Ultrafast Lattice-Bonding Dynamics in Tellurium

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Structural transitions driven by fs pulses

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order-disorder
(a.k.a. non-thermal melting)

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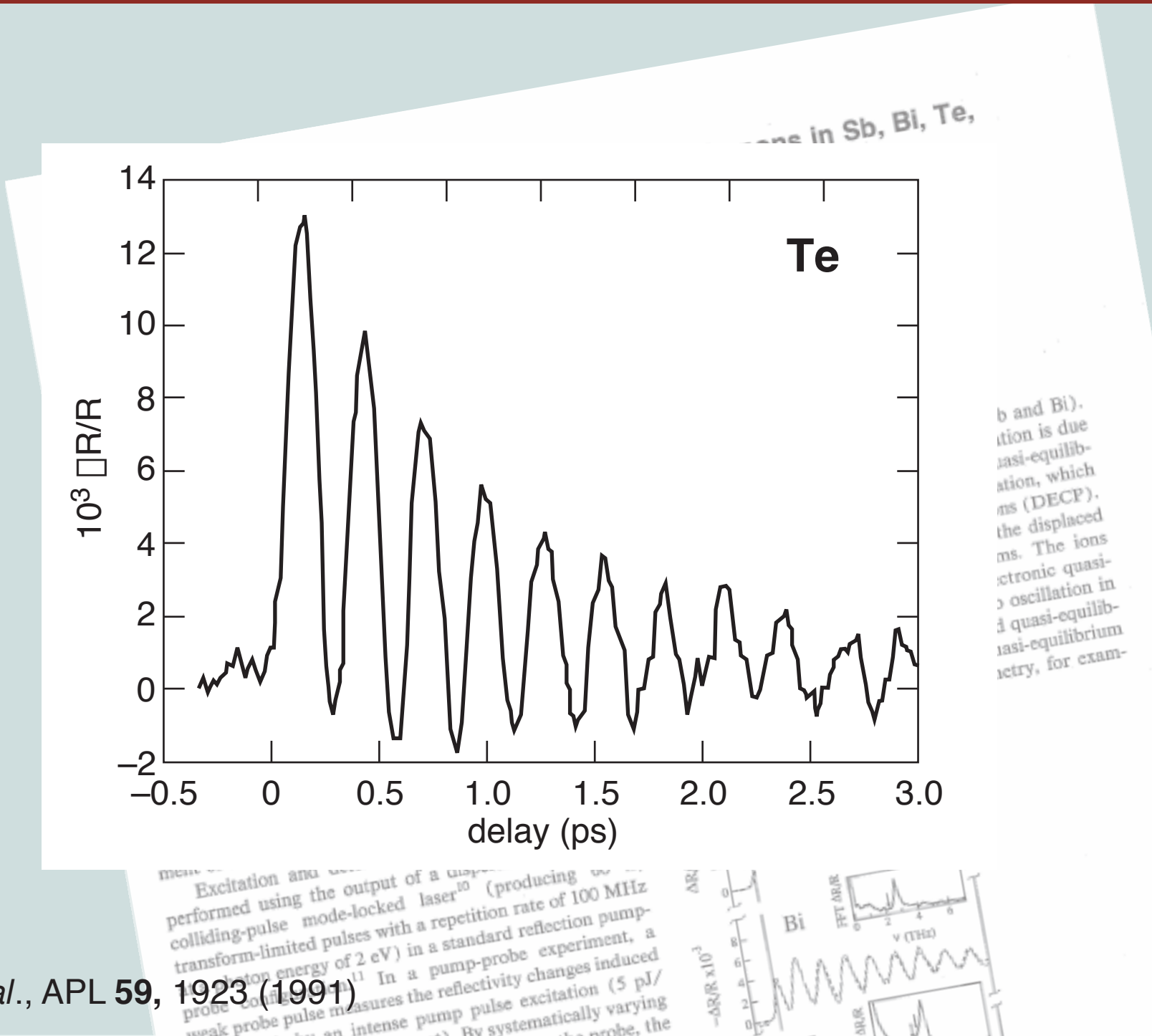
Structural transitions driven by fs pulses

