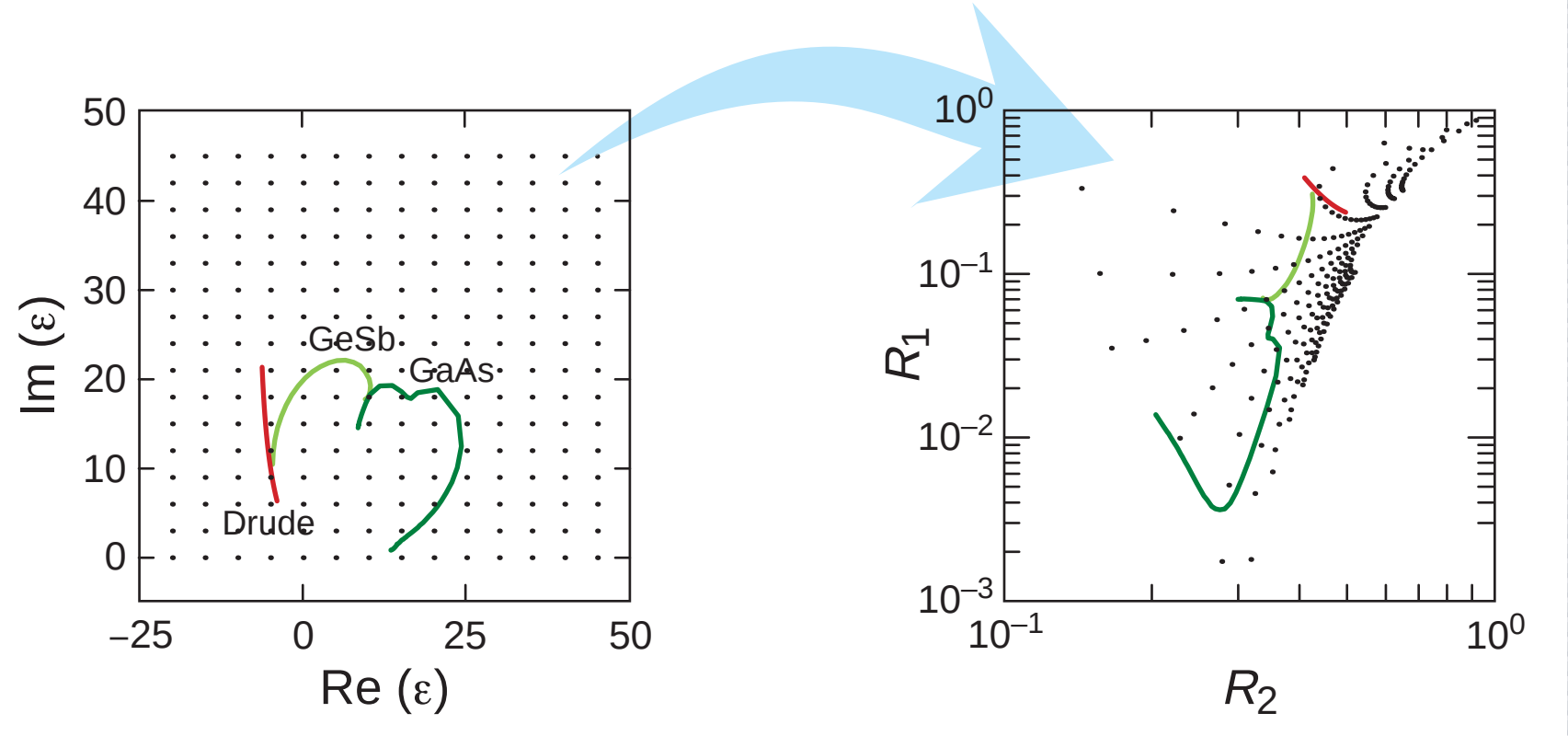
choice of angles



R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

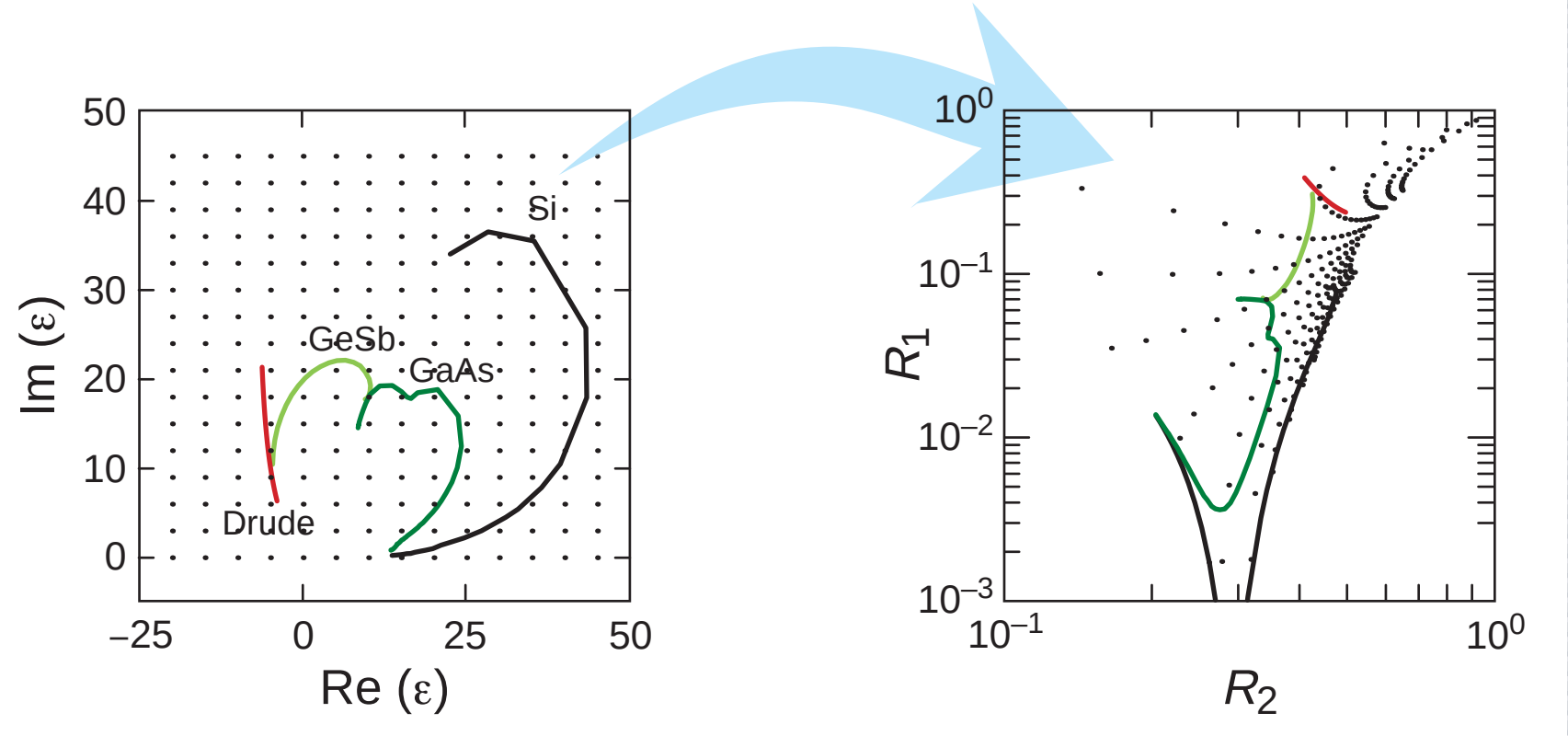
Technique

choice of angles



R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

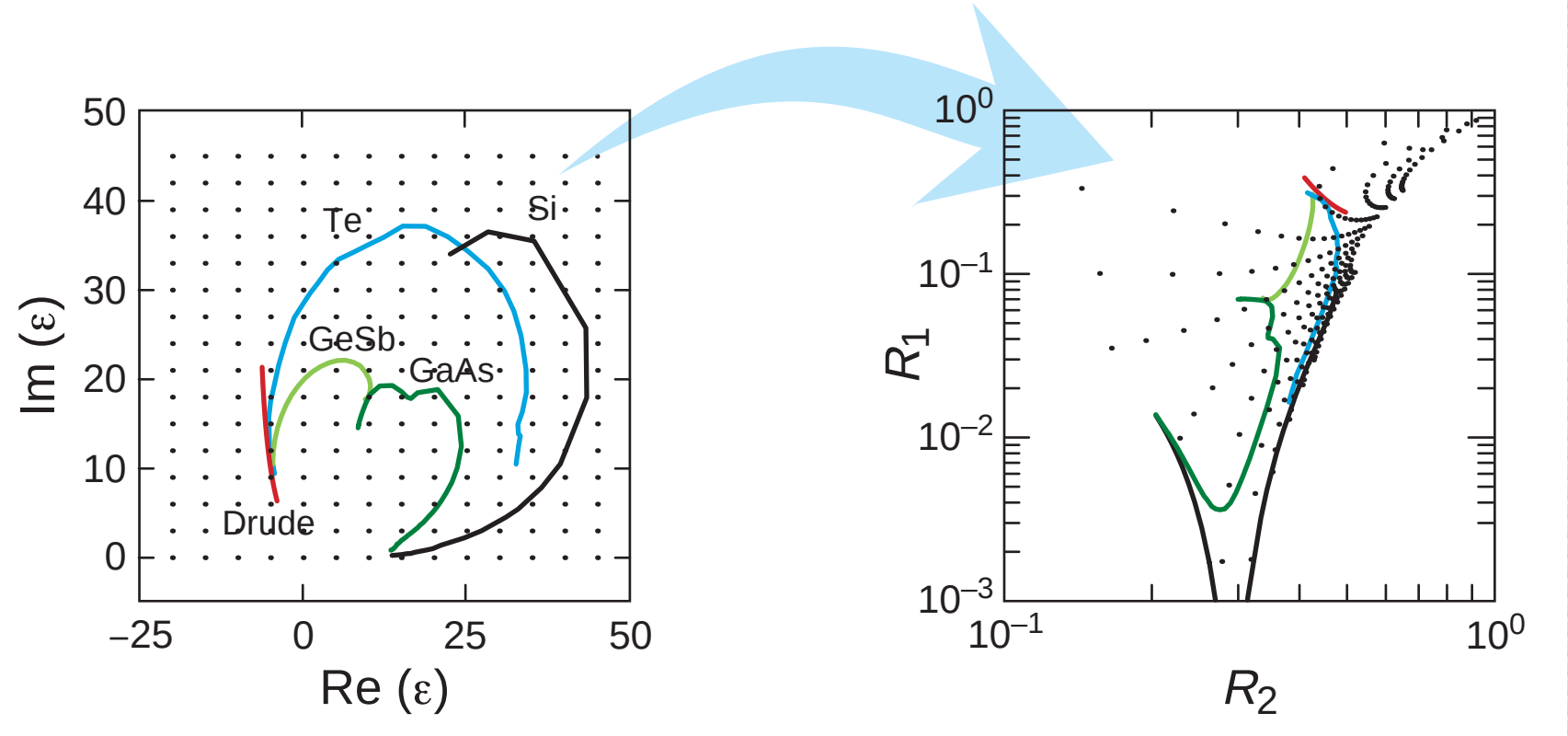
choice of angles



R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

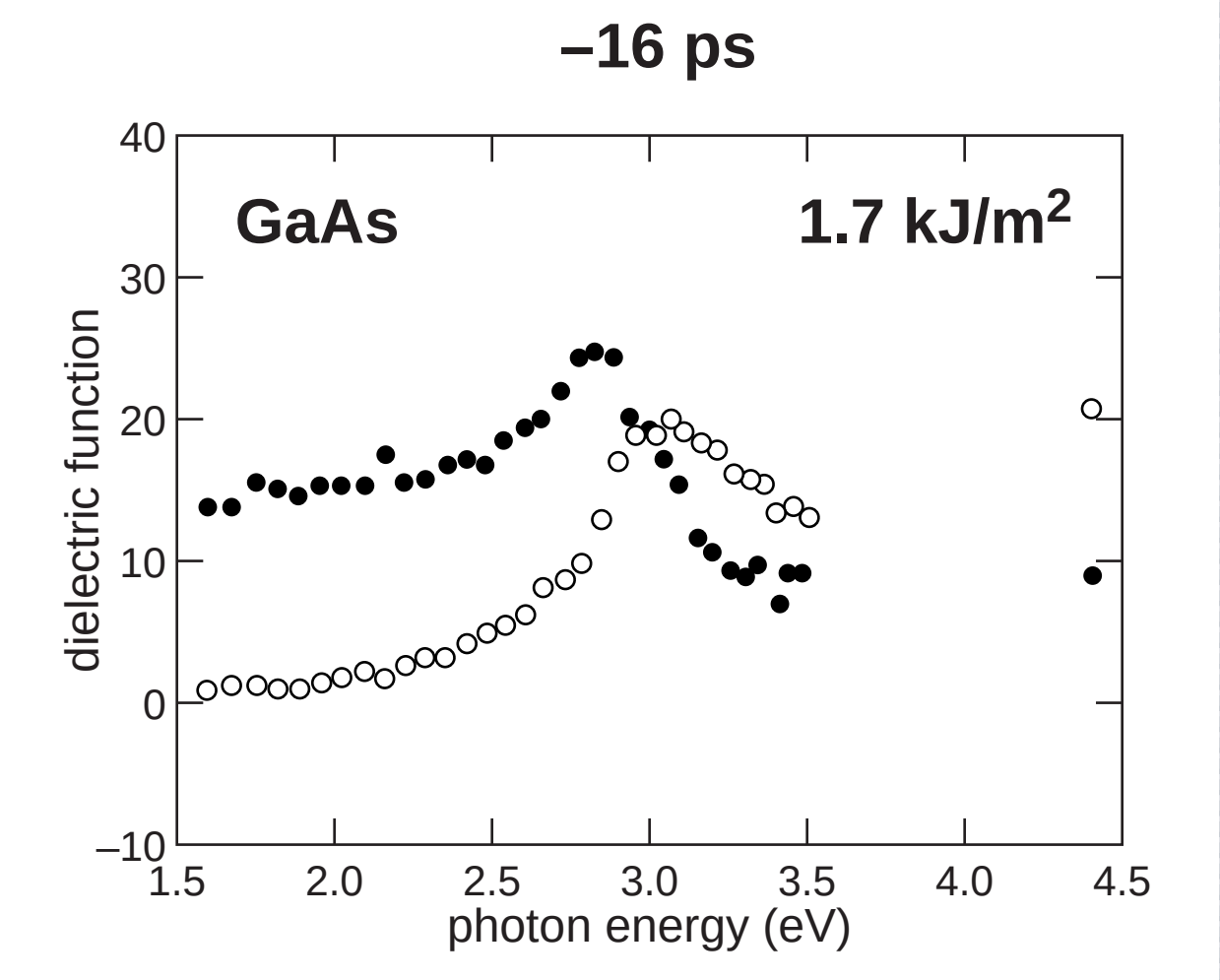
Technique

choice of angles



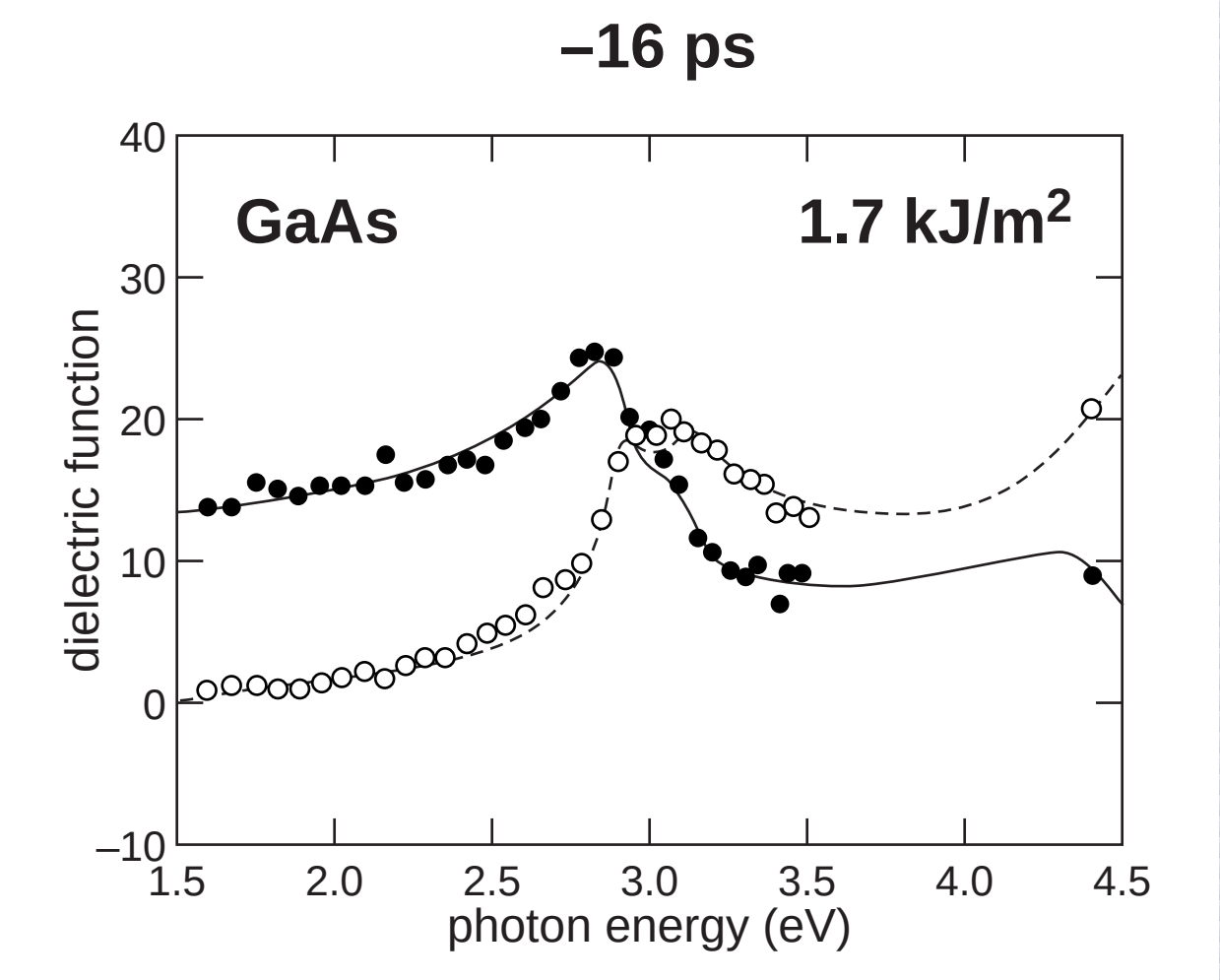
R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