order-disorder
(a.k.a. non-thermal melting)

order-order

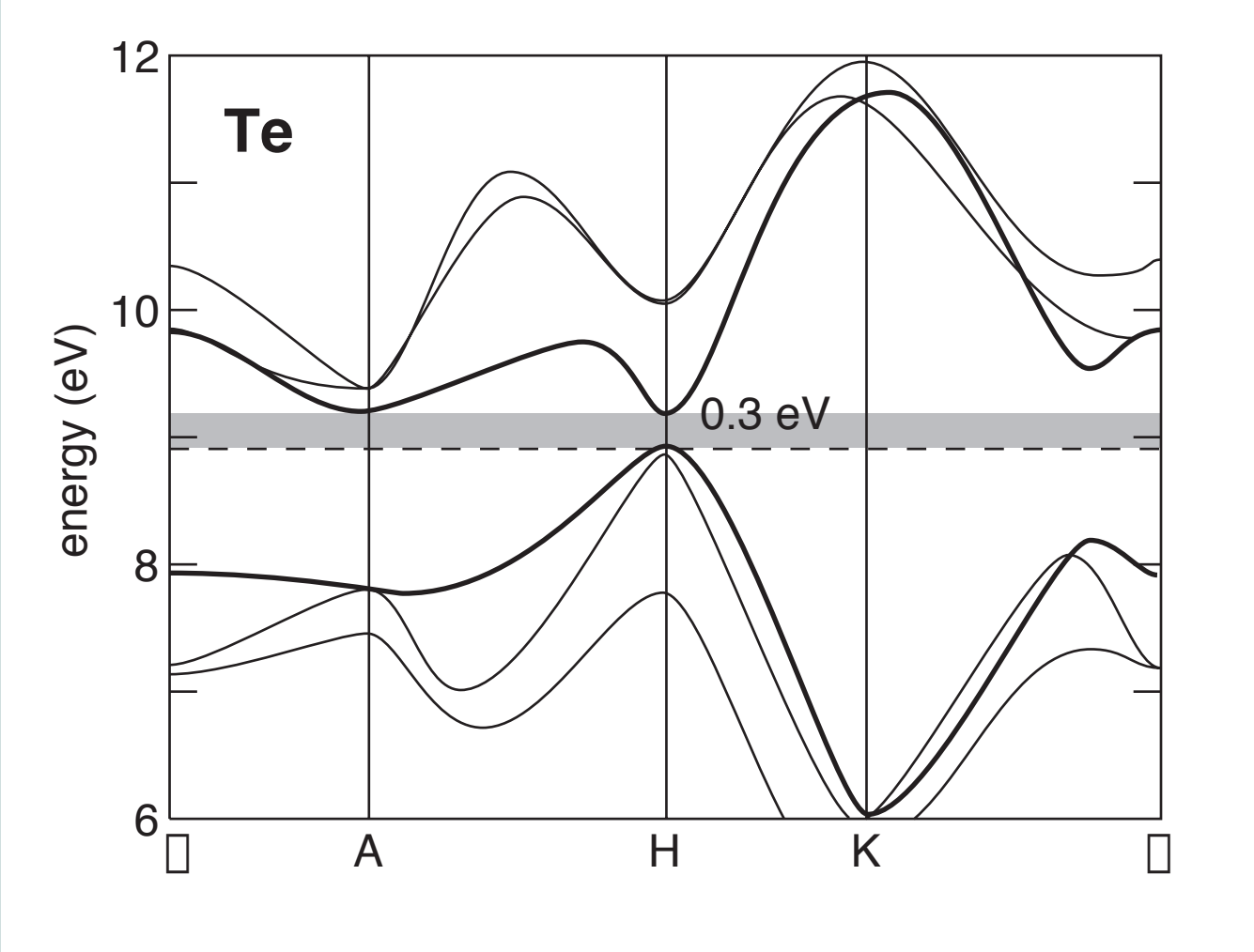
associated electronic transition
(i.e. metal-insulator)

INTRODUCTION



Cheng *et al*., APL **59,** 1923 (1991)

Te is close to band-crossing transition



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

OUTLINE

Experimental technique

time-resolved dielectric function

Results

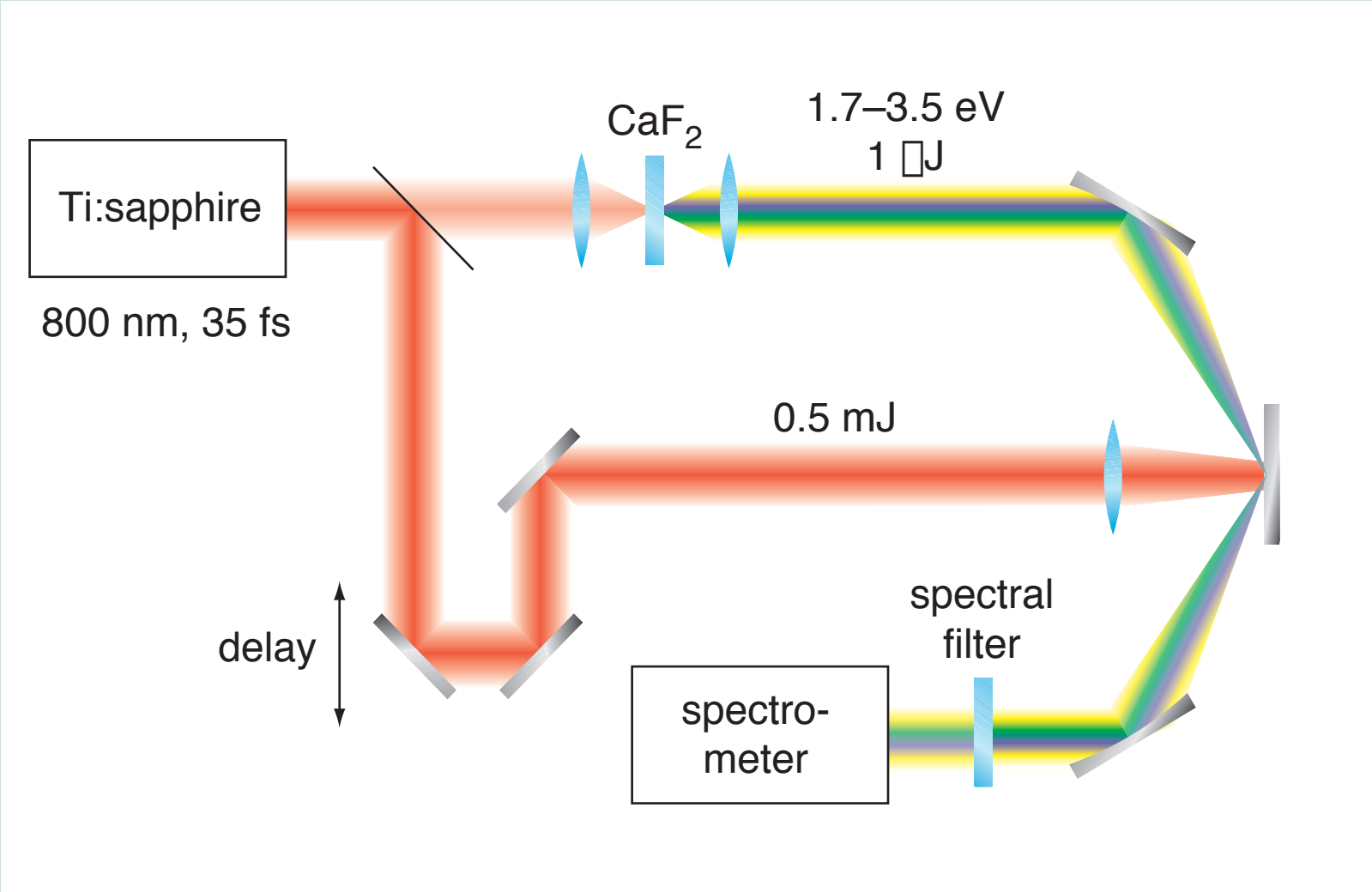
effects of coherent phonons on $\epsilon(\omega)$