Technique



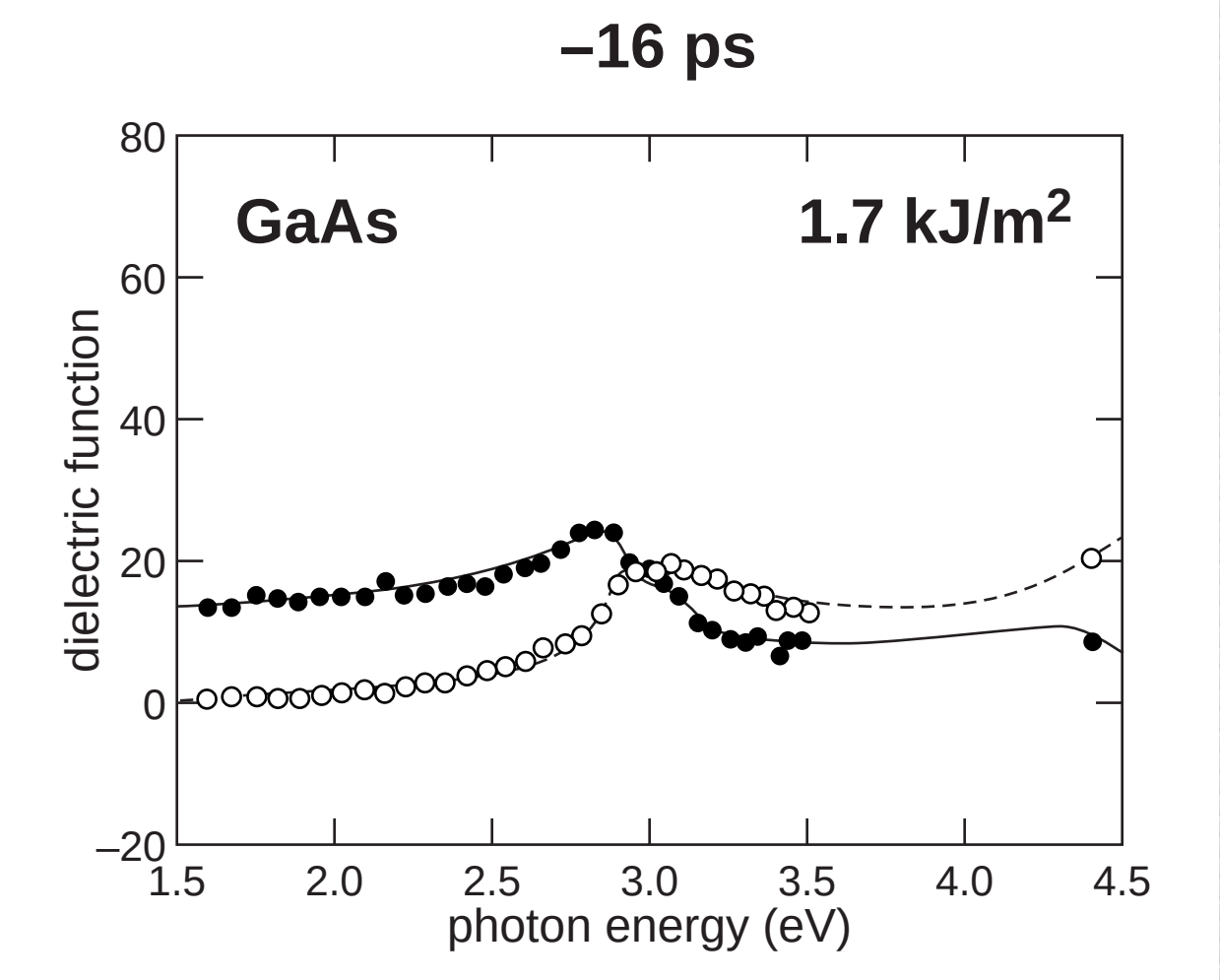
Phys. Rev. Lett. **80, 185 (1998)**

Technique



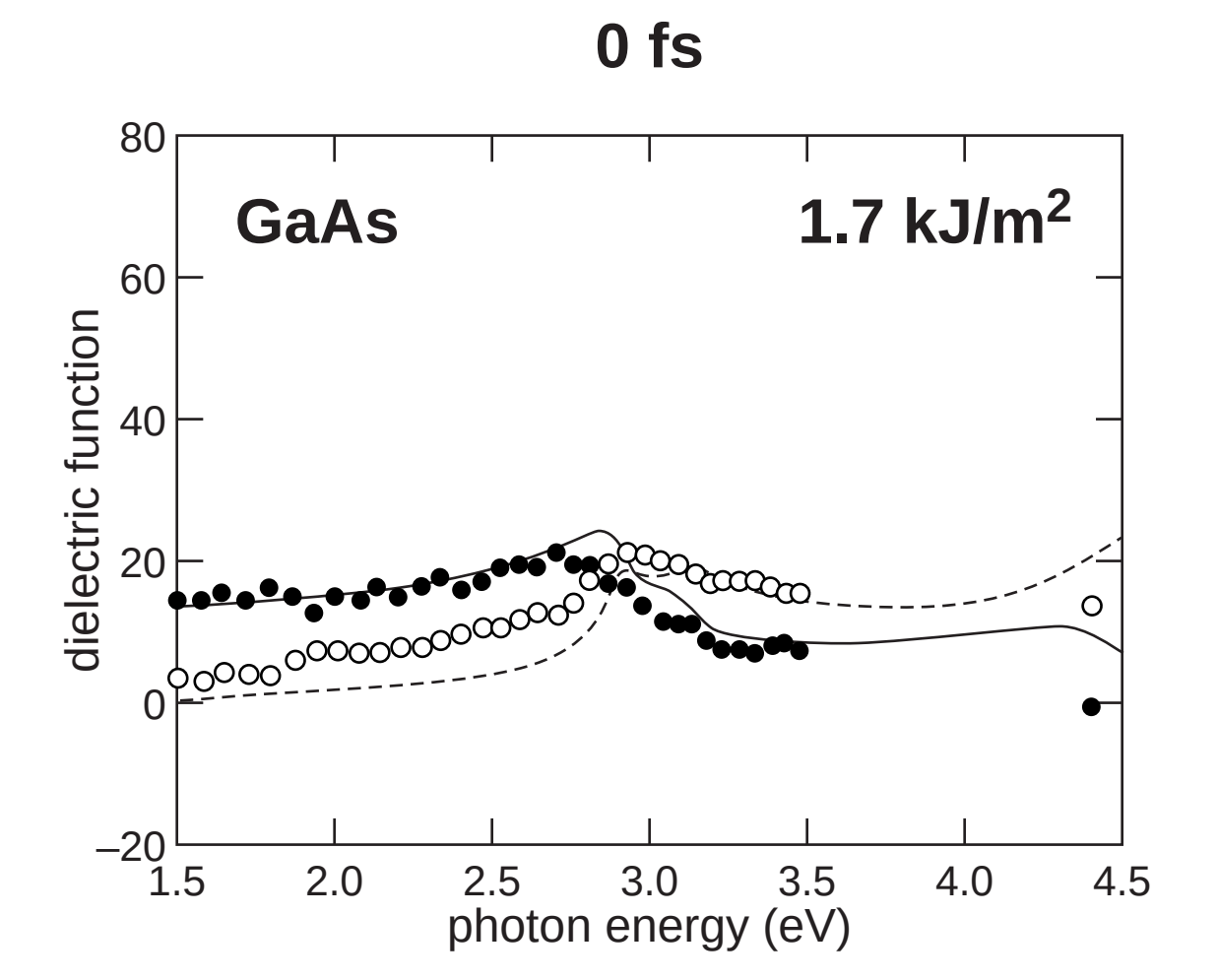
***Phys. Rev. Lett.* 80, 185 (1998)**

Technique



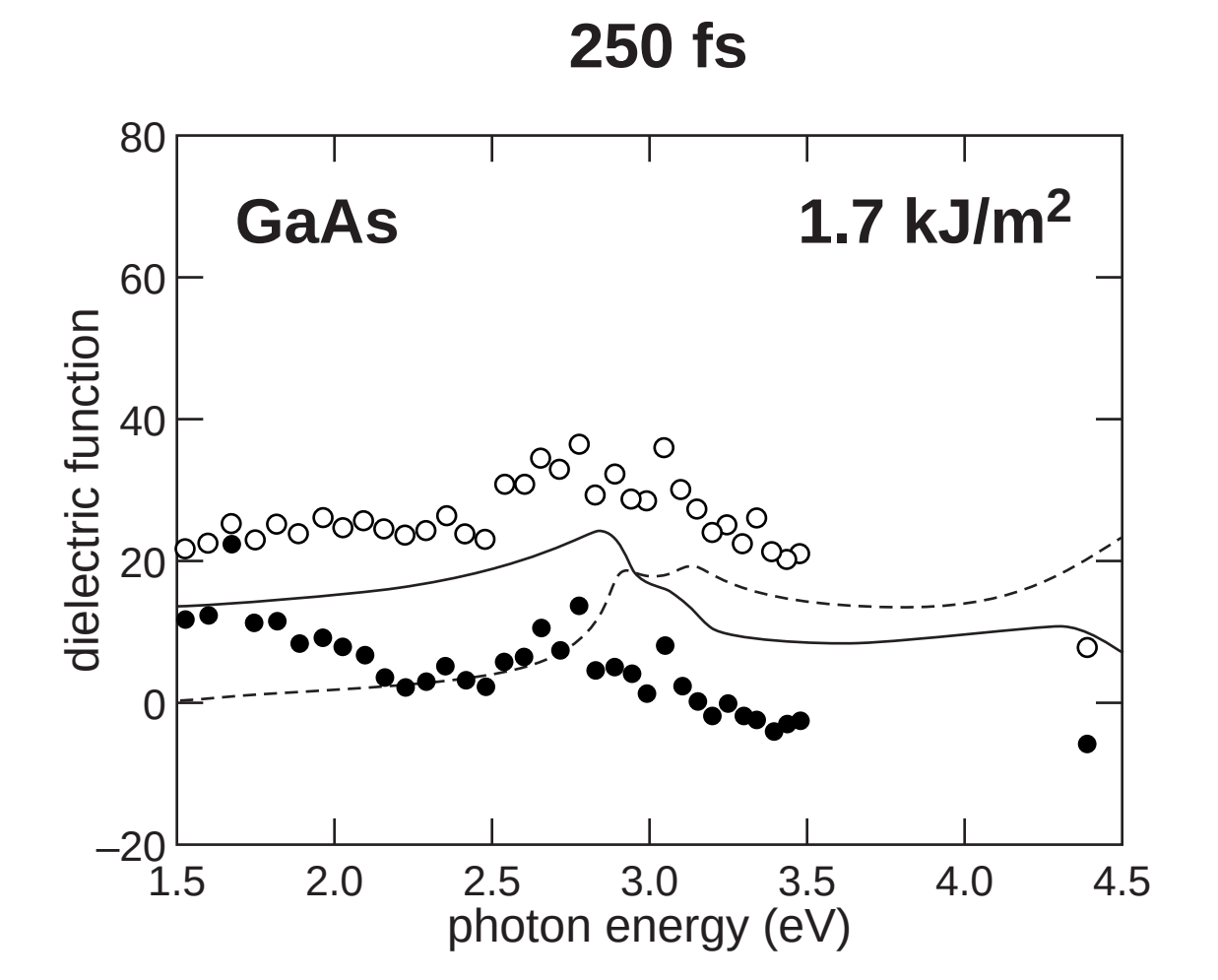
***Phys. Rev. Lett.* 80, 185 (1998)**

Technique



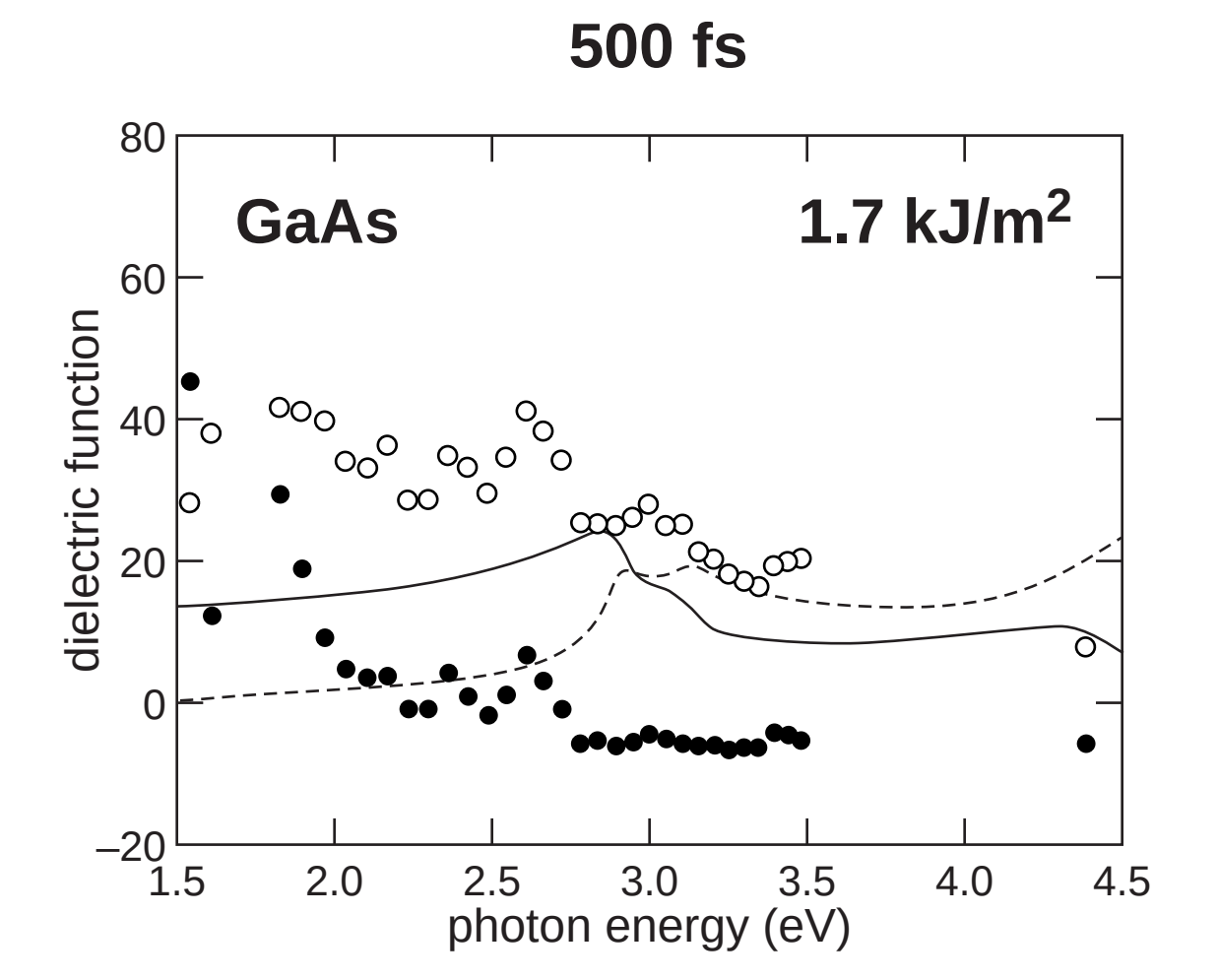
Phys. Rev. Lett. **80, 185 (1998)**

Technique



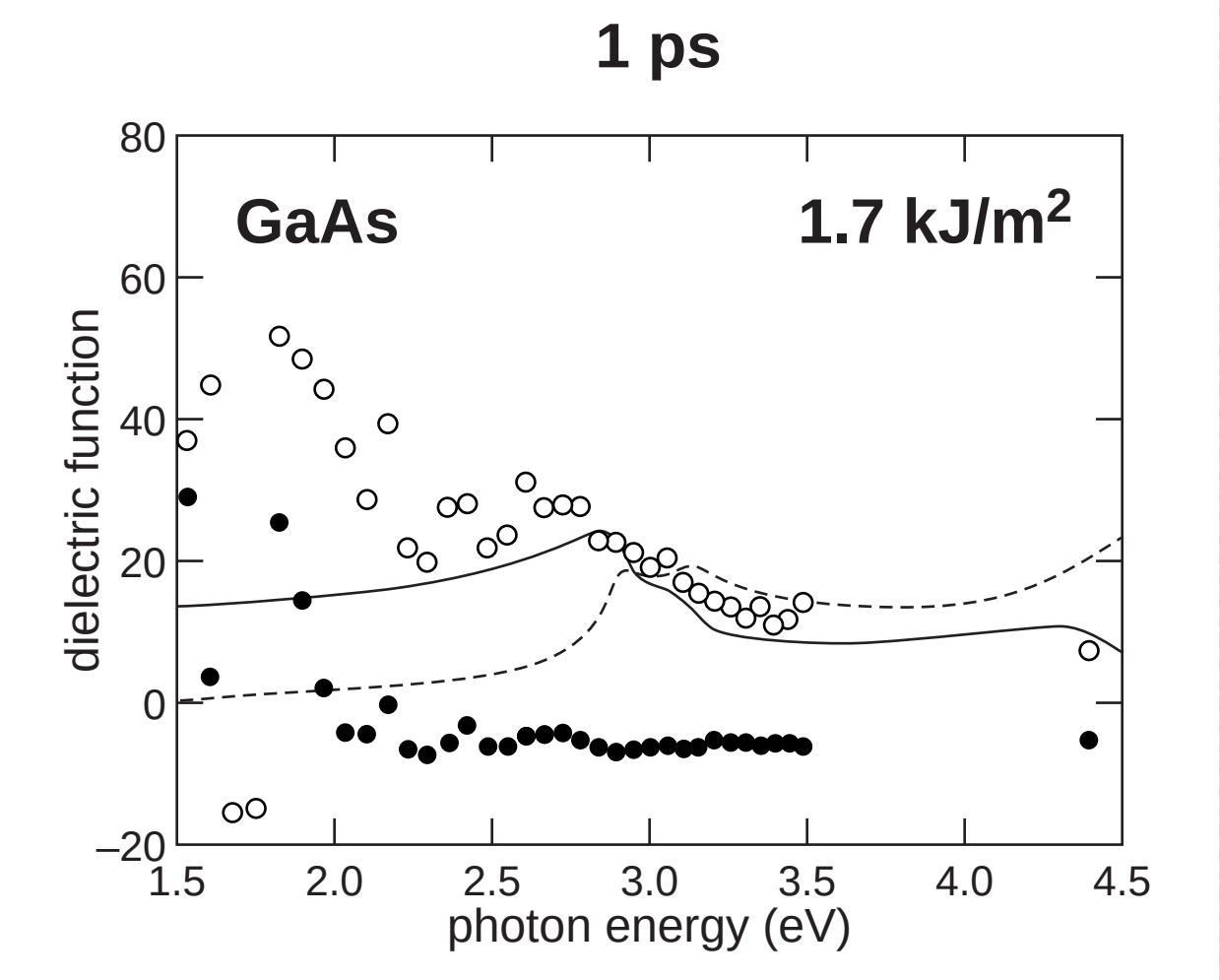
***Phys. Rev. Lett.* 80, 185 (1998)**

Technique



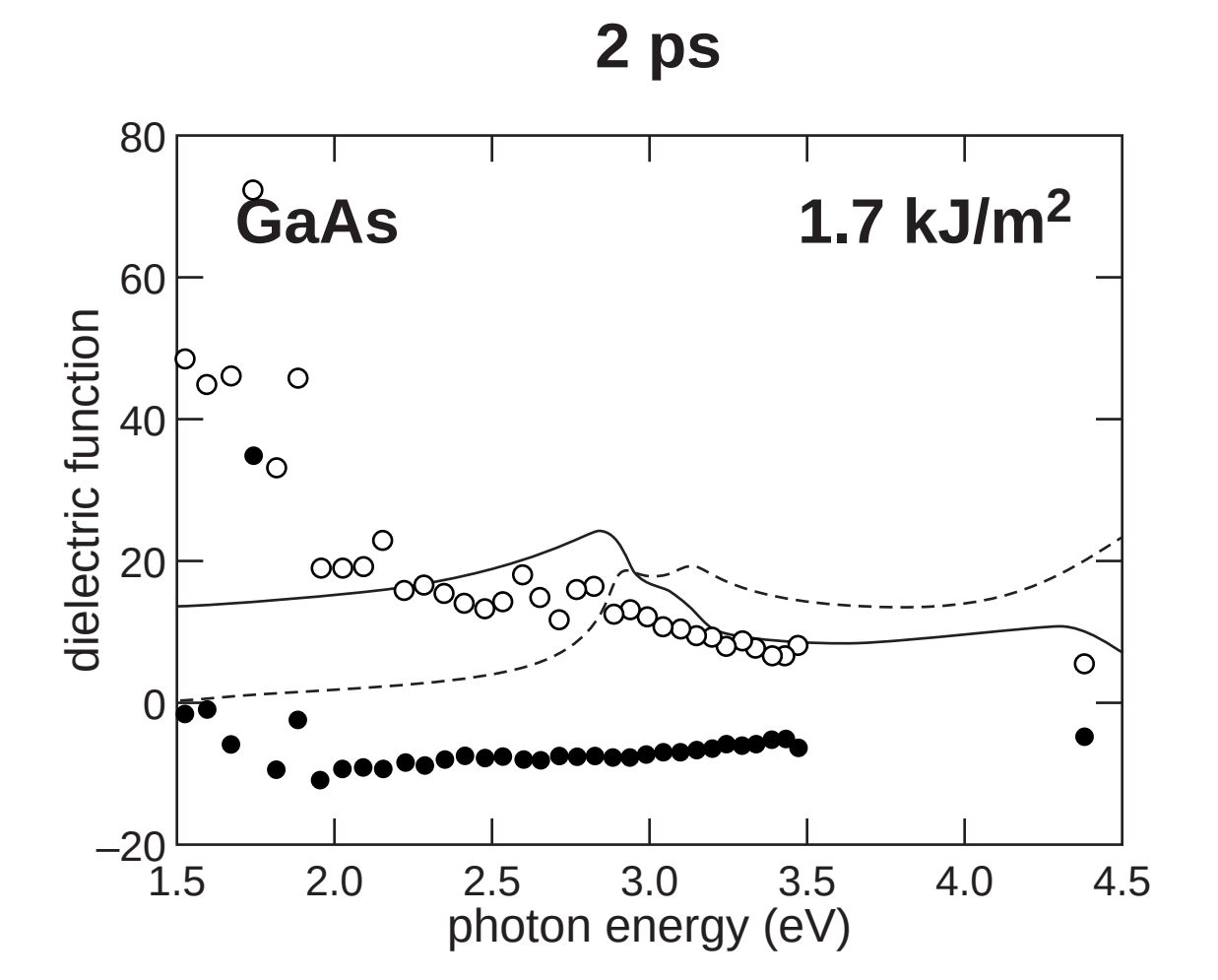
Phys. Rev. Lett. 80, 185 (1998)

Technique



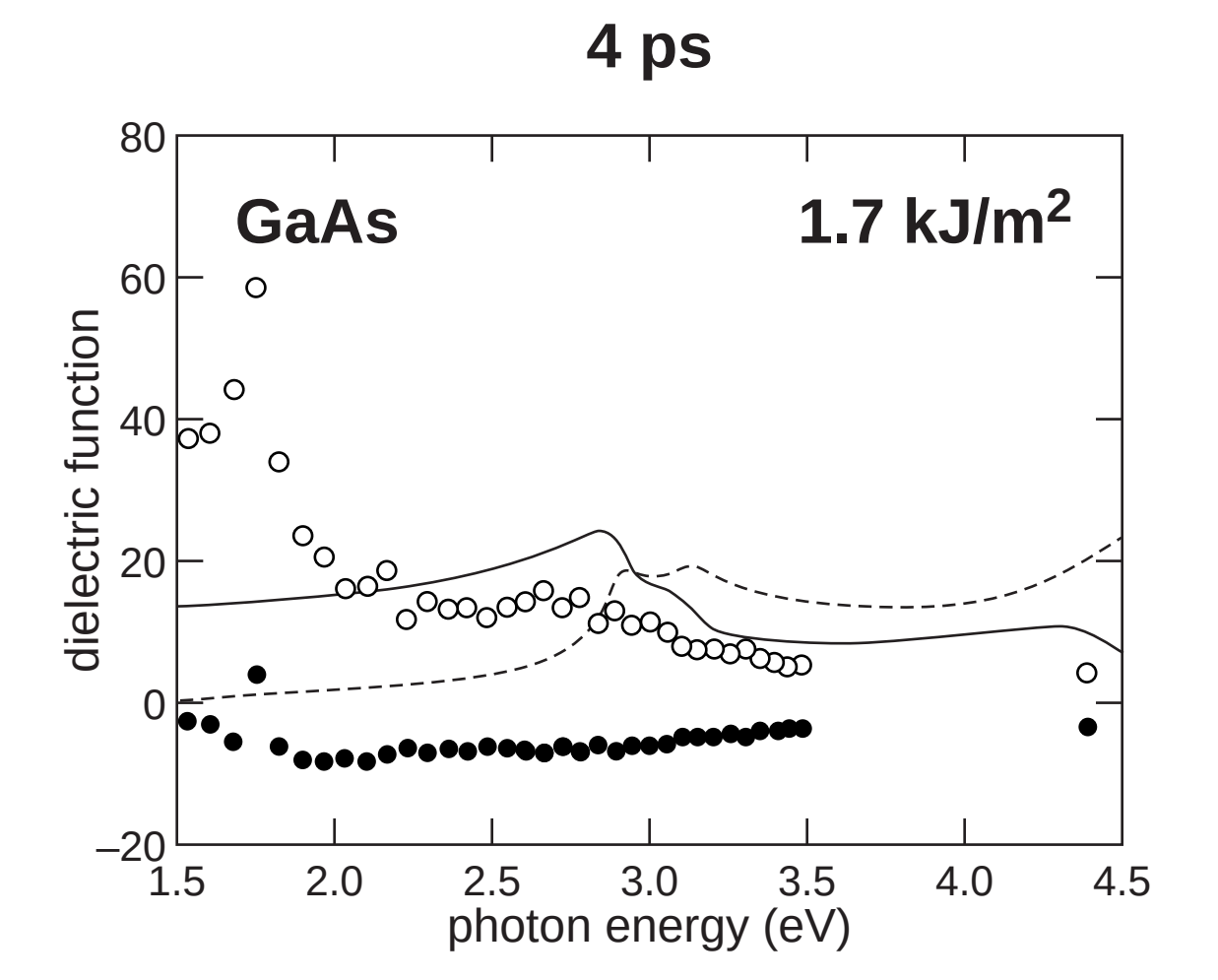
***Phys. Rev. Lett.* 80, 185 (1998)**

Technique



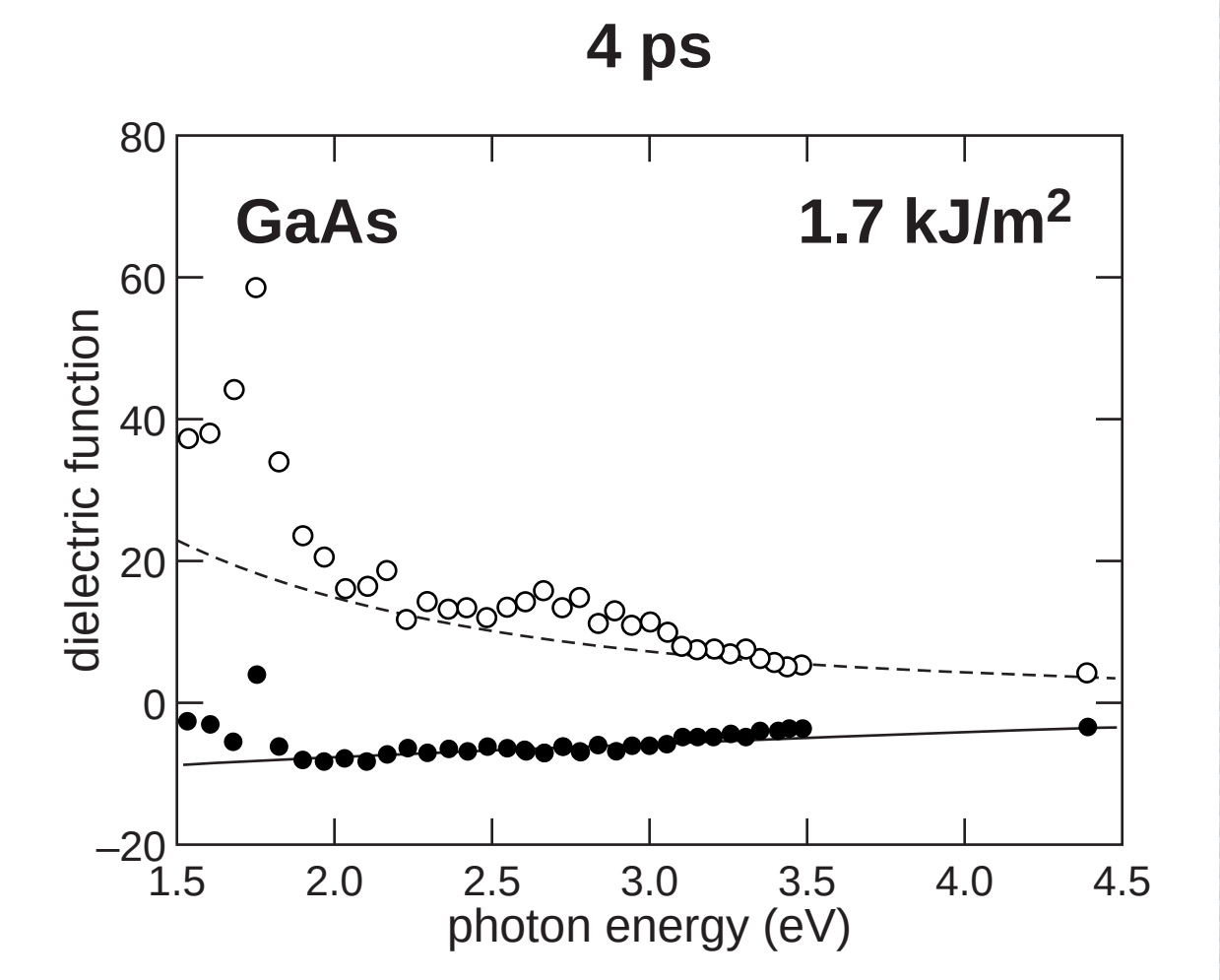
Phys. Rev. Lett. 80, 185 (1998)