Discussion

two-atom model

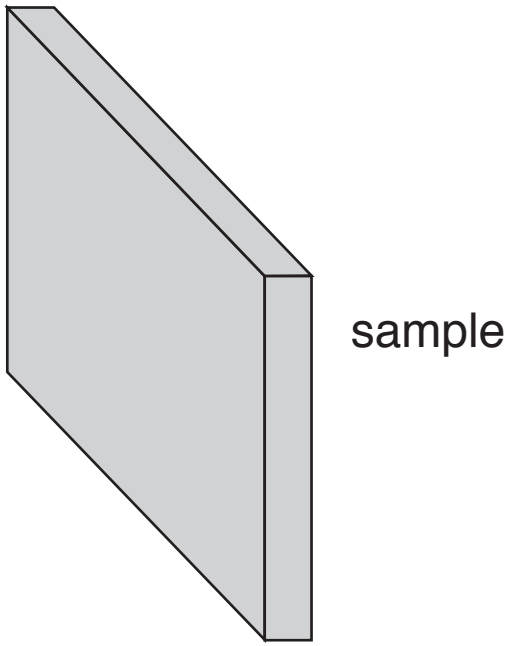
DFT calculations

Time-resolved ellipsometry

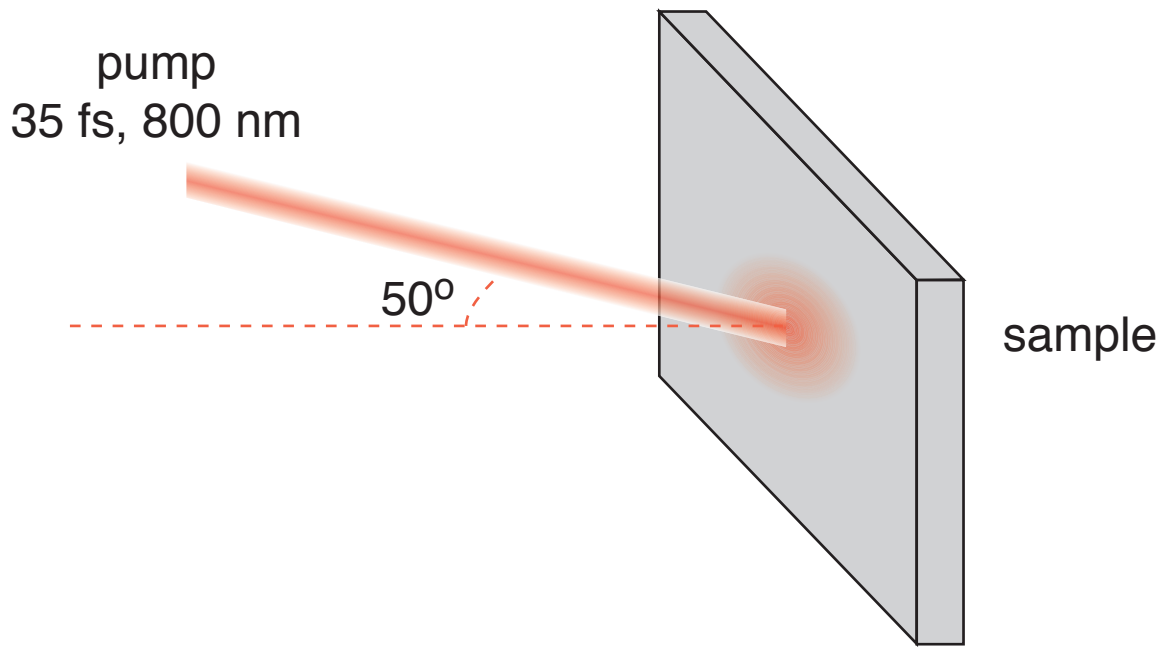


White-light pump-probe setup

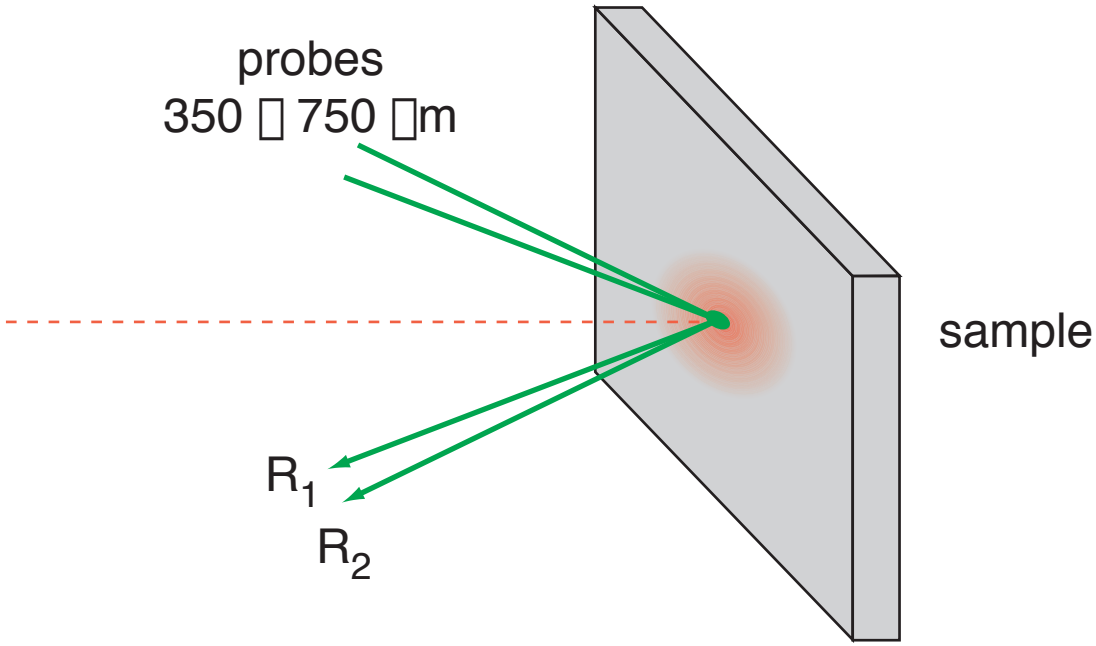
Time-resolved ellipsometry



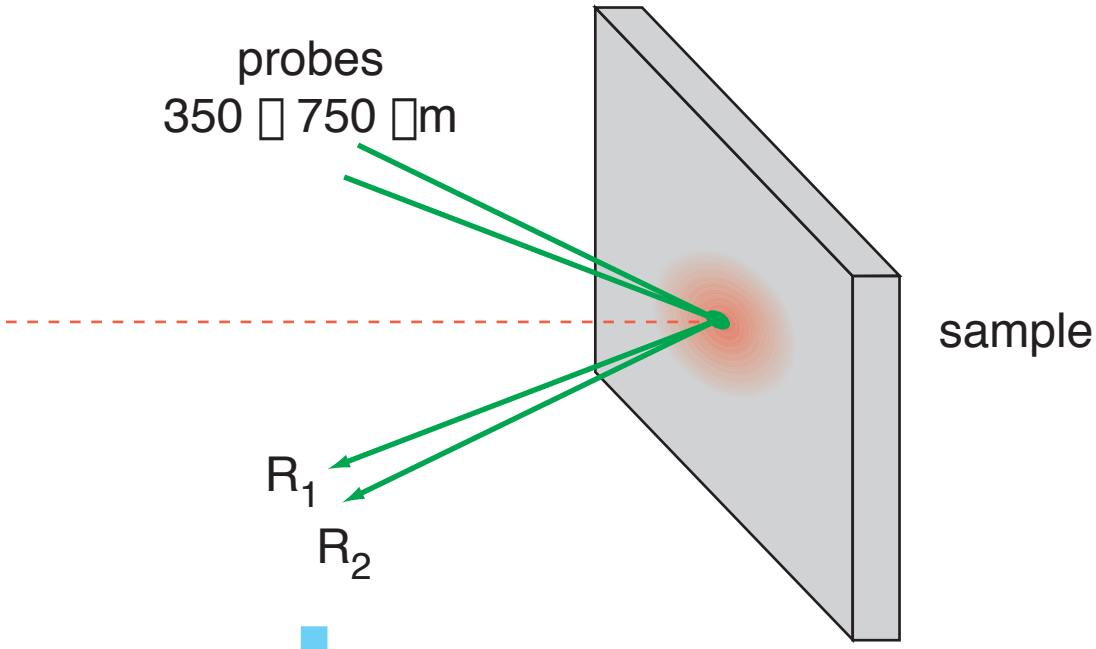