Technique



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

Technique



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

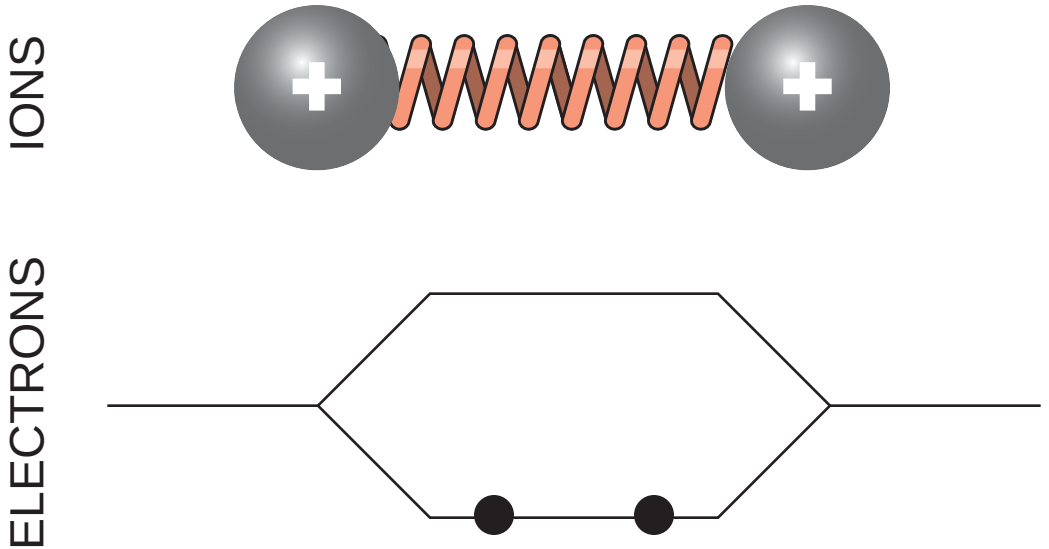
Technique

- direct observation of semiconductor-to-metal transition
- order-disorder transition
- transition structural, not electronic

Outline

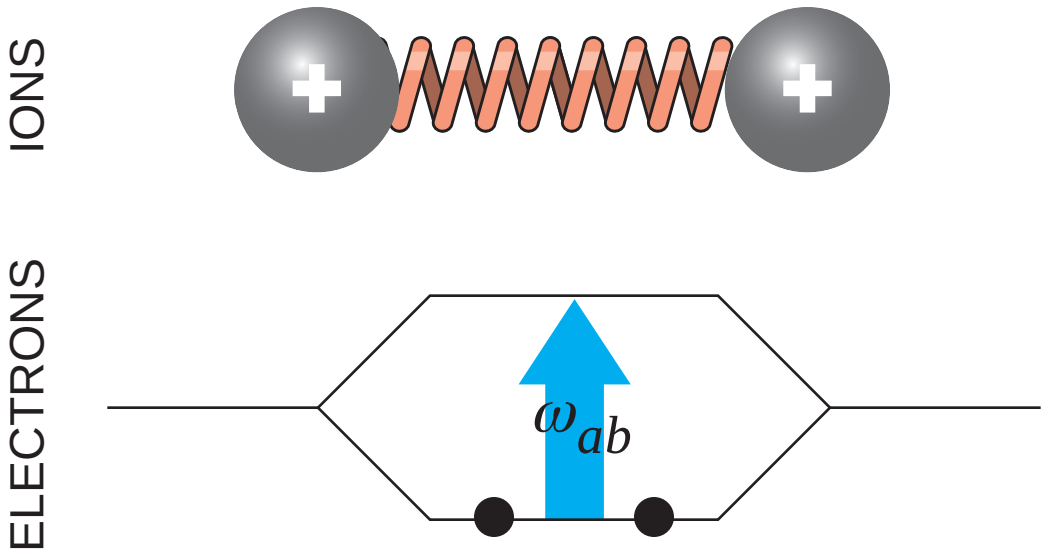
- technique
- **results**
- discussion

Displacive excitation



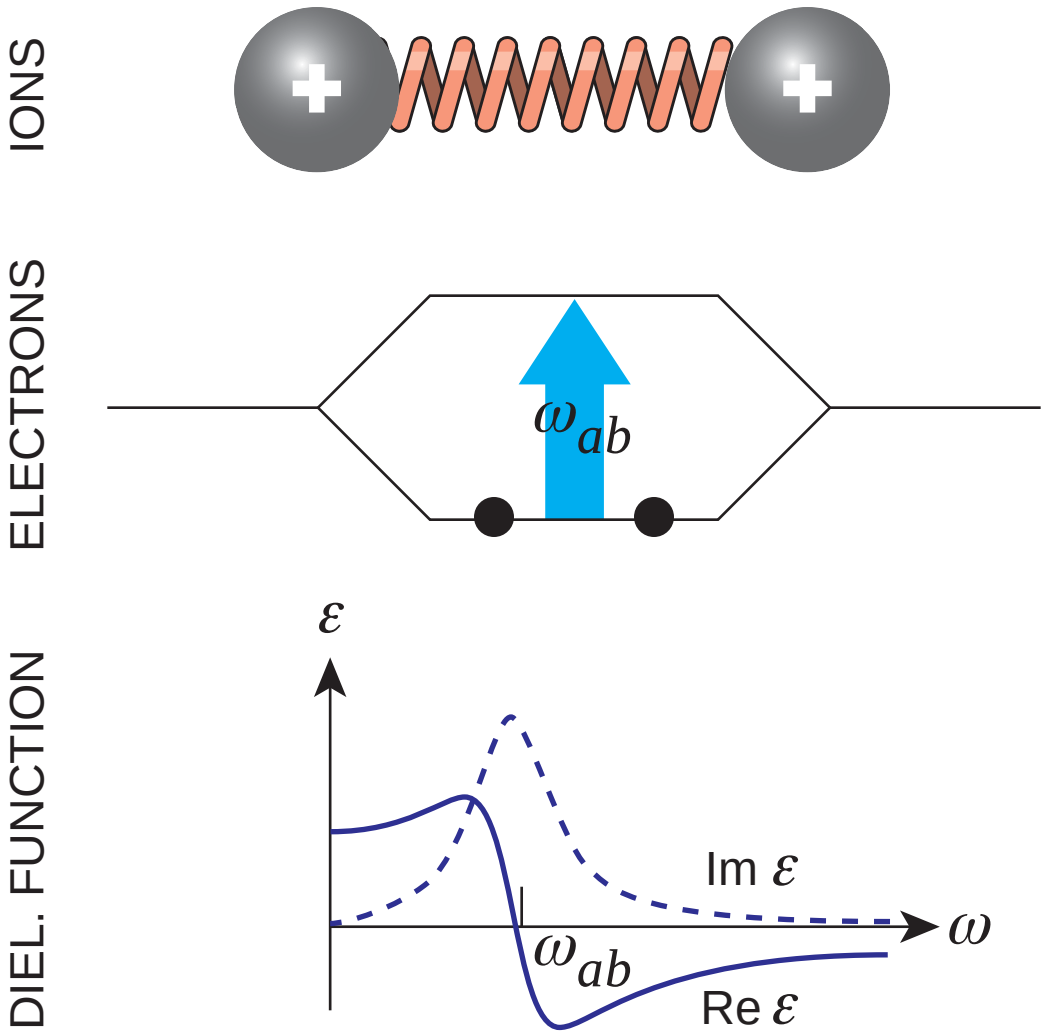
“two-atom model”

Displacive excitation



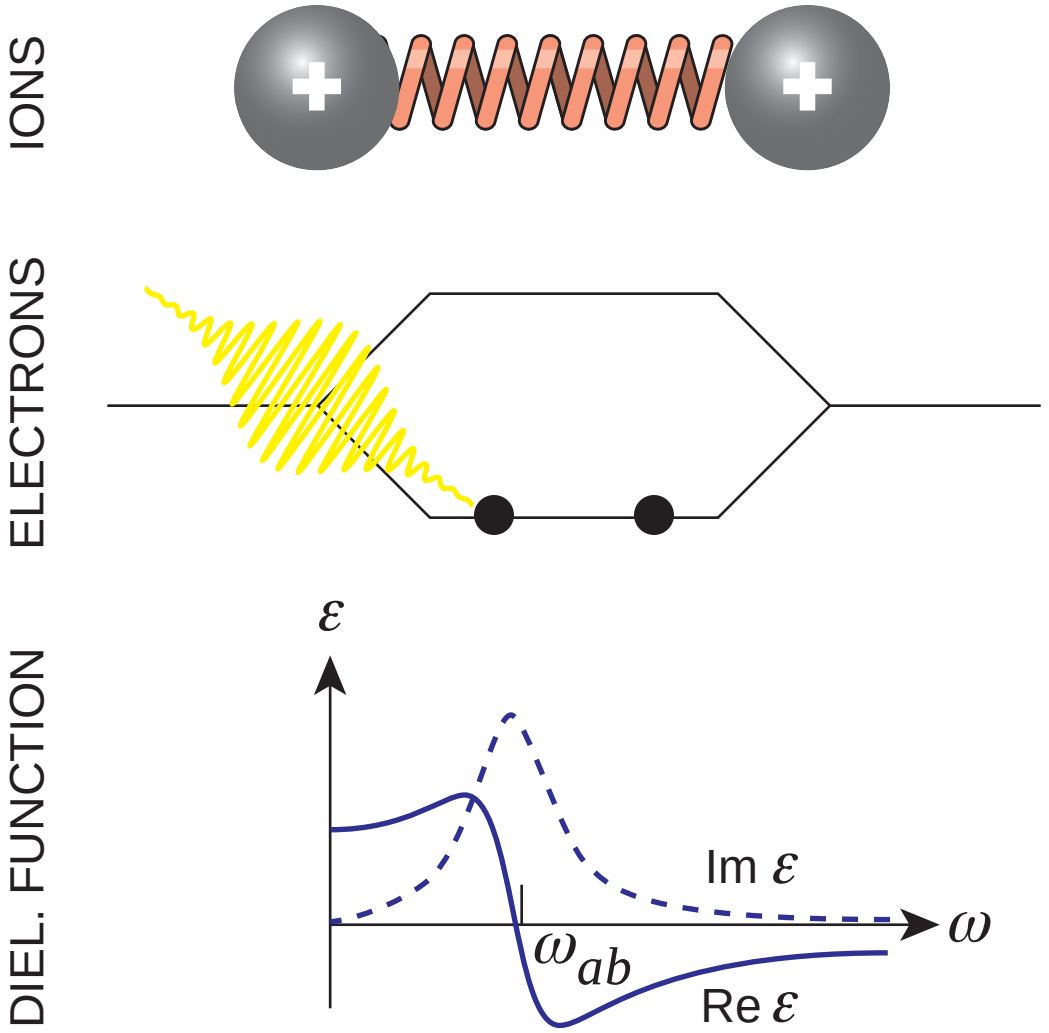
bonding-antibonding splitting

Displacive excitation



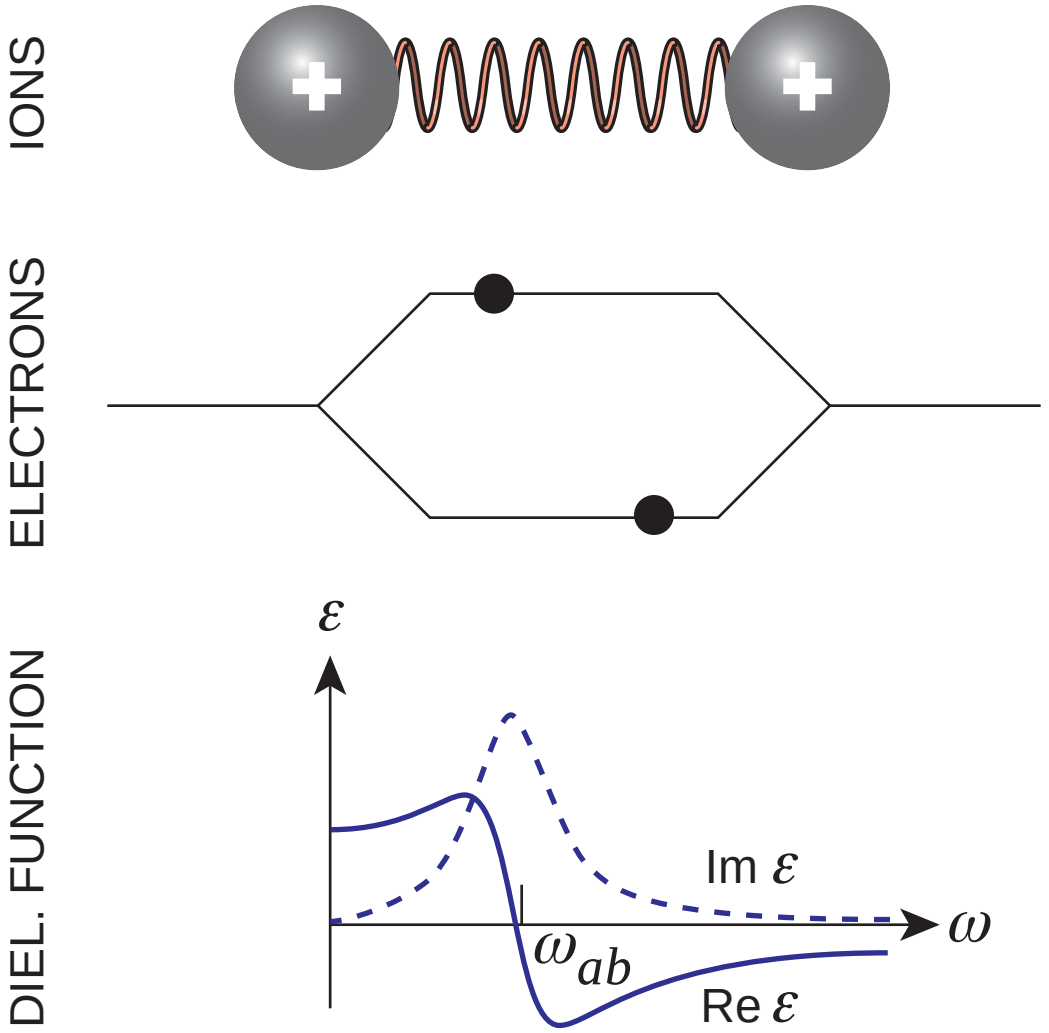
Lorentz model

Displacive excitation



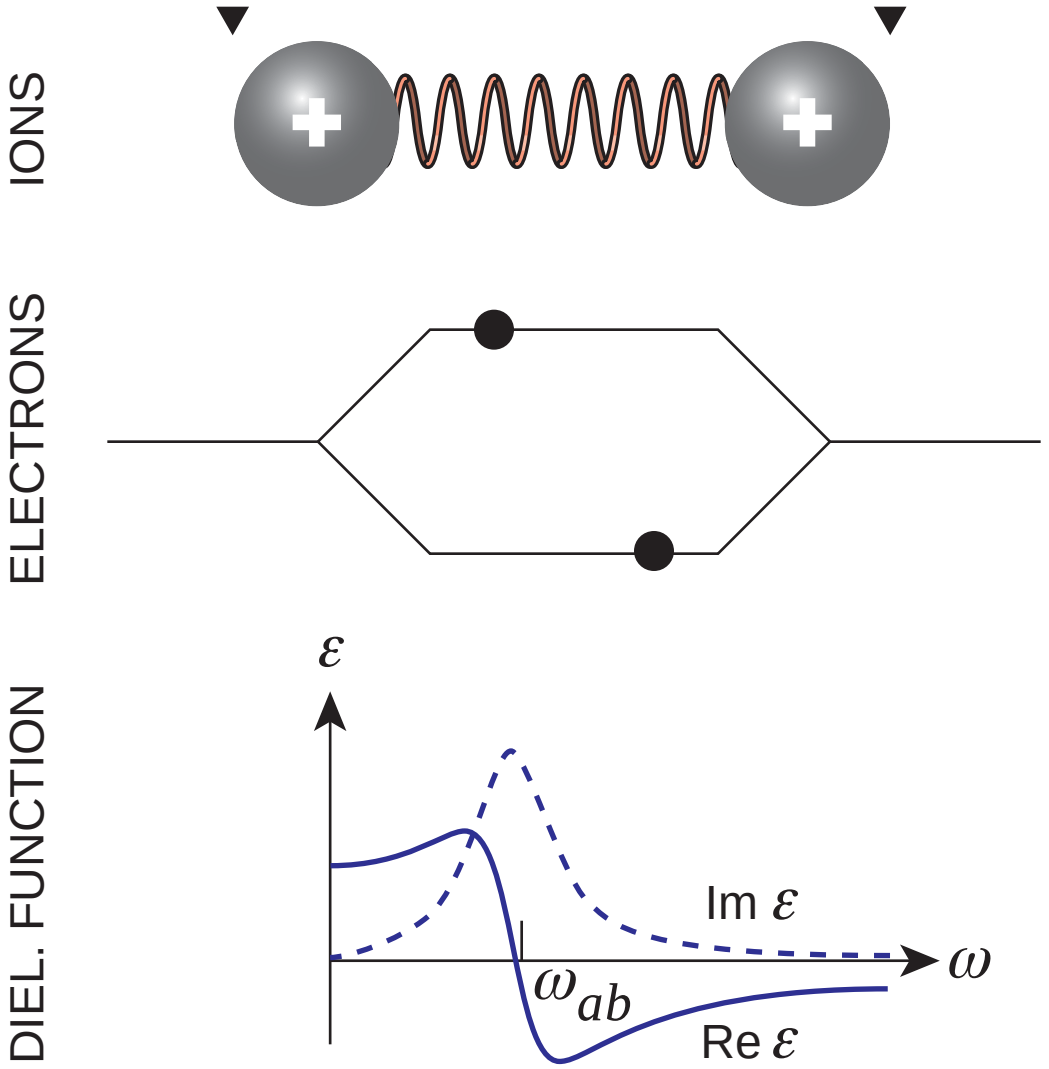
photon promotes electron…

Displacive excitation



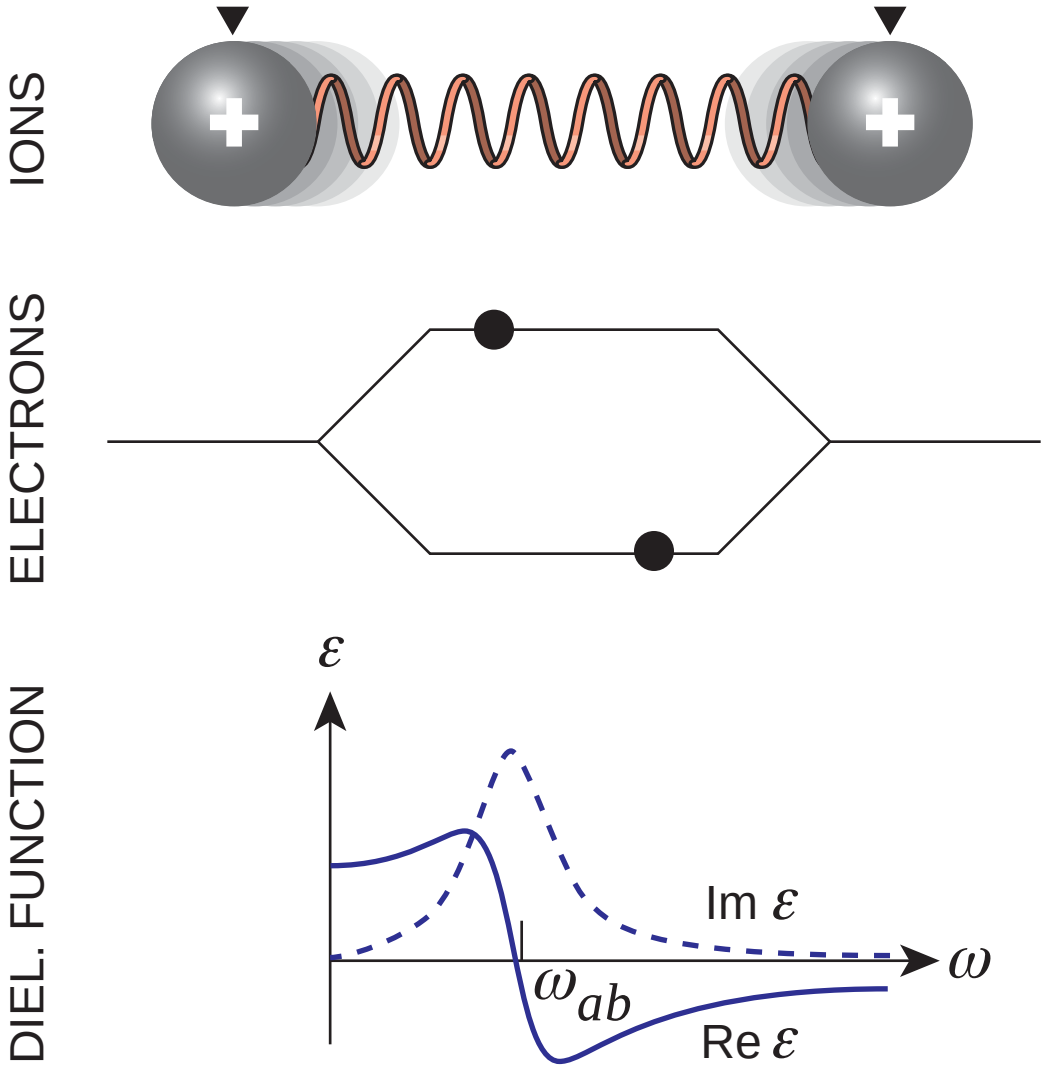
… weakening binding force…

Displacive excitation



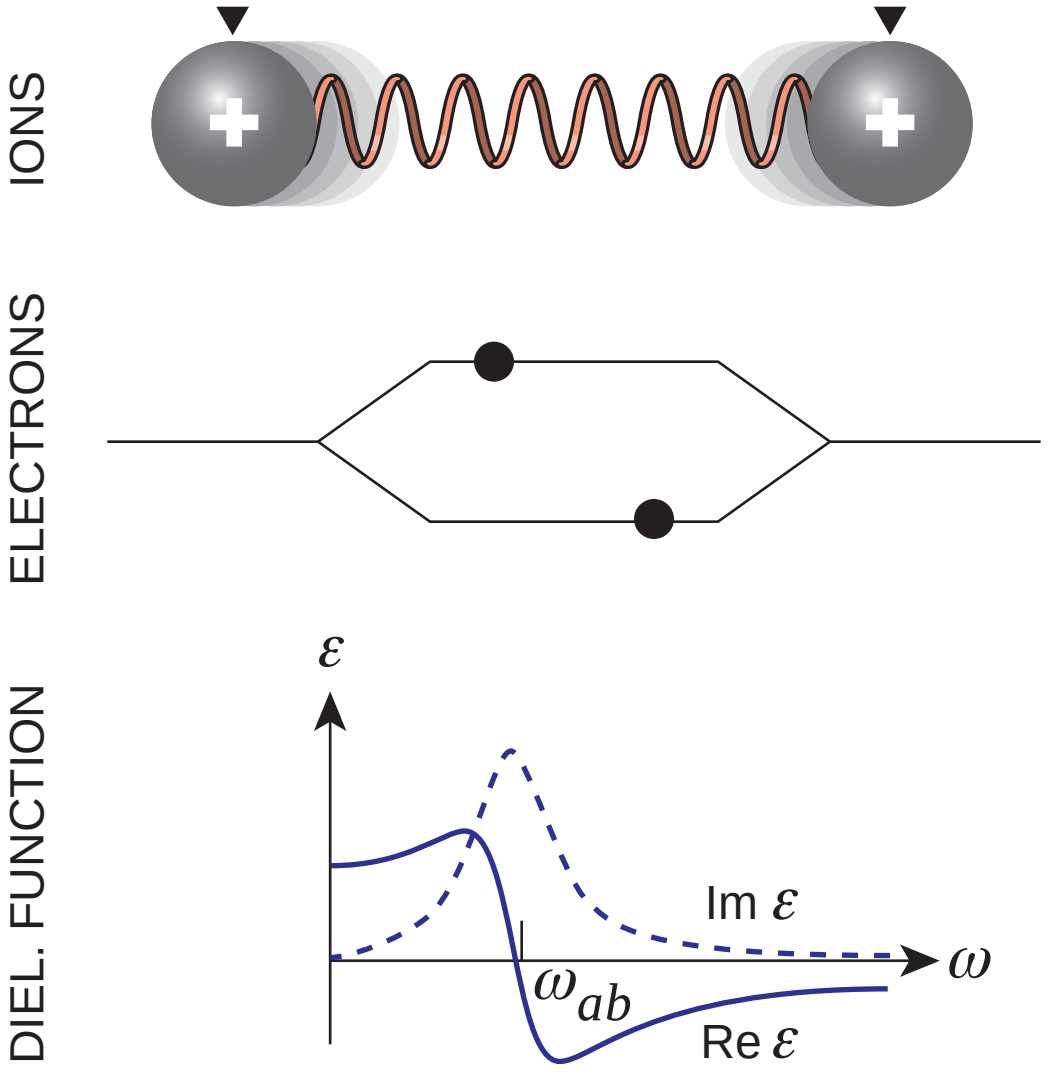
… establishing new equilibrium positions

Displacive excitation



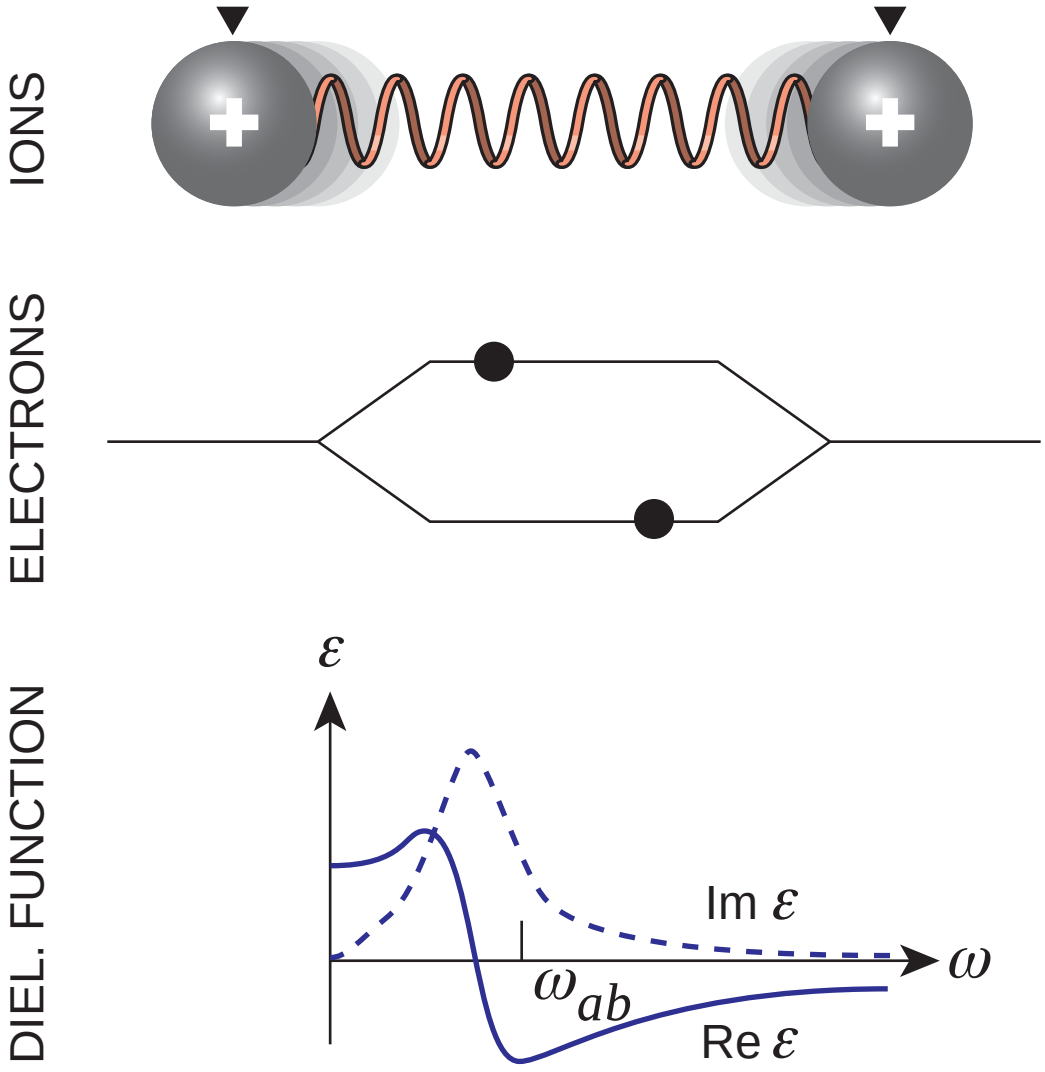
ions move to new equilibrium positions…

Displacive excitation



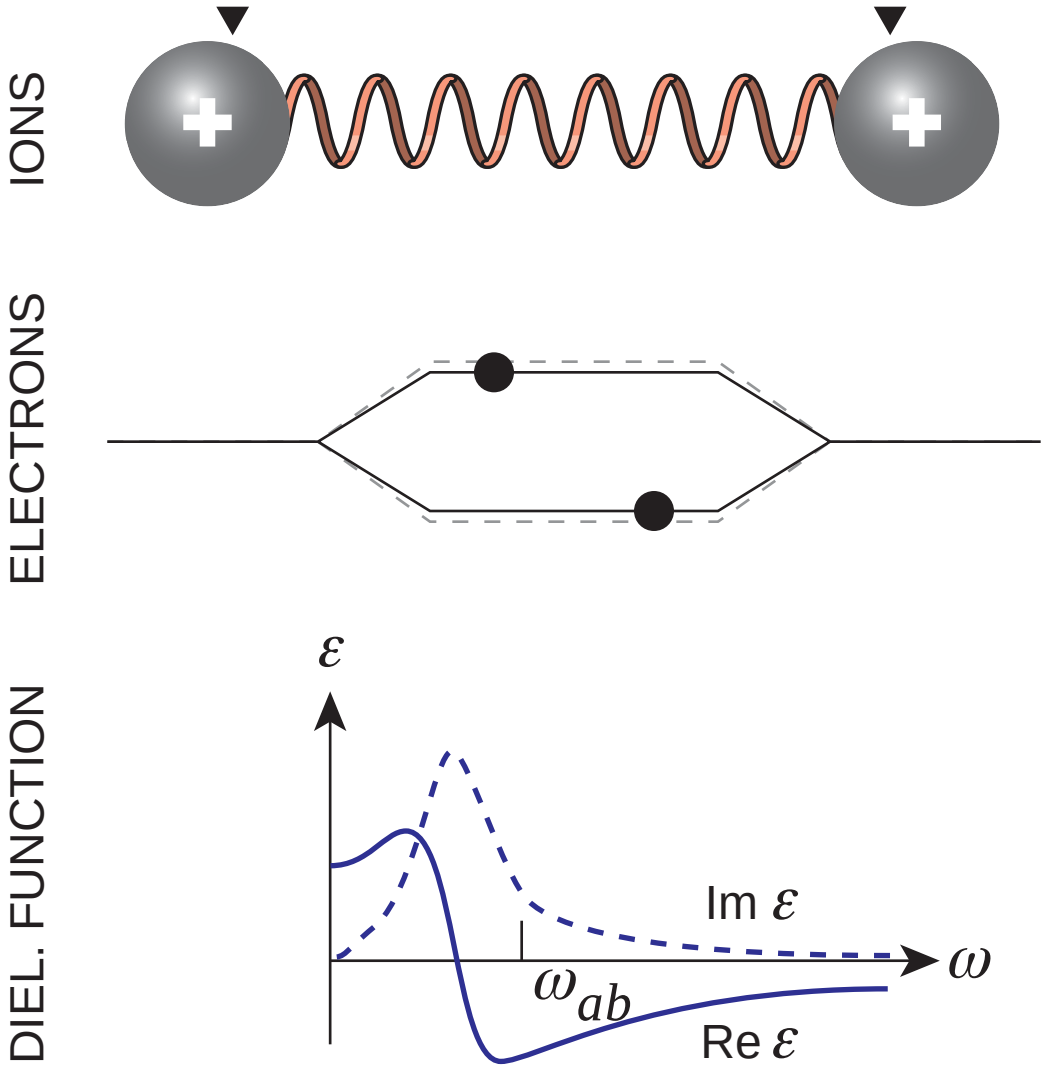
… diminishing splitting…

Displacive excitation



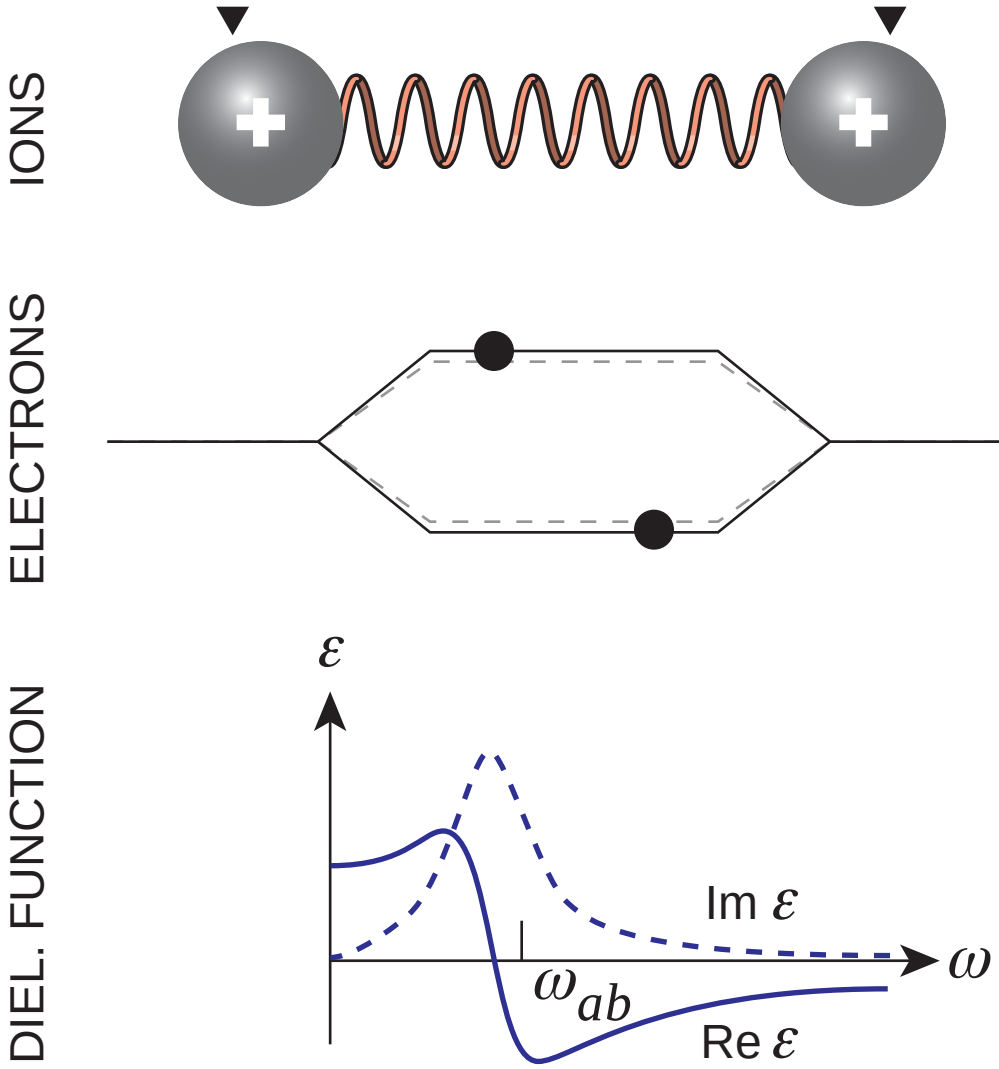
… and red-shifting the dielectric function

Displacive excitation



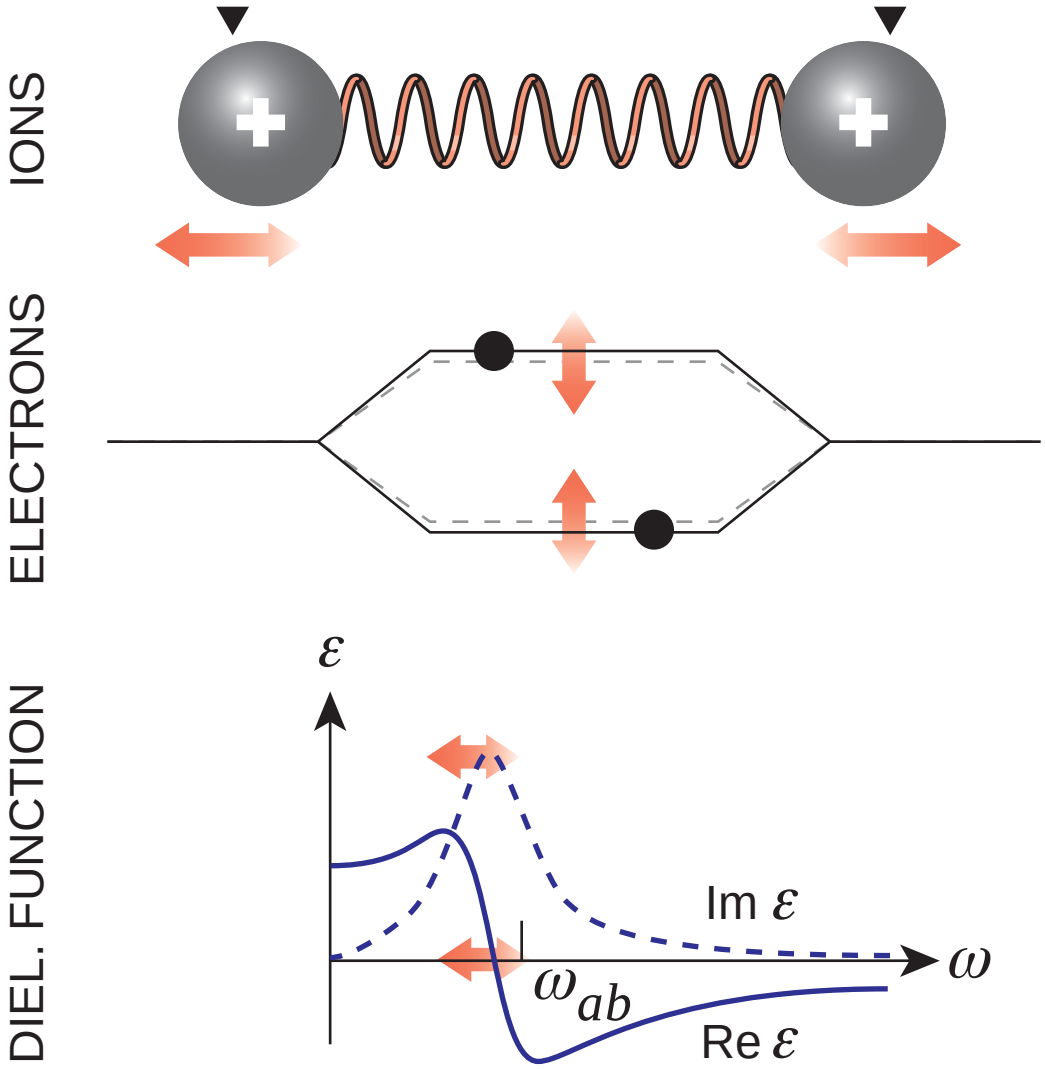
ions overshoot equilibrium position…

Displacive excitation



… reversing travel and overshooting again

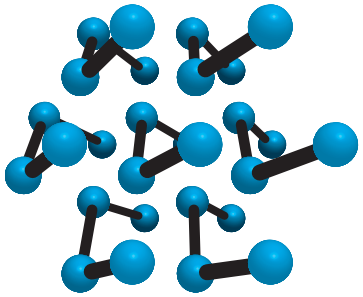
Displacive excitation



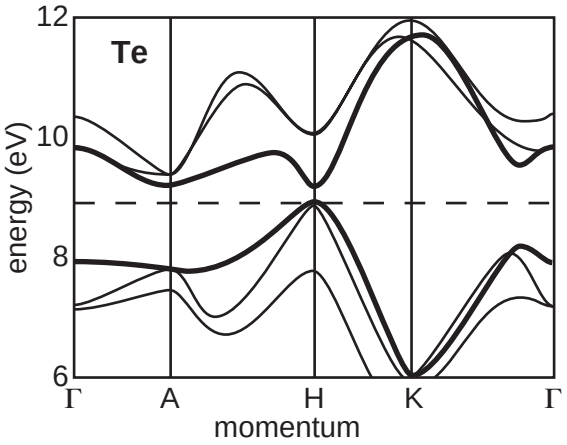
oscillation around “displaced” equilibrium position

Displacive excitation

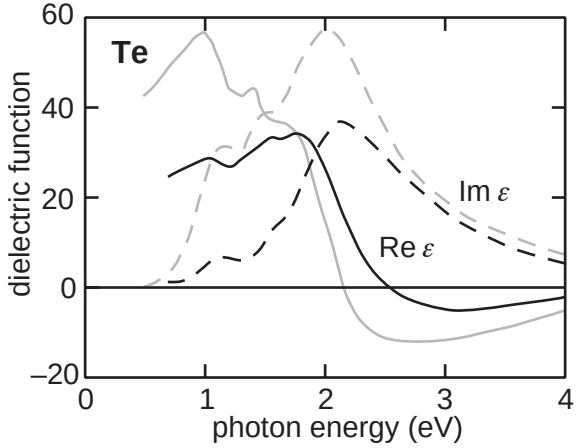
structure



band structure

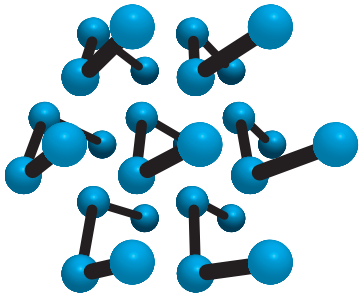


dielectric function

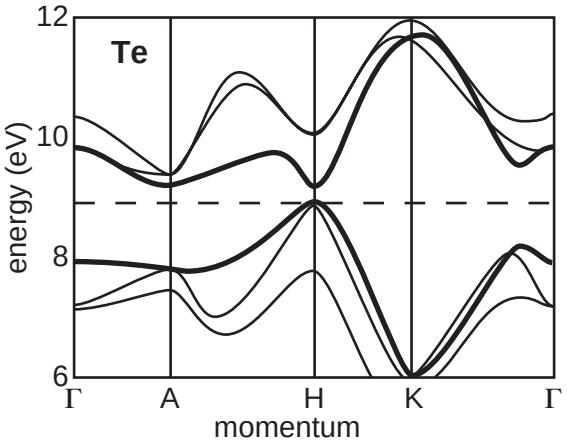


Displacive excitation

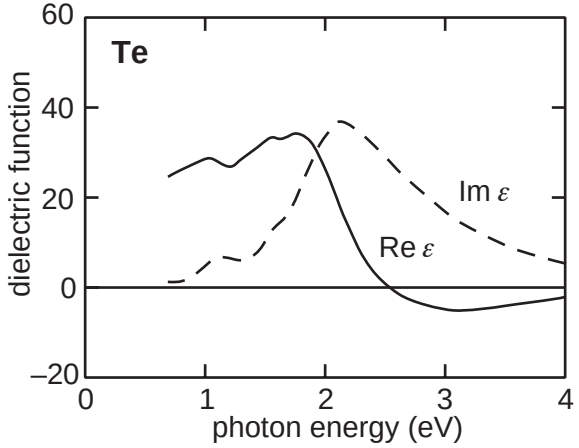
structure



band structure

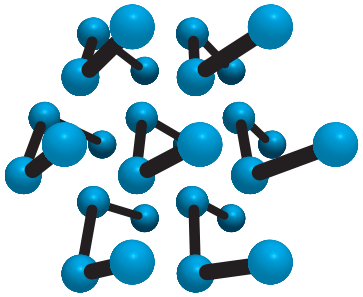


dielectric function

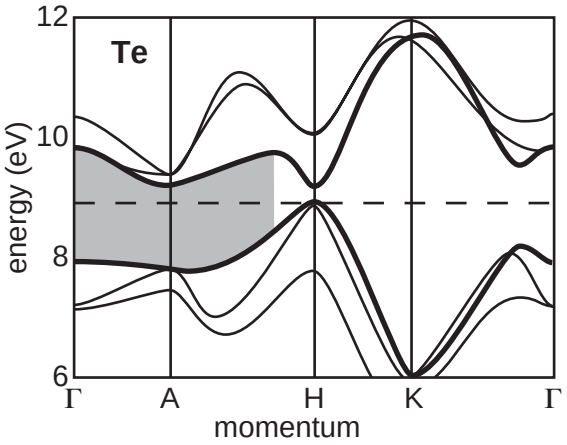


Displacive excitation

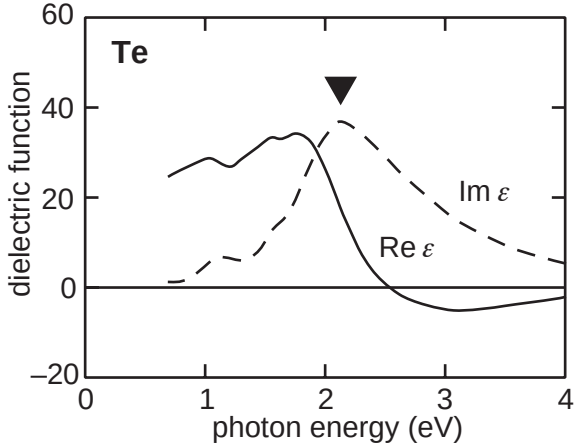
structure



band structure

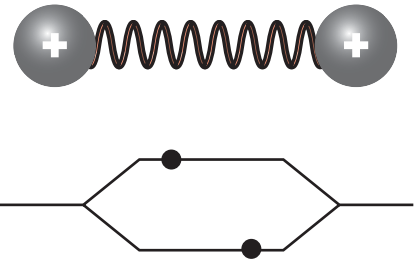


dielectric function

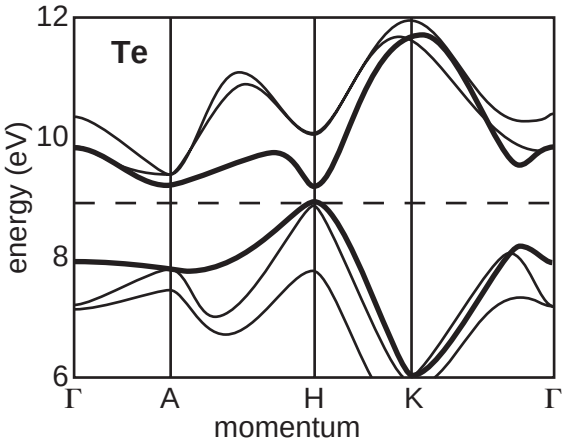


Displacive excitation

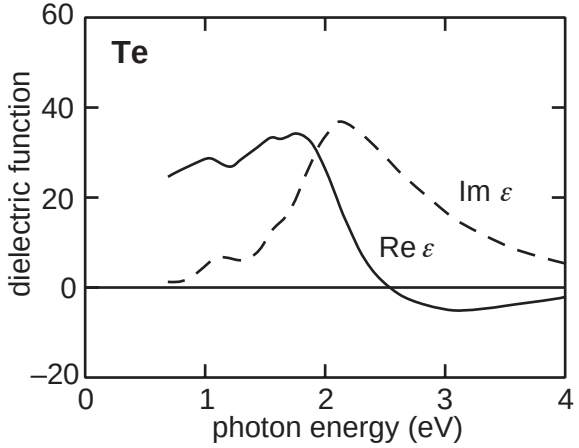
two-atom model



band structure

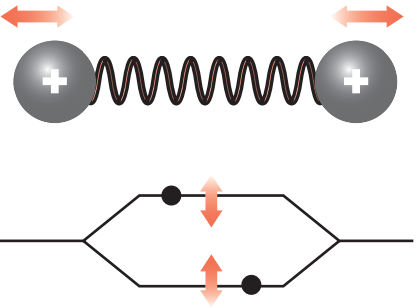


dielectric function

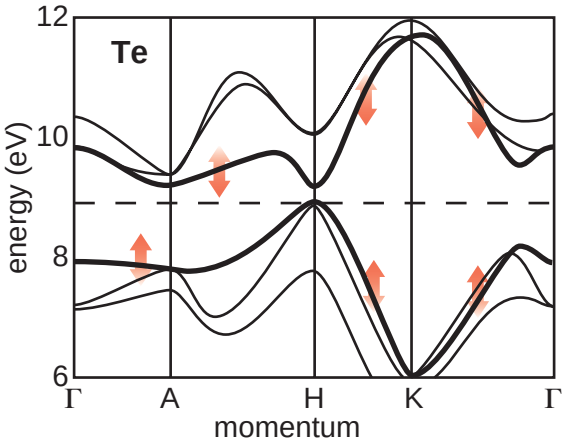


Displacive excitation

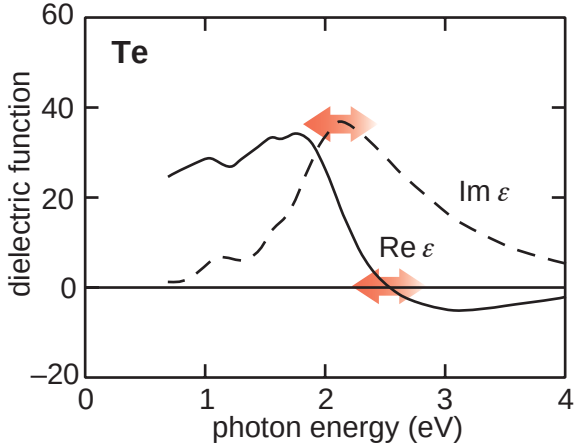
two-atom model



band structure



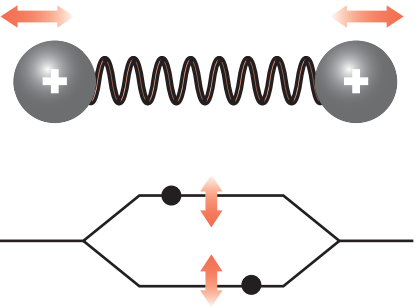
dielectric function



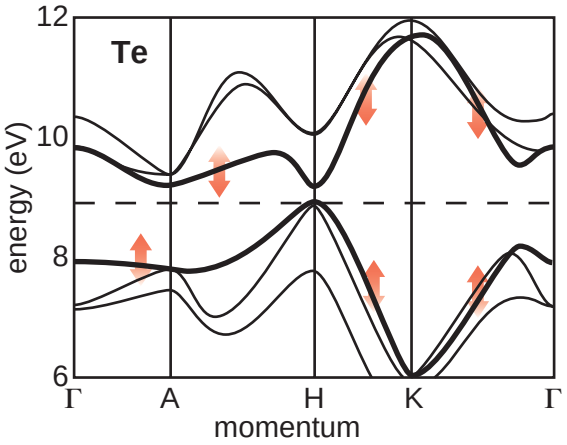
red-shift followed by oscillation

Displacive excitation

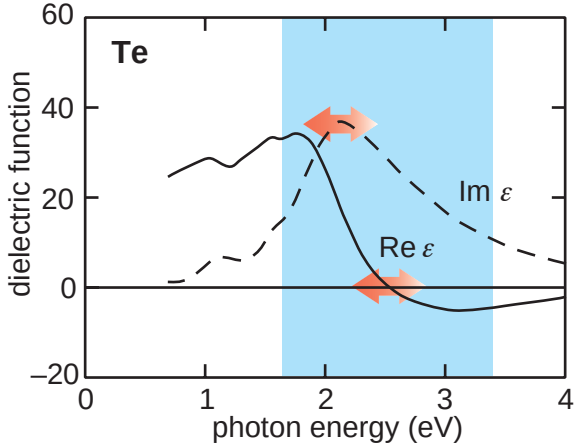
two-atom model



band structure

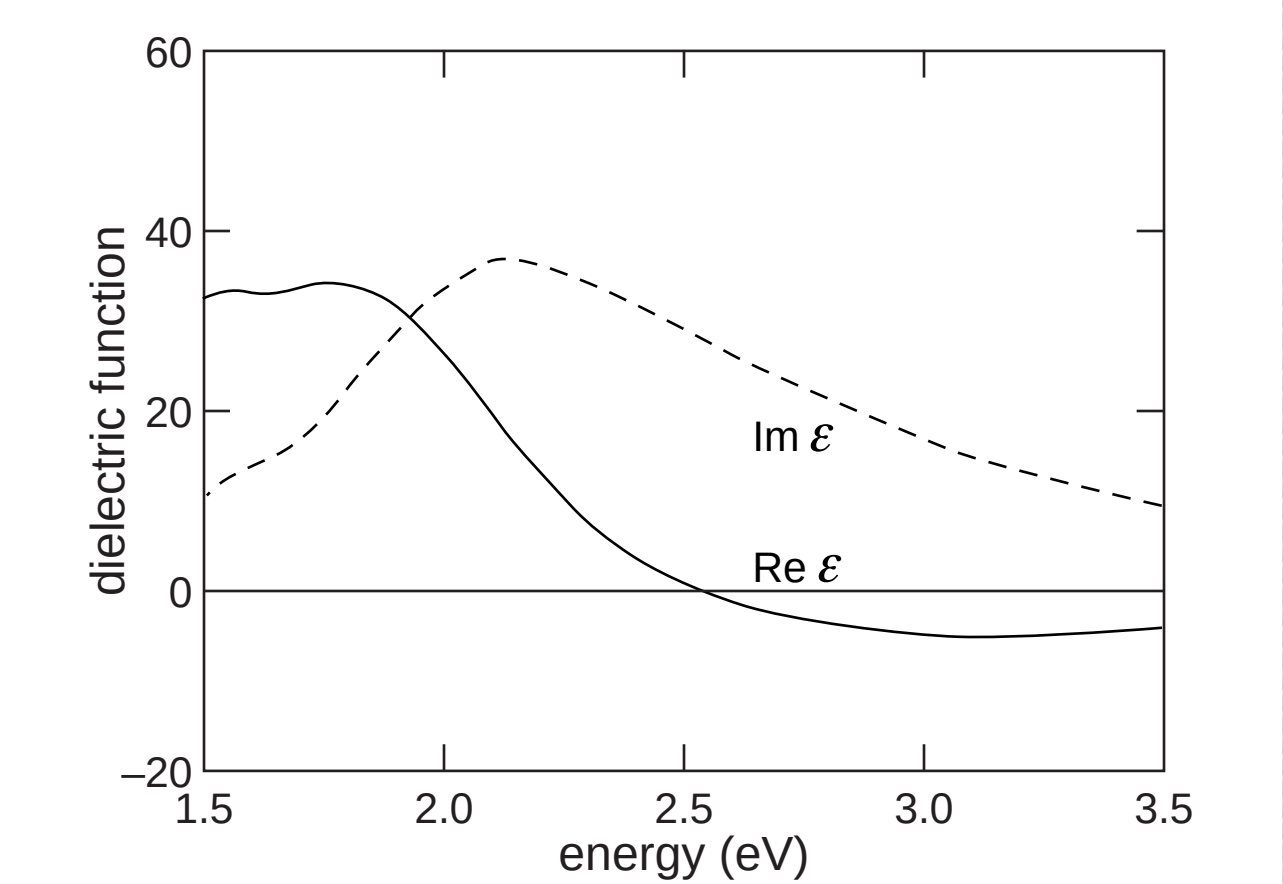


dielectric function

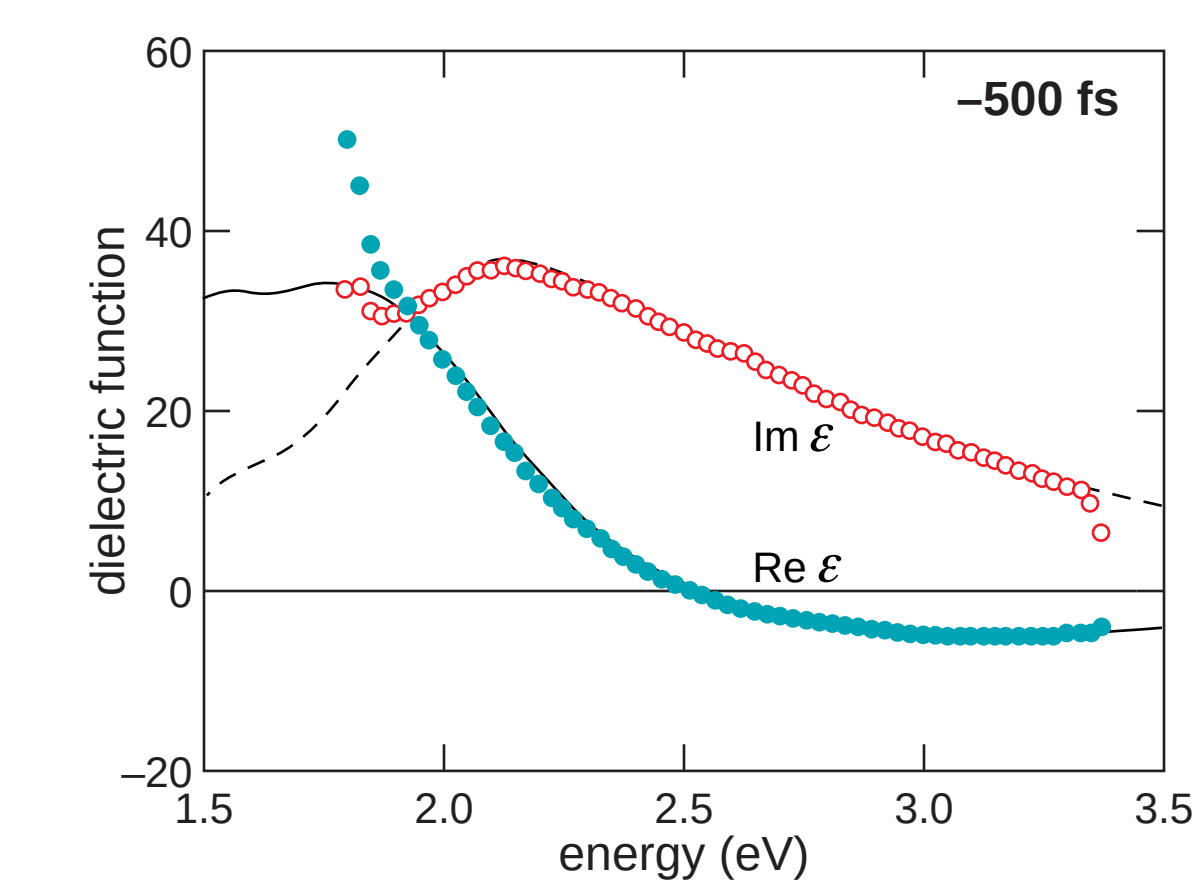


red-shift followed by oscillation

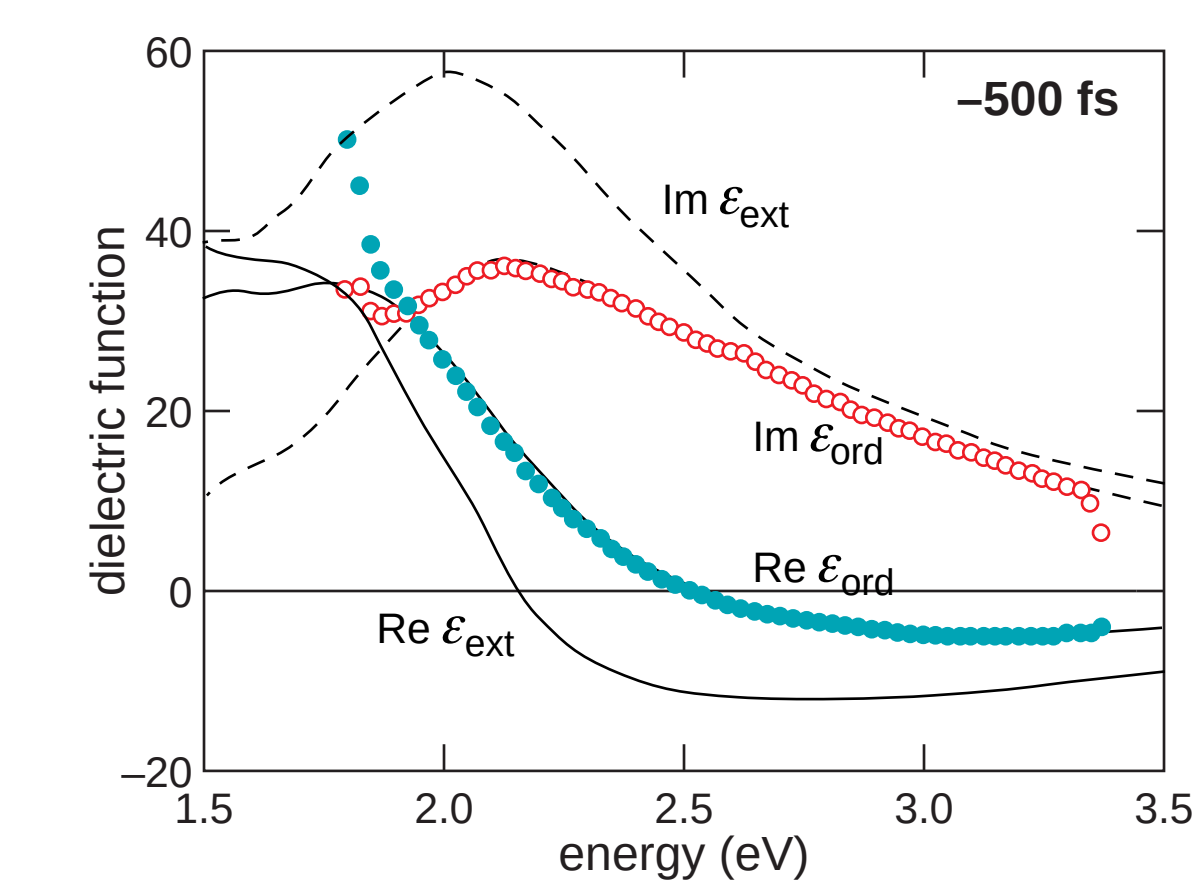
Results


60
40
20
0
–20
dielectric function
Im ε
Re ε
1.5
2.0
2.5
3.0
3.5
energy (eV)

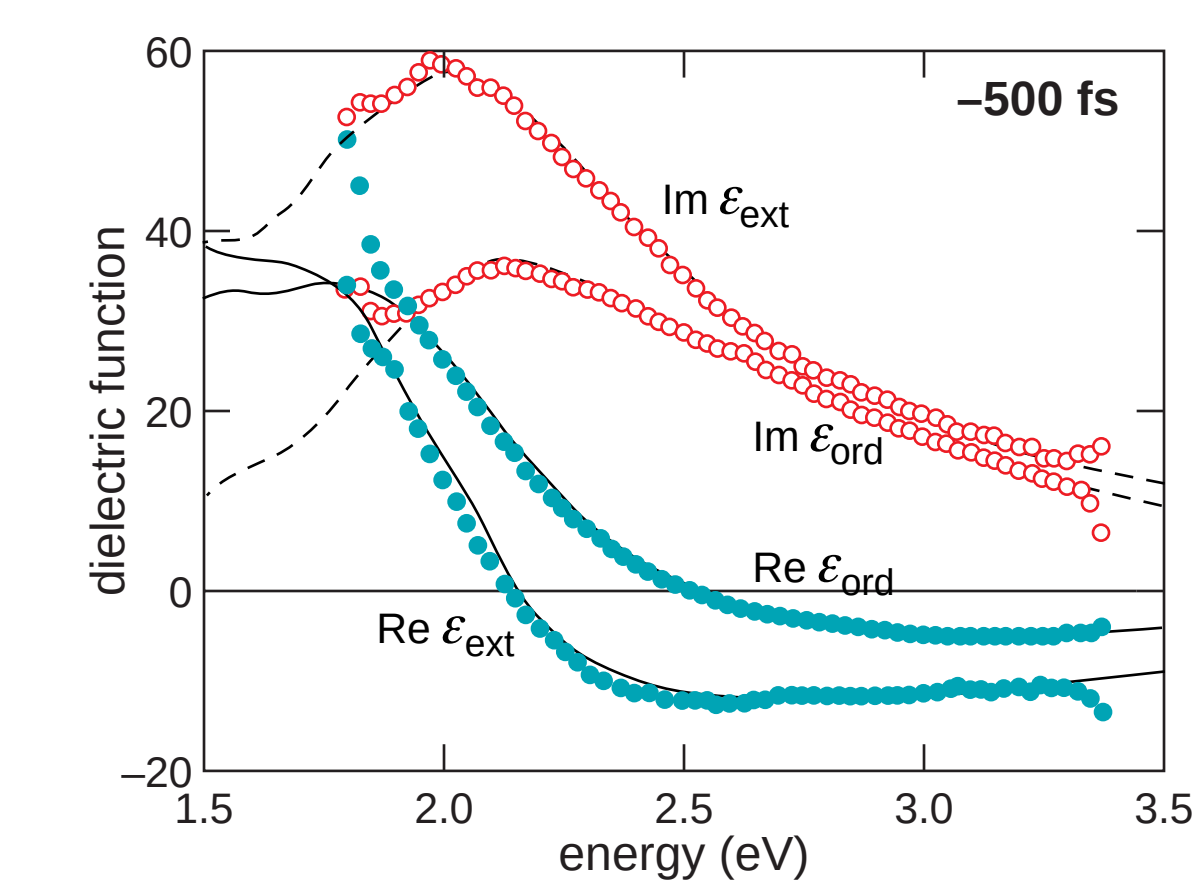
Results


60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
dielectric function
energy (eV)
–500 fs
Im ε
Re ε

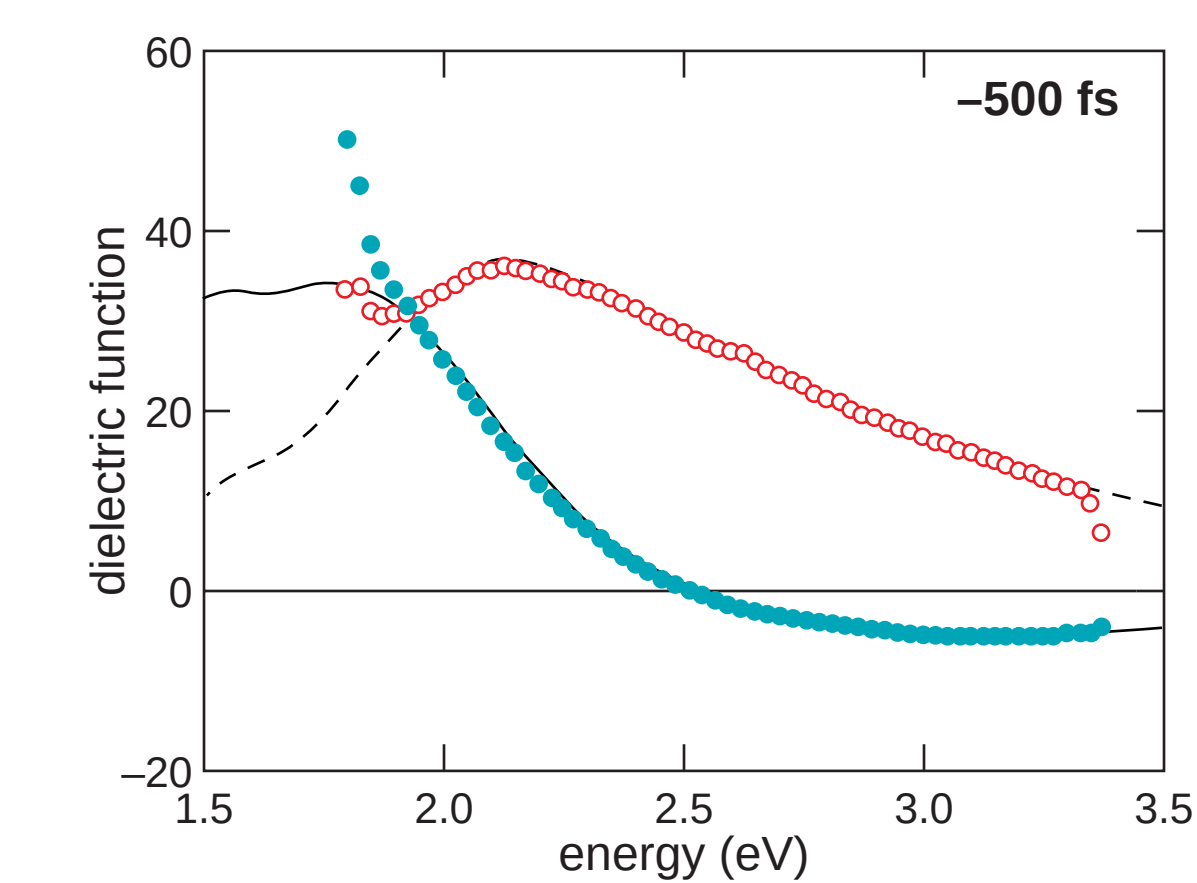
Results


–500 fs
Im ε_{ext}
Im ε_{ord}
Re ε_{ord}
Re ε_{ext}
dielectric function
energy (eV)
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5

Results


–500 fs
Im ε_{ext}
Im ε_{ord}
Re ε_{ord}
Re ε_{ext}
dielectric function
energy (eV)
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5

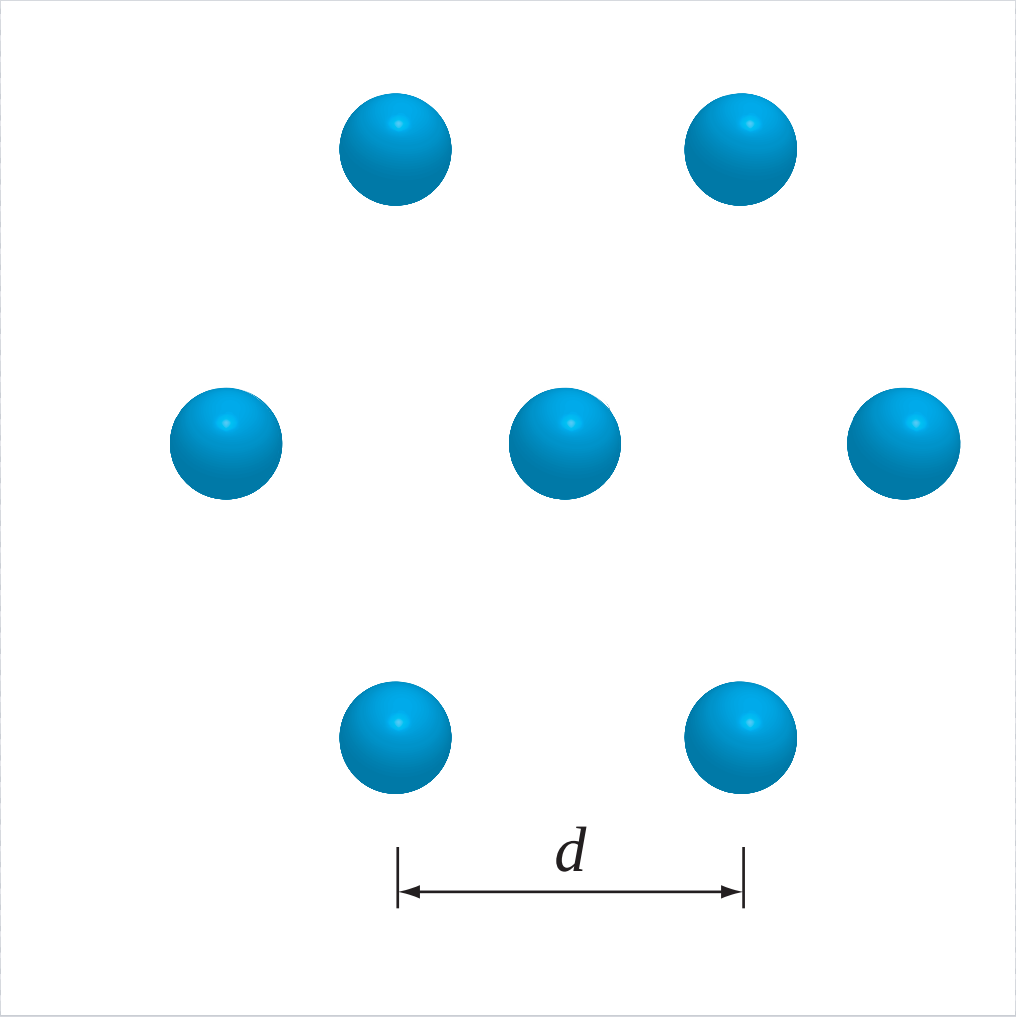
Results


–500 fs
dielectric function
energy (eV)
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5

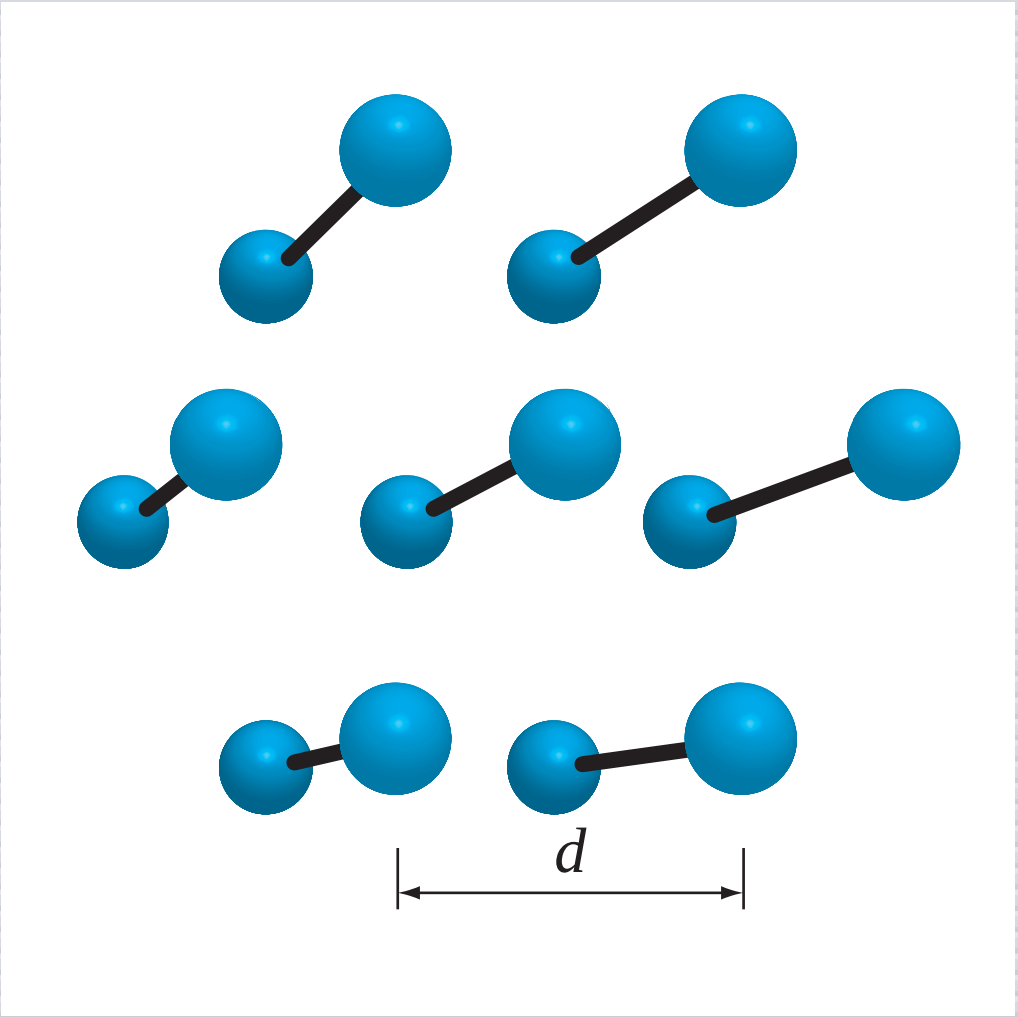
Outline

- technique
- results
- discussion

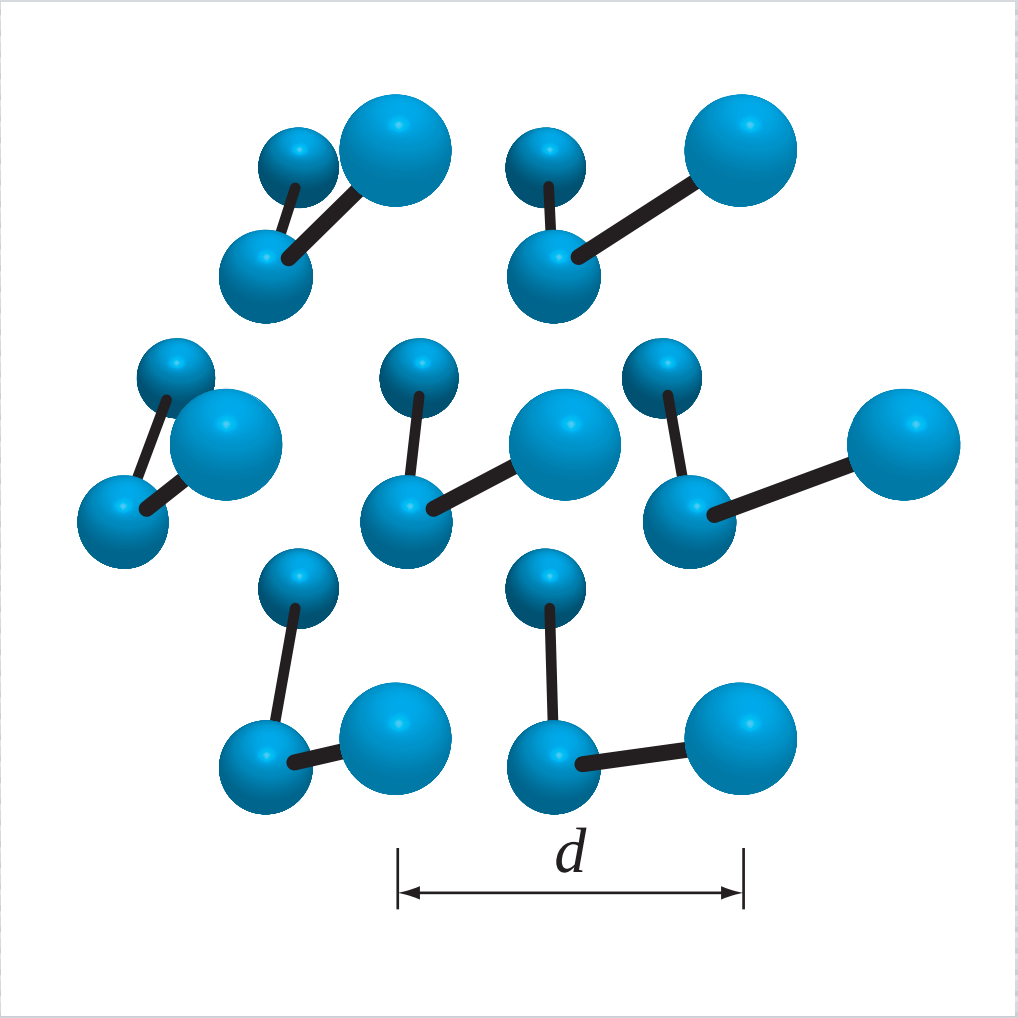
Tellurium structure



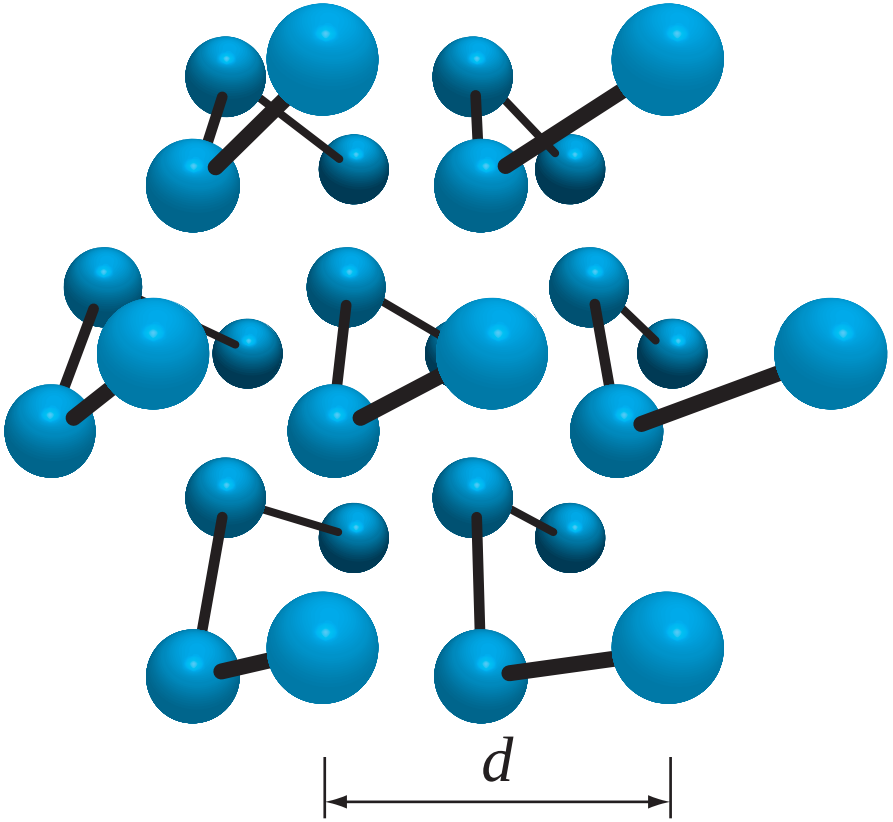
Tellurium structure



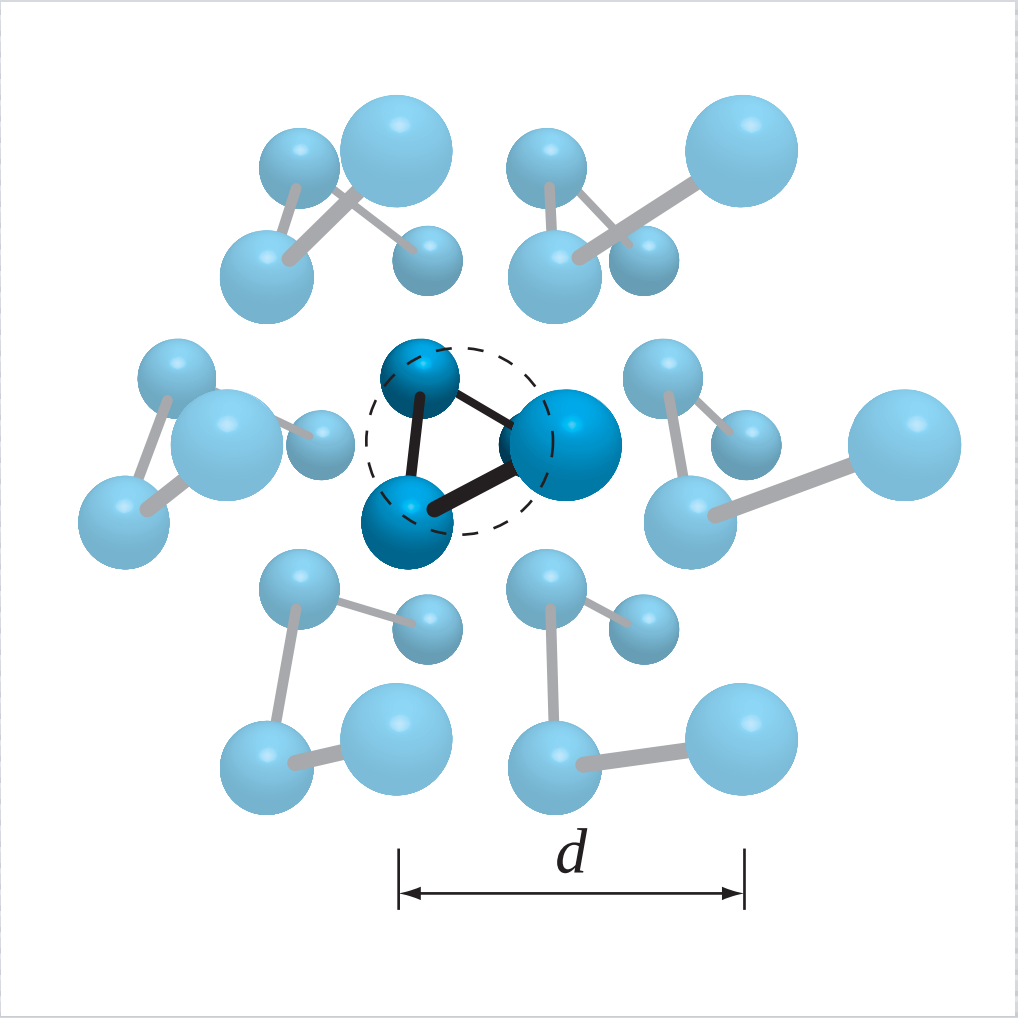
Tellurium structure



Tellurium structure

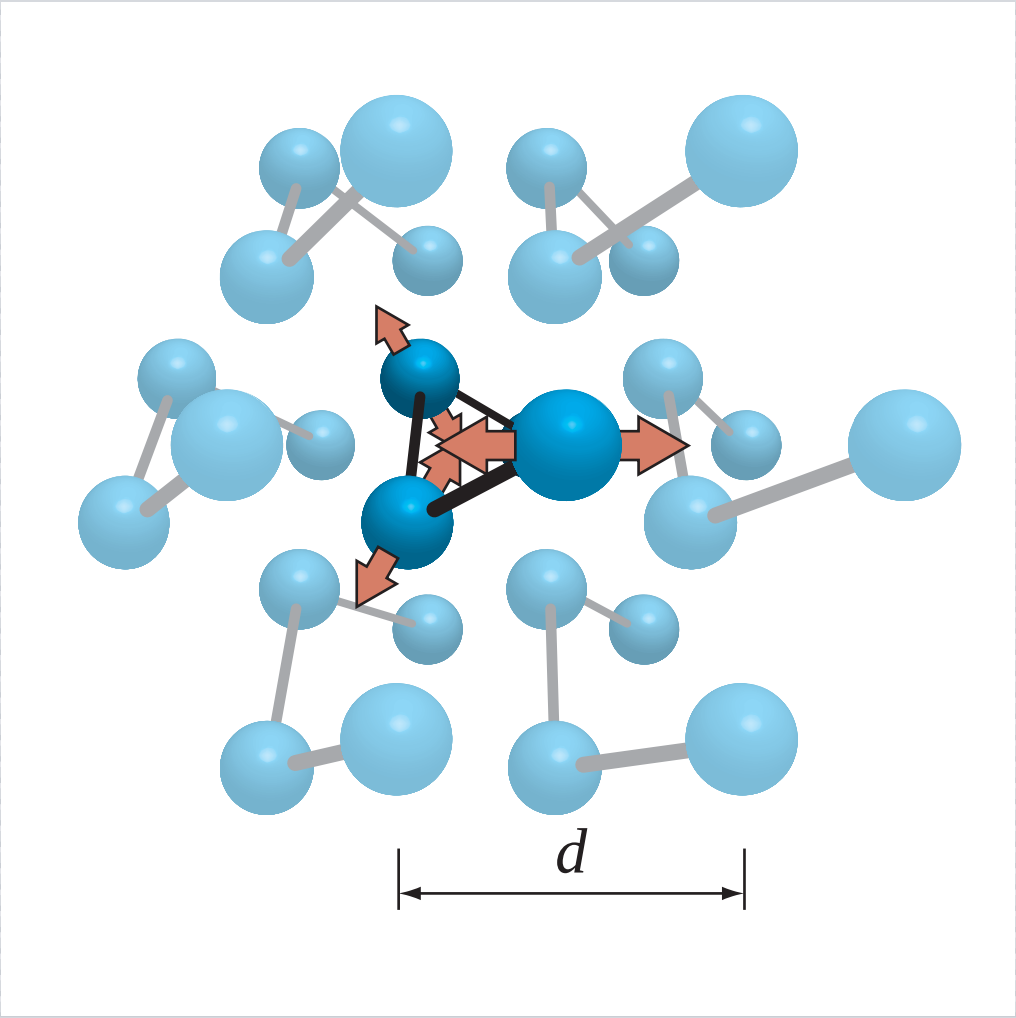


Tellurium structure



helical radius $x = 0.26d$

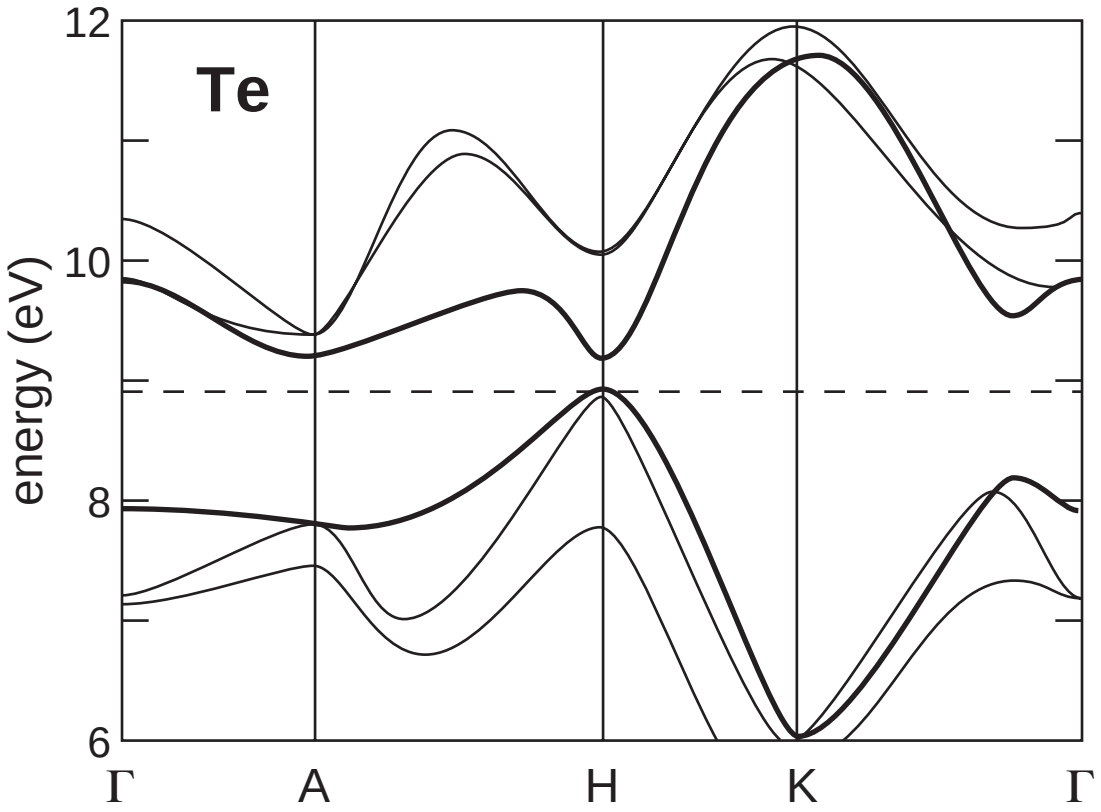
Tellurium structure



A_1 mode modulates *x*

Tellurium band structure

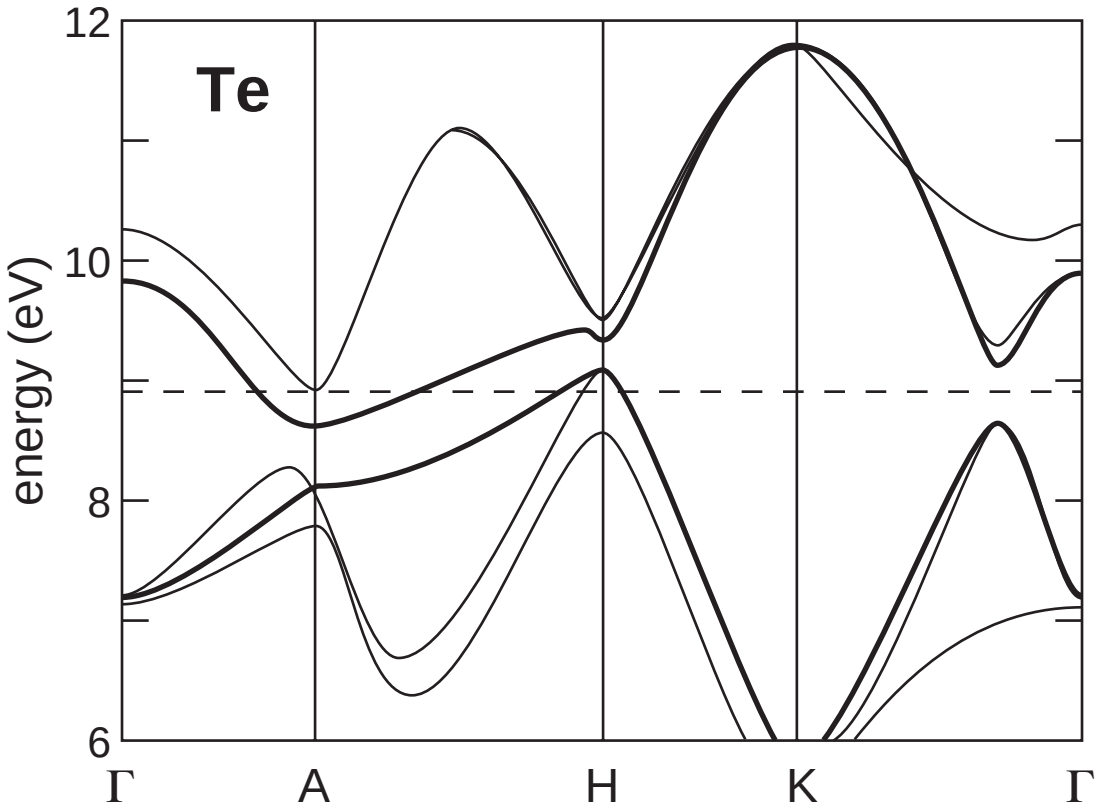
band structure very sensitive to *x*



P. Tangney (Princeton) and S. Fahey (Cork), *private communication*

Tellurium band structure

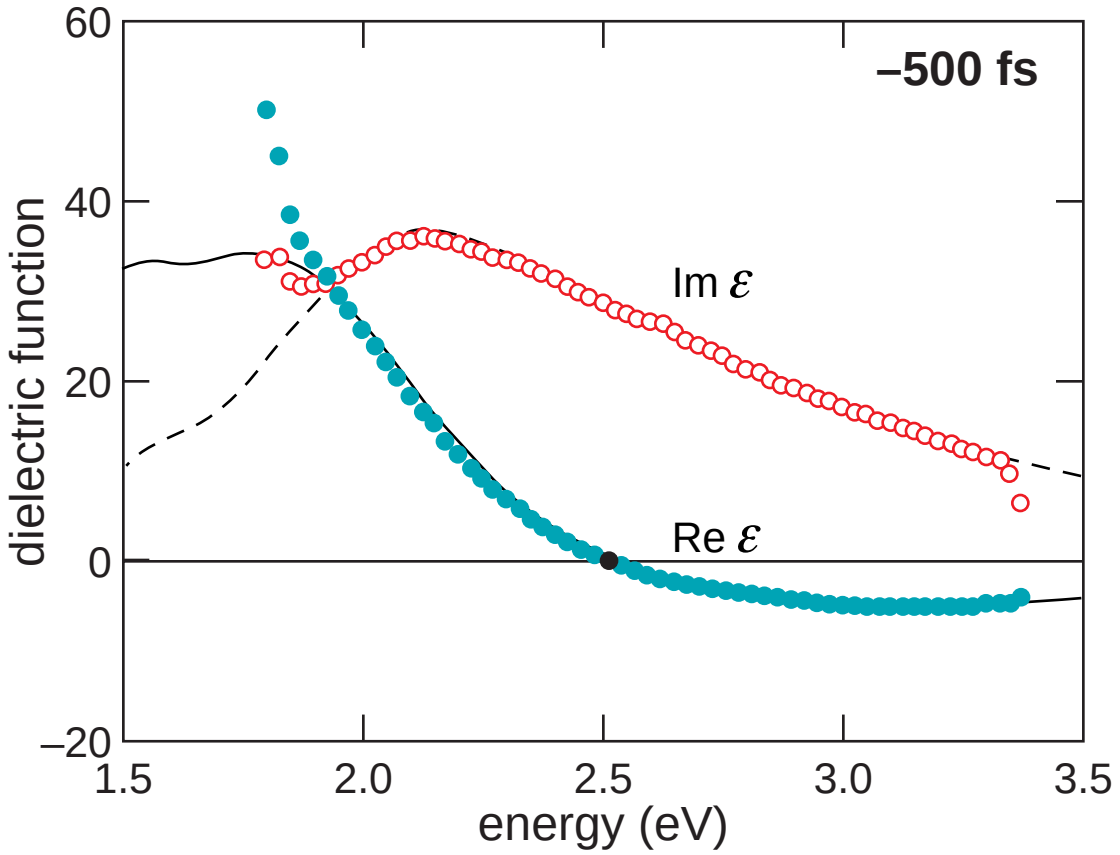
bands cross when *x* changes by 6%



P. Tangney (Princeton) and S. Fahey (Cork), *private communication*

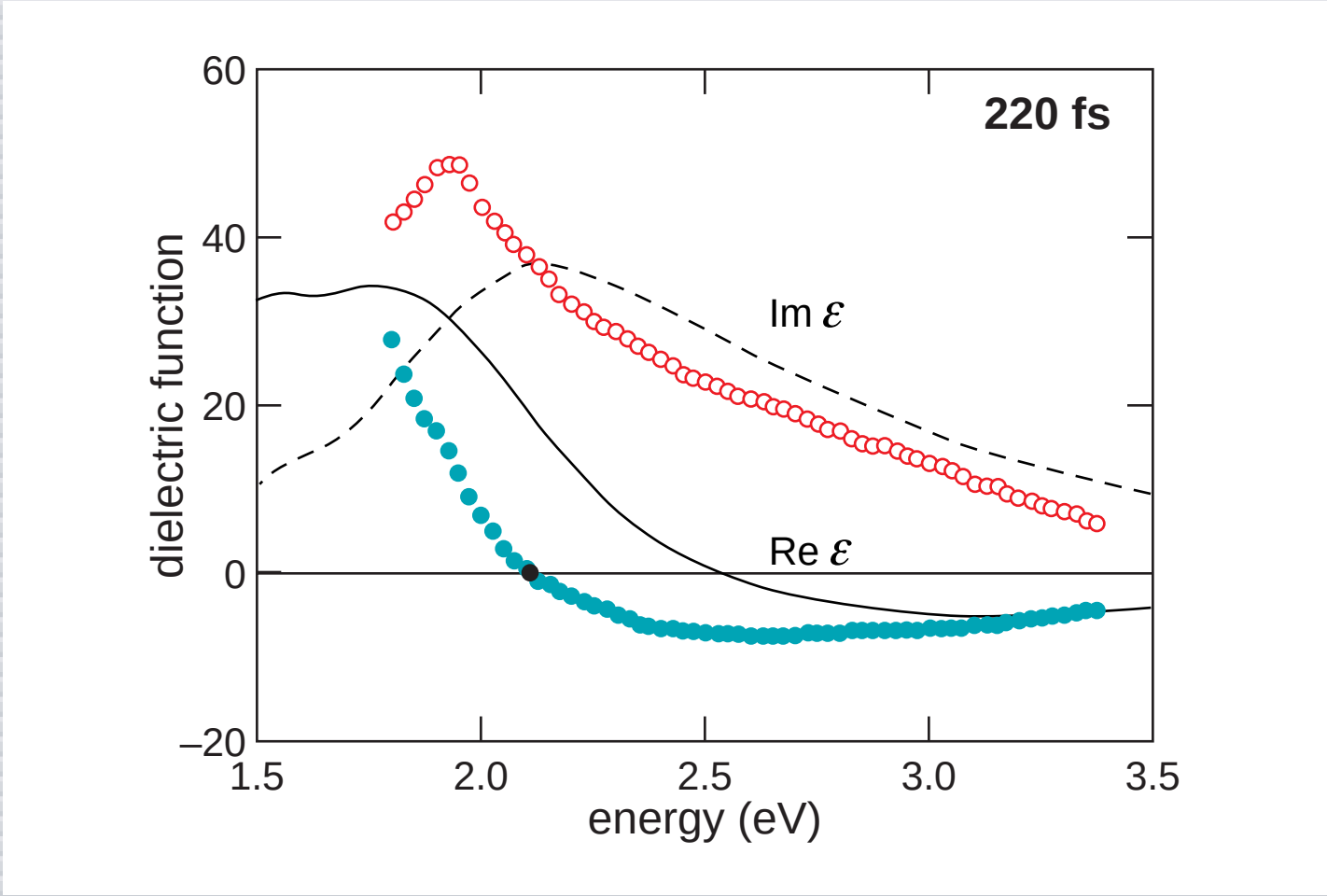
Discussion

track zero-crossing of real part



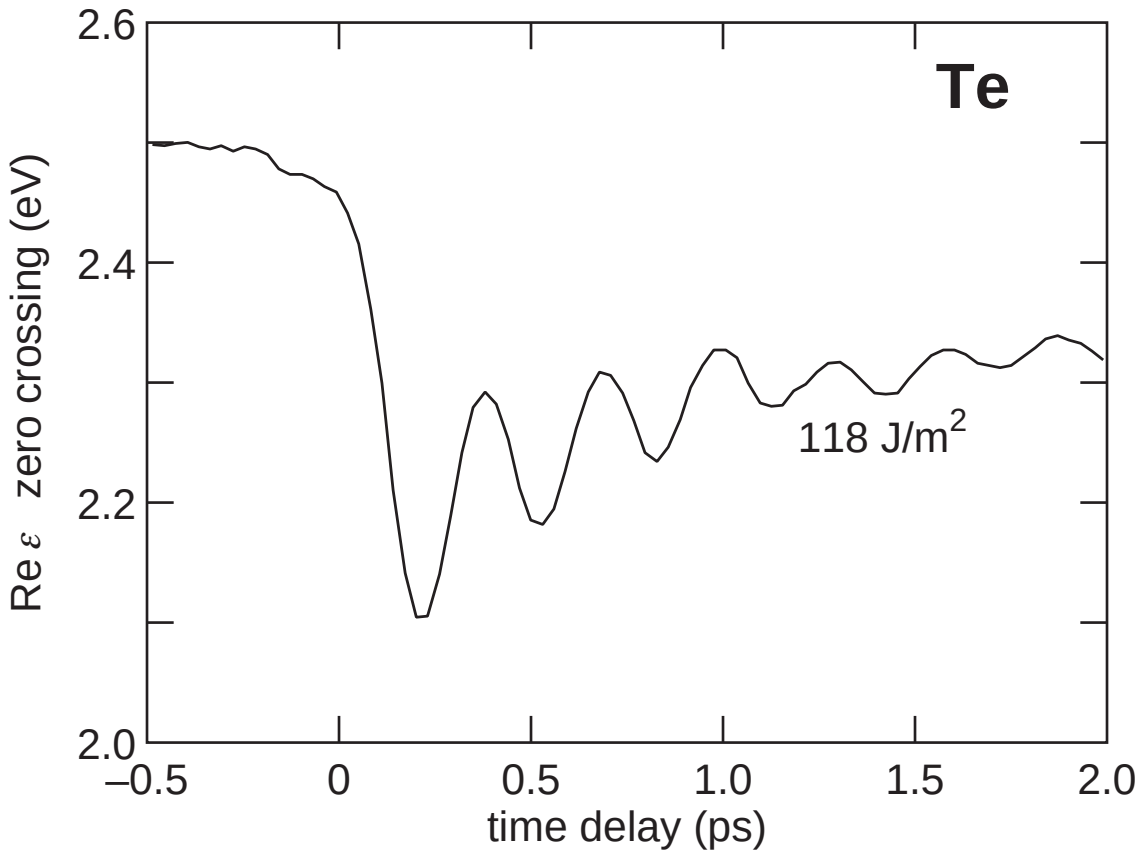
Discussion

track zero-crossing of real part



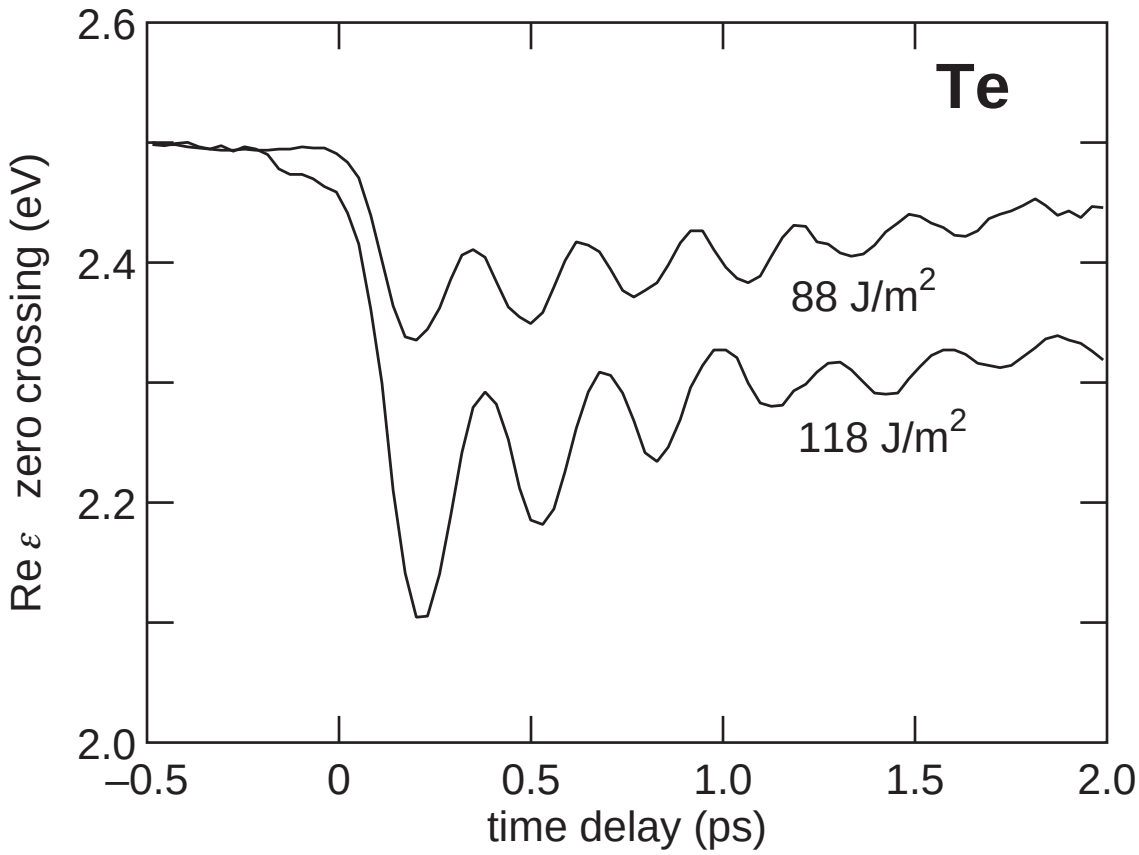
Discussion

track zero-crossing of real part



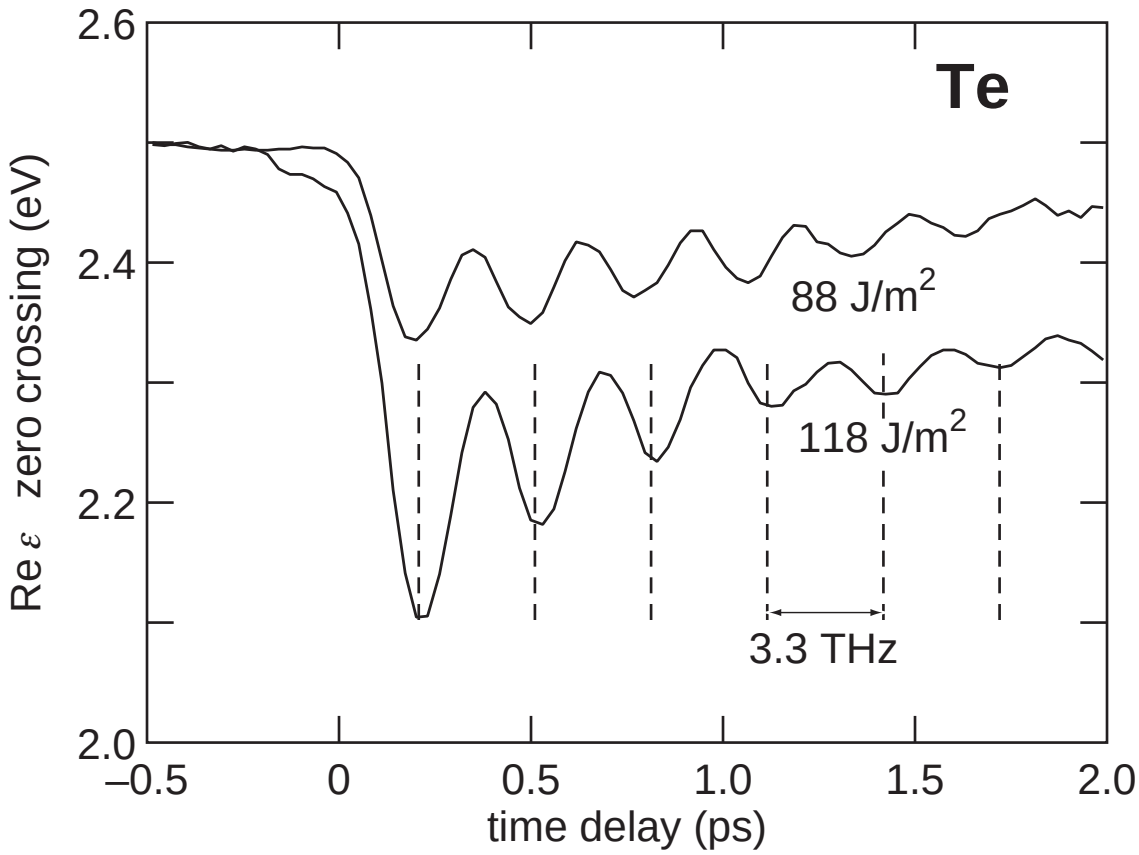
Discussion

higher fluence: larger amplitude oscillations



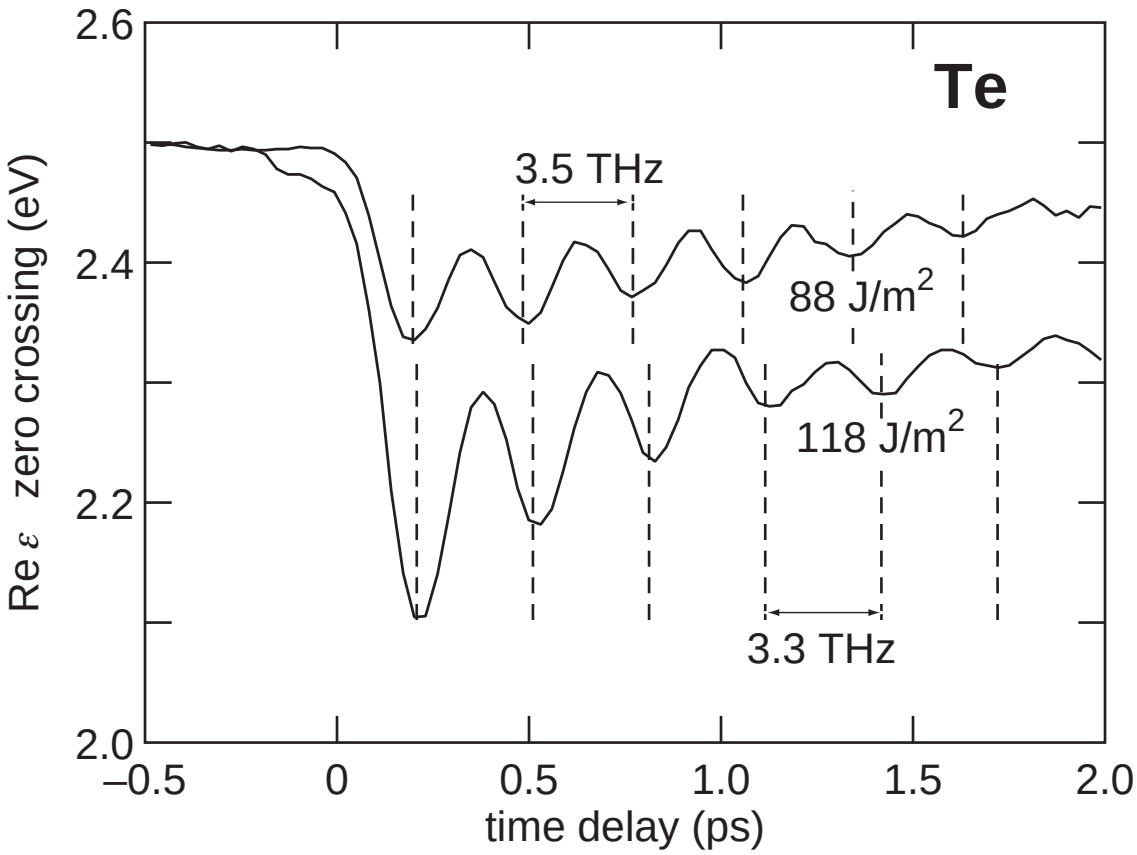
Discussion

frequency less than 3.6 THz equilibrium value



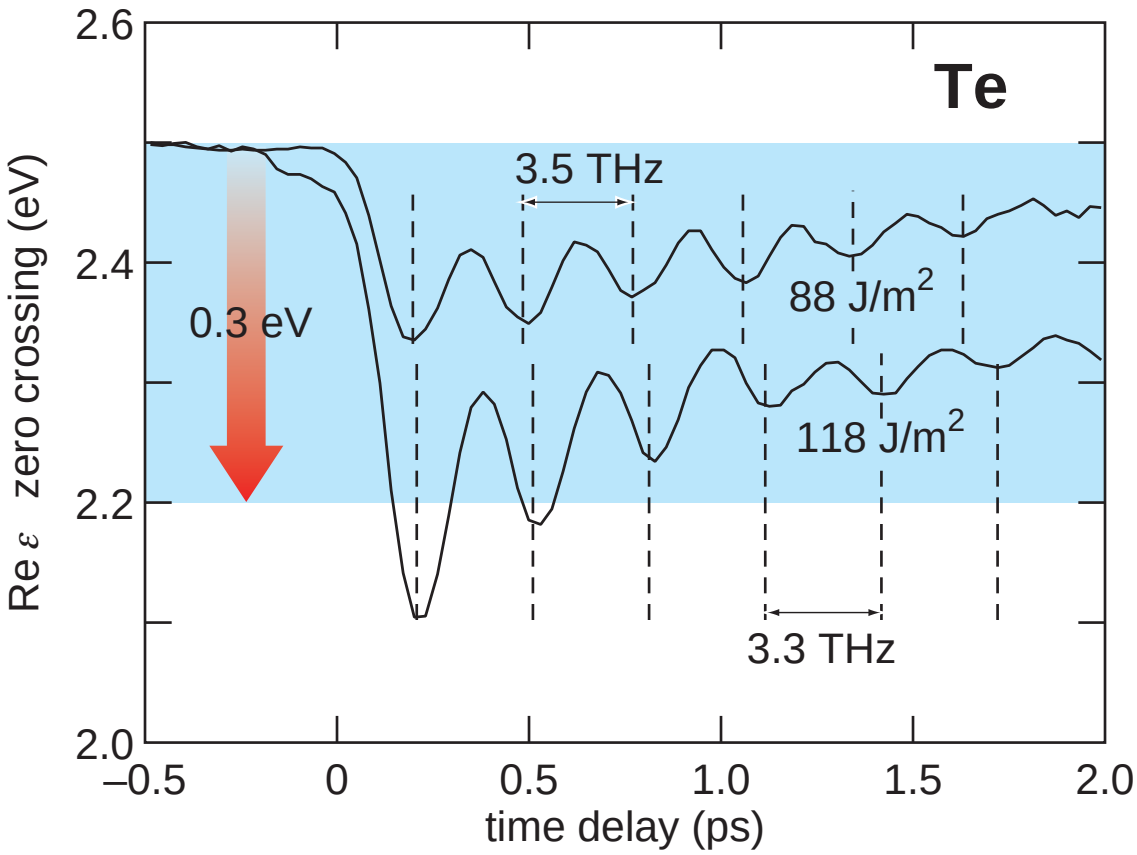
Discussion

softening of phonon mode



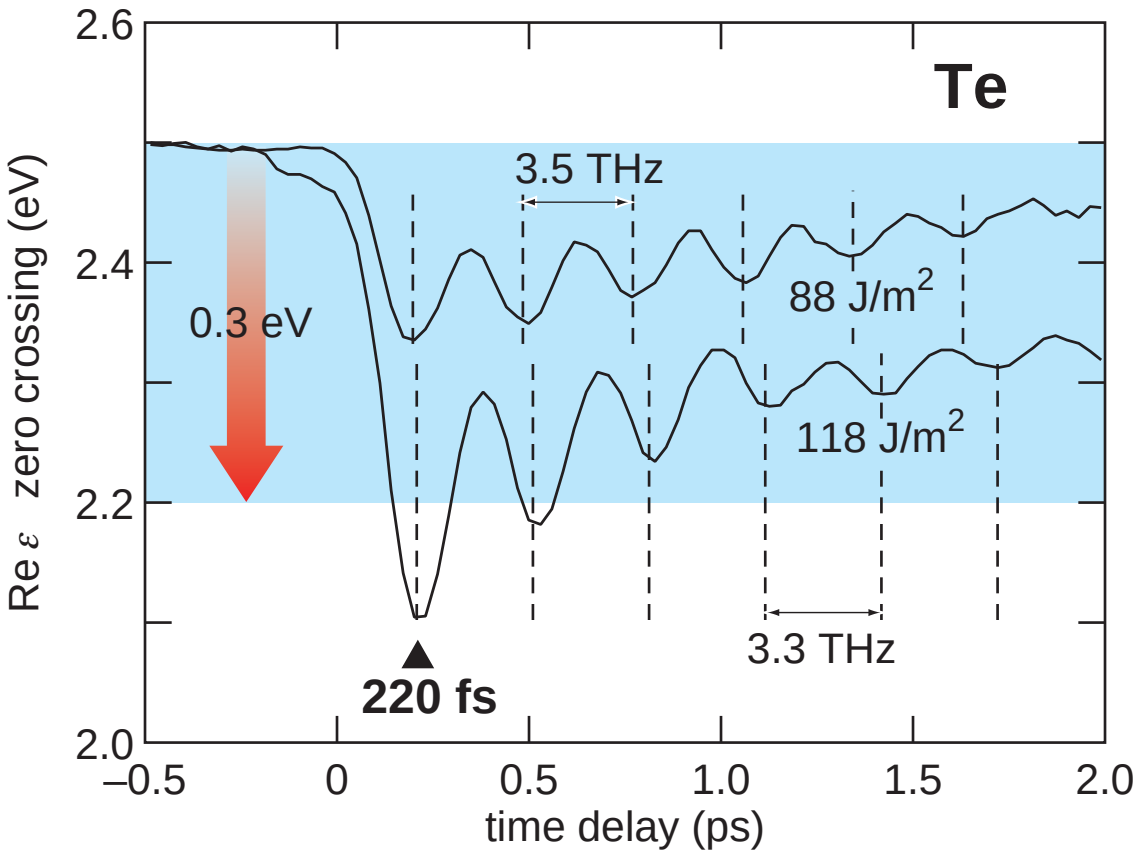
Discussion

compare shift to band gap



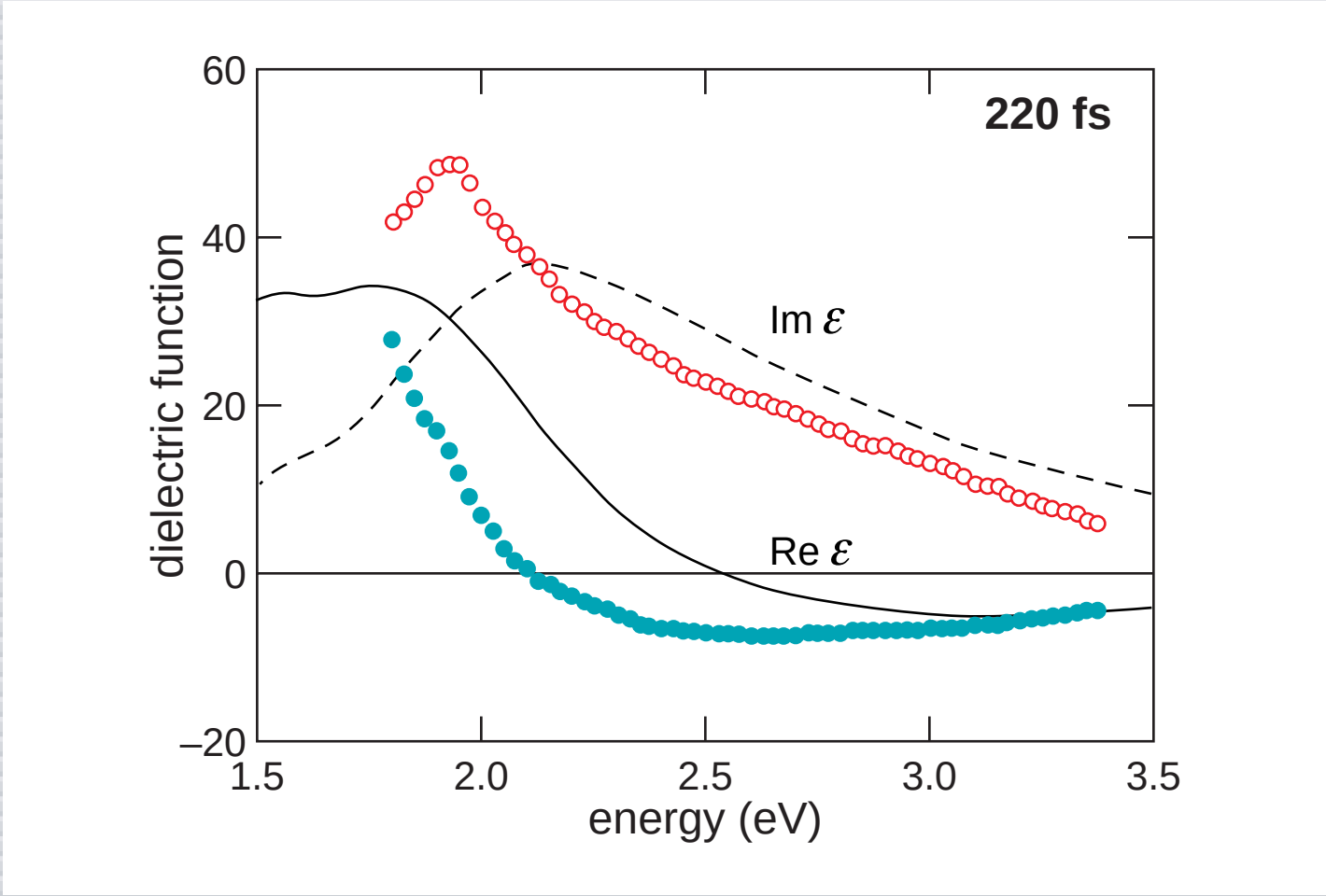
Discussion

shift exceeds band gap at 220 fs…

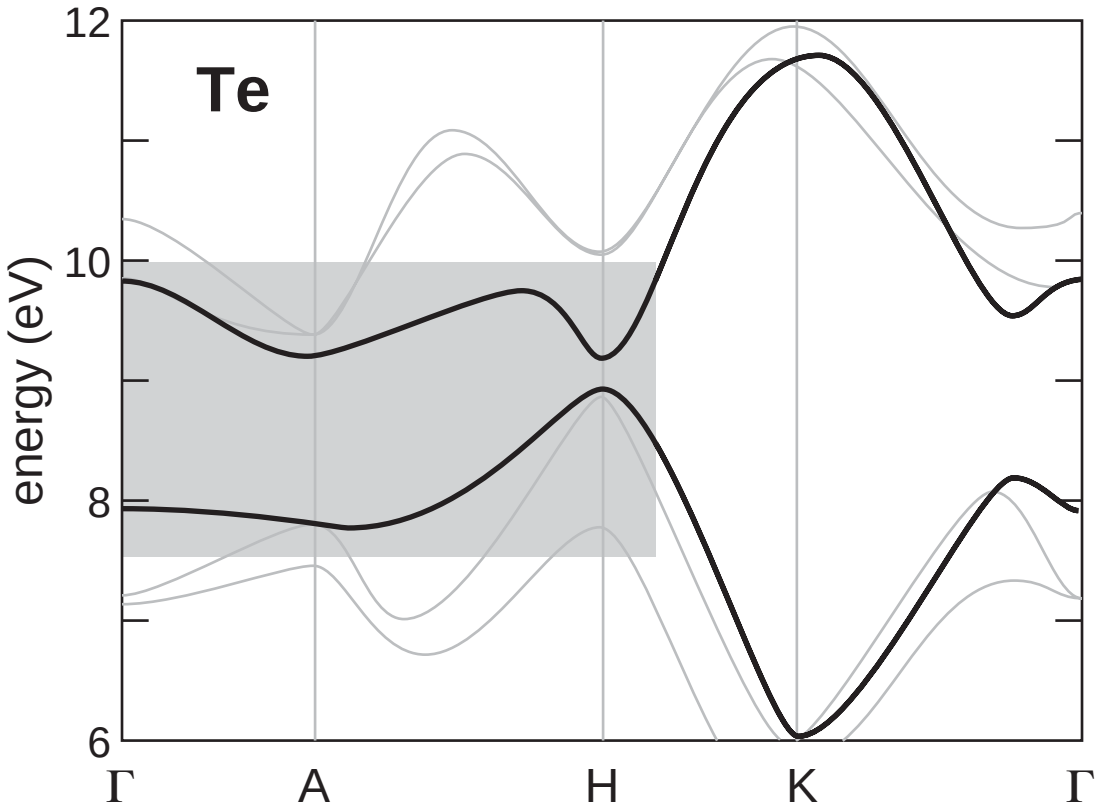


Discussion

… but dielectric function non-metallic!

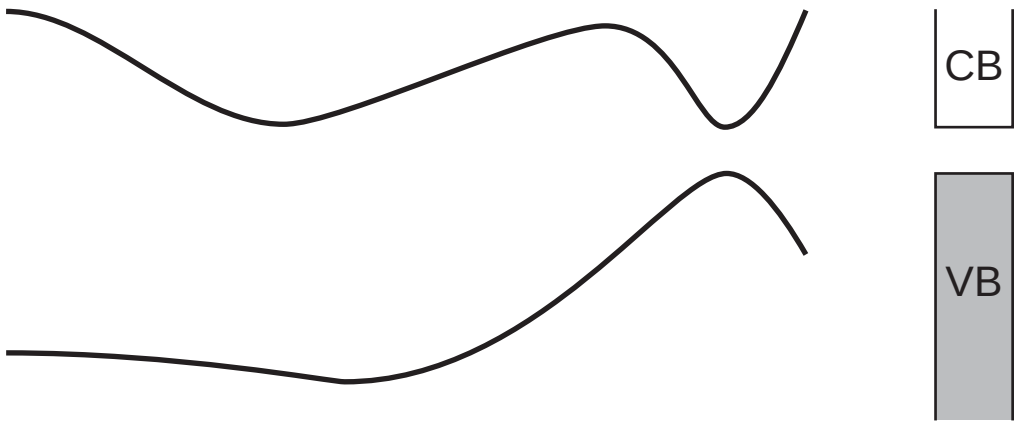


Discussion



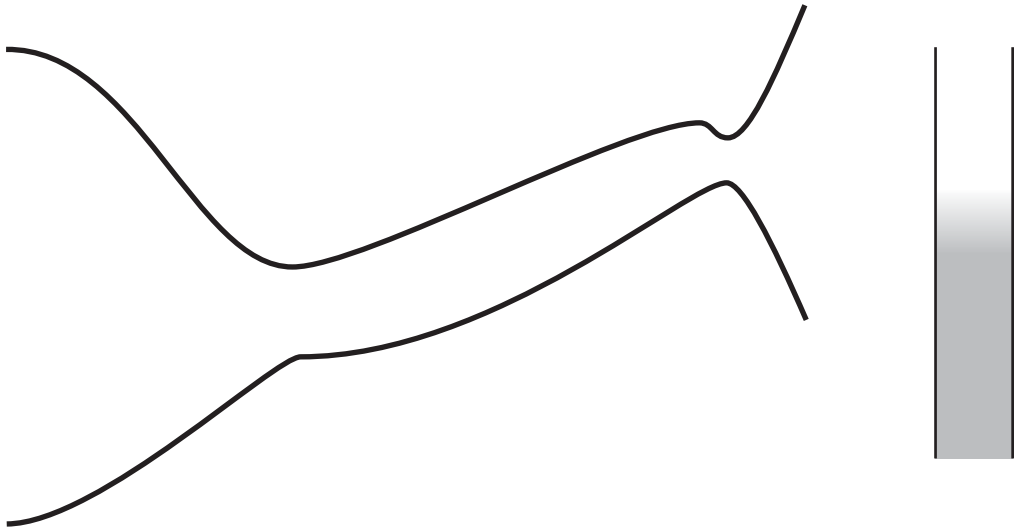
Discussion

semiconducting because of 0.3 eV gap



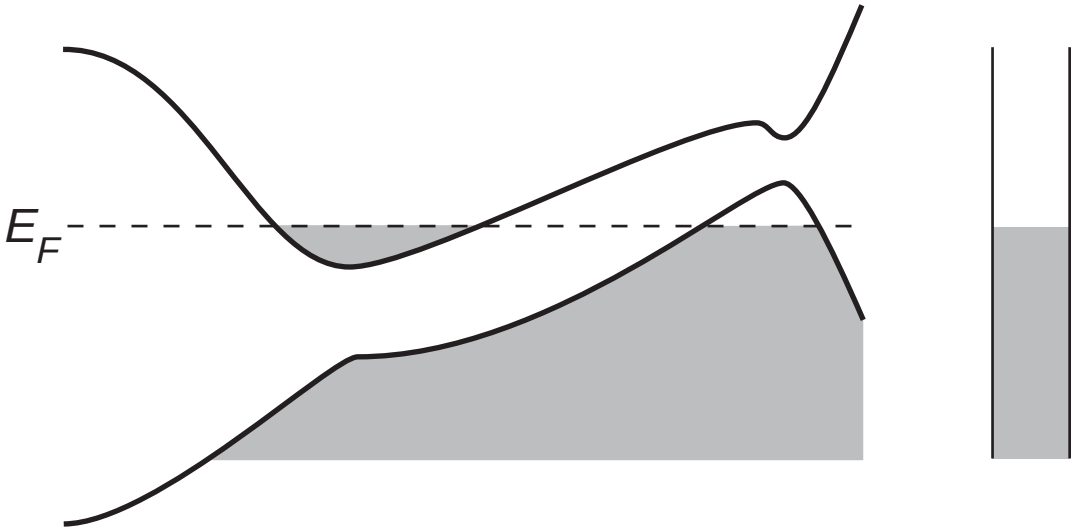
Discussion

after bands cross…



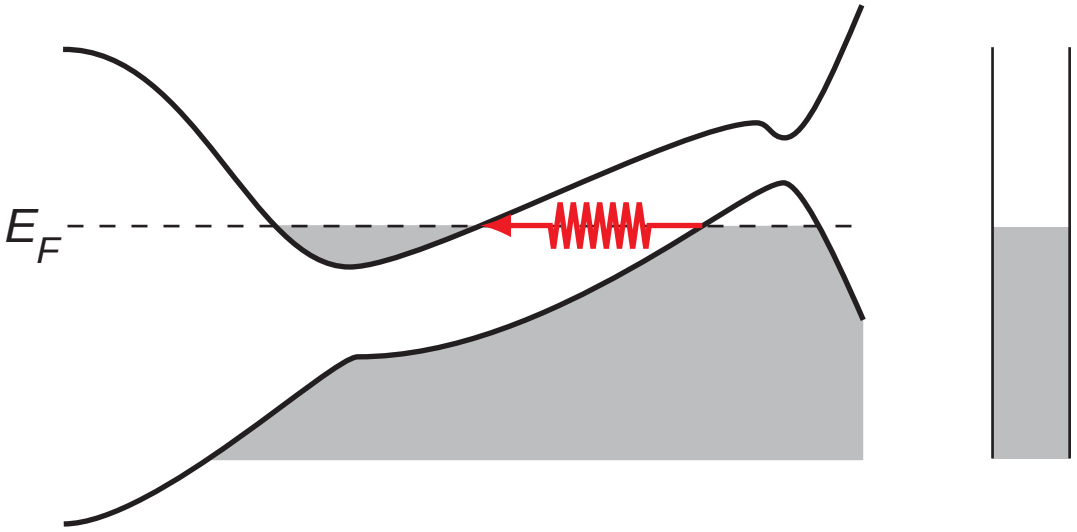
Discussion

… tellurium can become metallic…



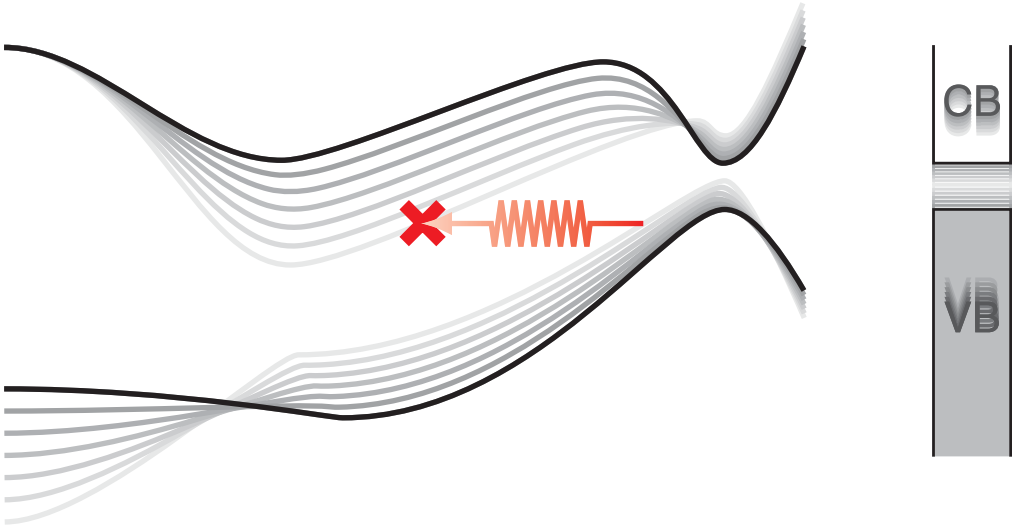
Discussion

… provided phonons scatter electrons



Discussion

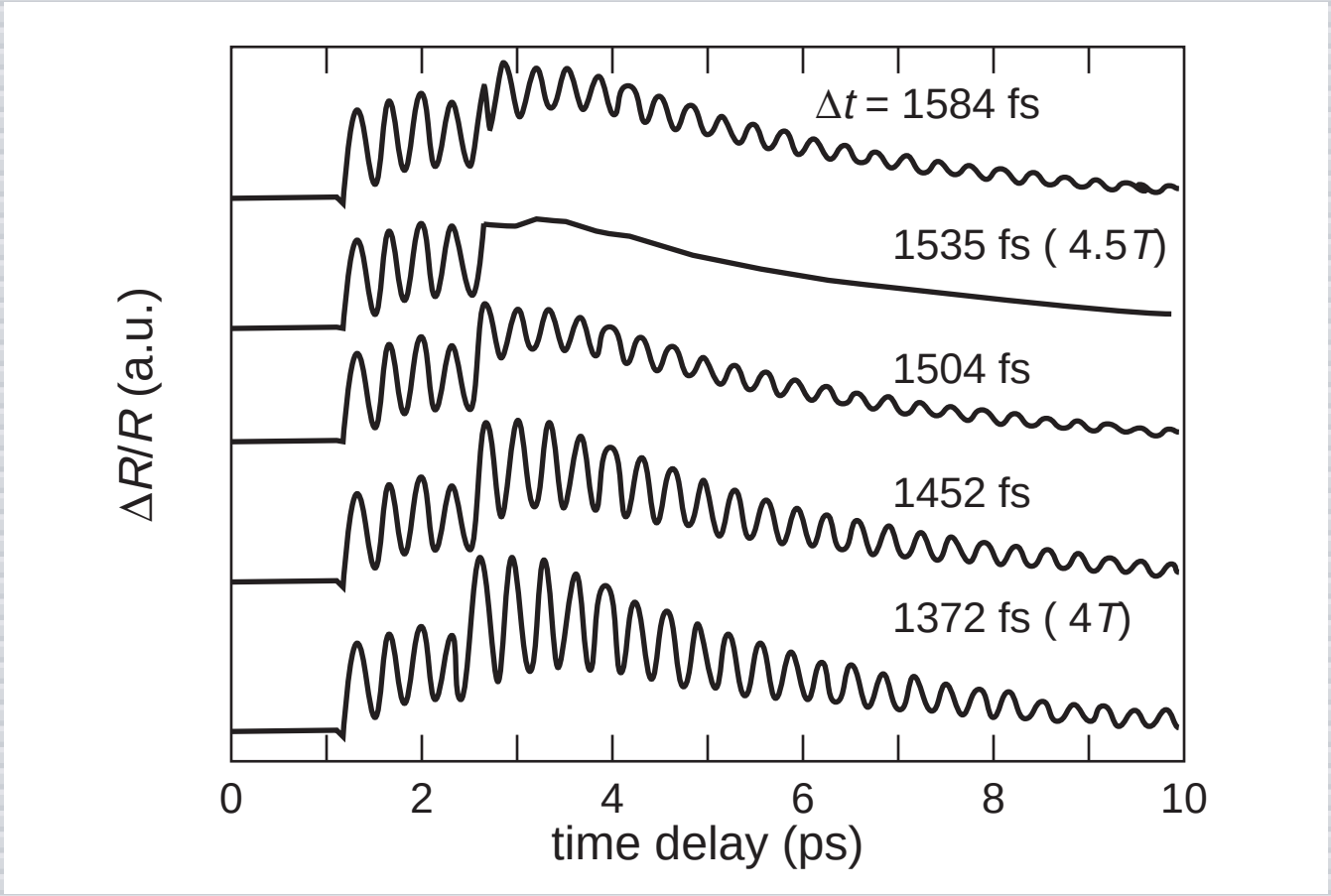
if $\tau_{scatter} > T_{phonon}$, 'frustrated' metal



What's next?

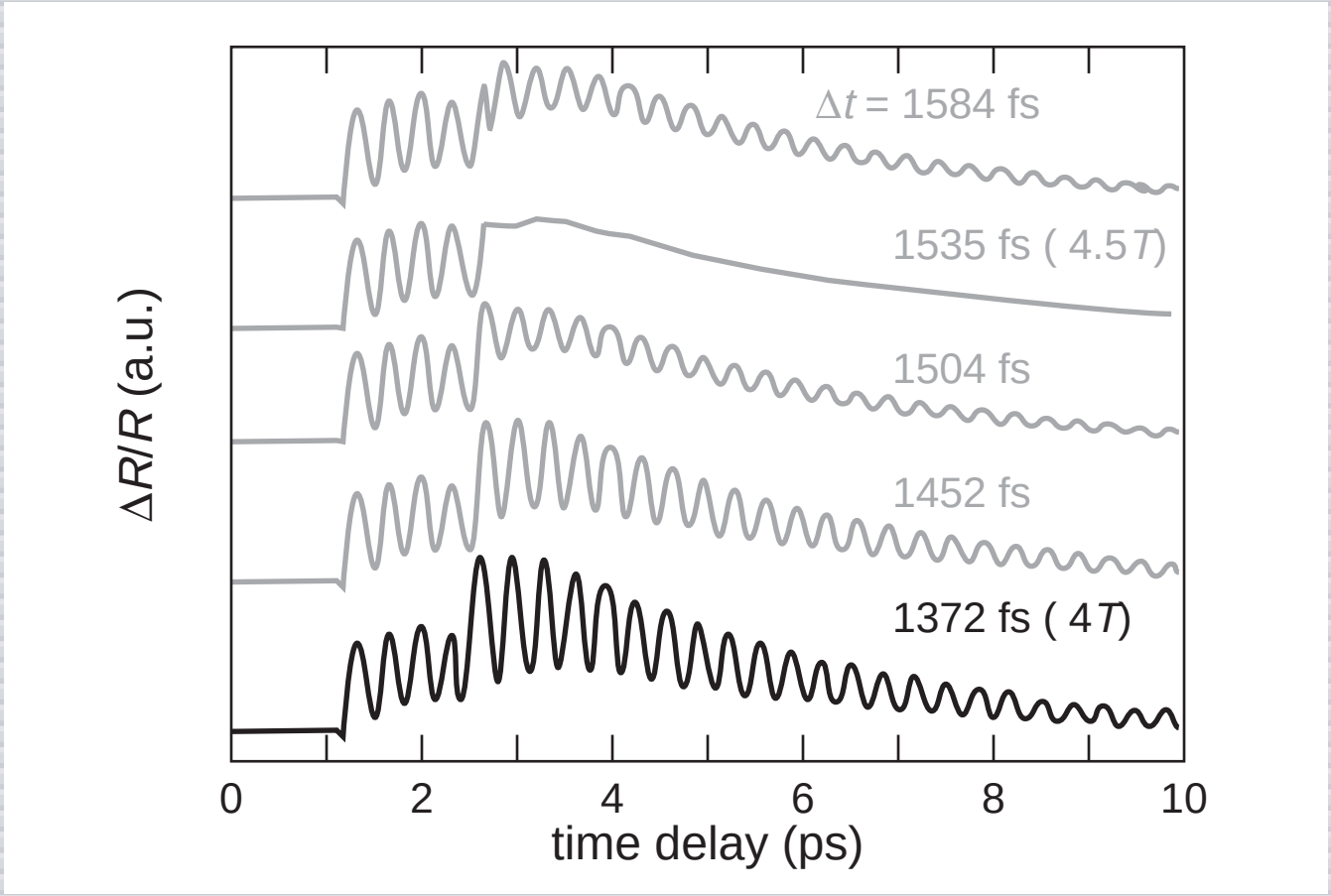
- larger amplitude phonons
- different materials
- density functional theory modeling
- multiple pulse excitation for coherent control

What's next?



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

What's next?



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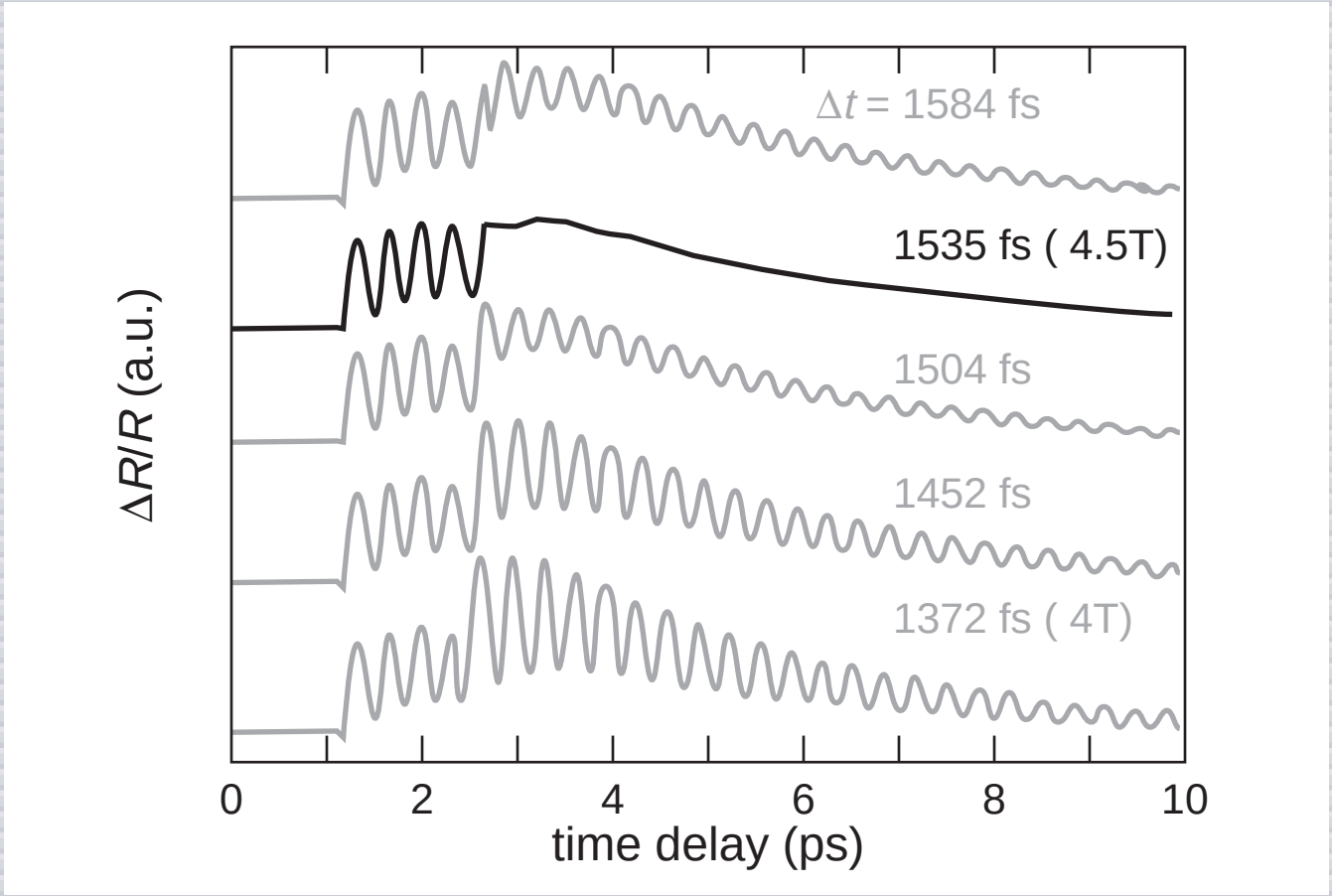
What's next?



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

Summary

- **femtosecond ellipsometry:**
  observe transitions *as they occur*

Summary

- **femtosecond ellipsometry: observe transitions *as they occur***
- **dielectric function shows displacive excitation of coherent phonons**

Summary

- **femtosecond ellipsometry: observe transitions *as they occur***
- **dielectric function shows displacive excitation of coherent phonons**
- **no electronic transition in spite of structural change**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APPLIED SCIENCE

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