Time-resolved ellipsometry



Time-resolved ellipsometry

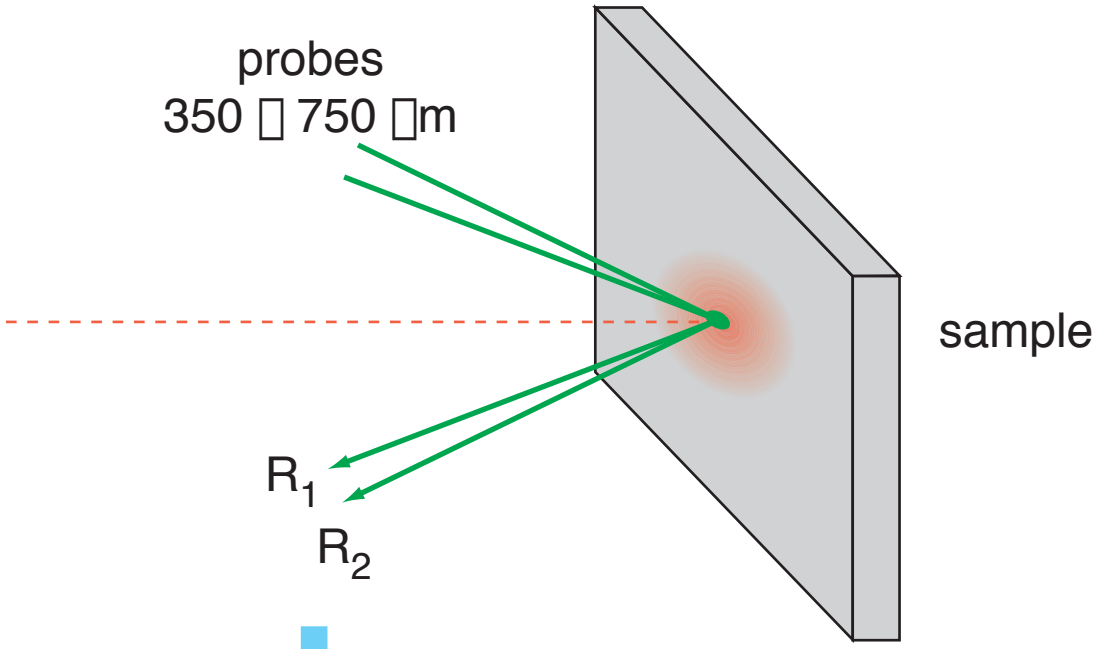


Time-resolved ellipsometry



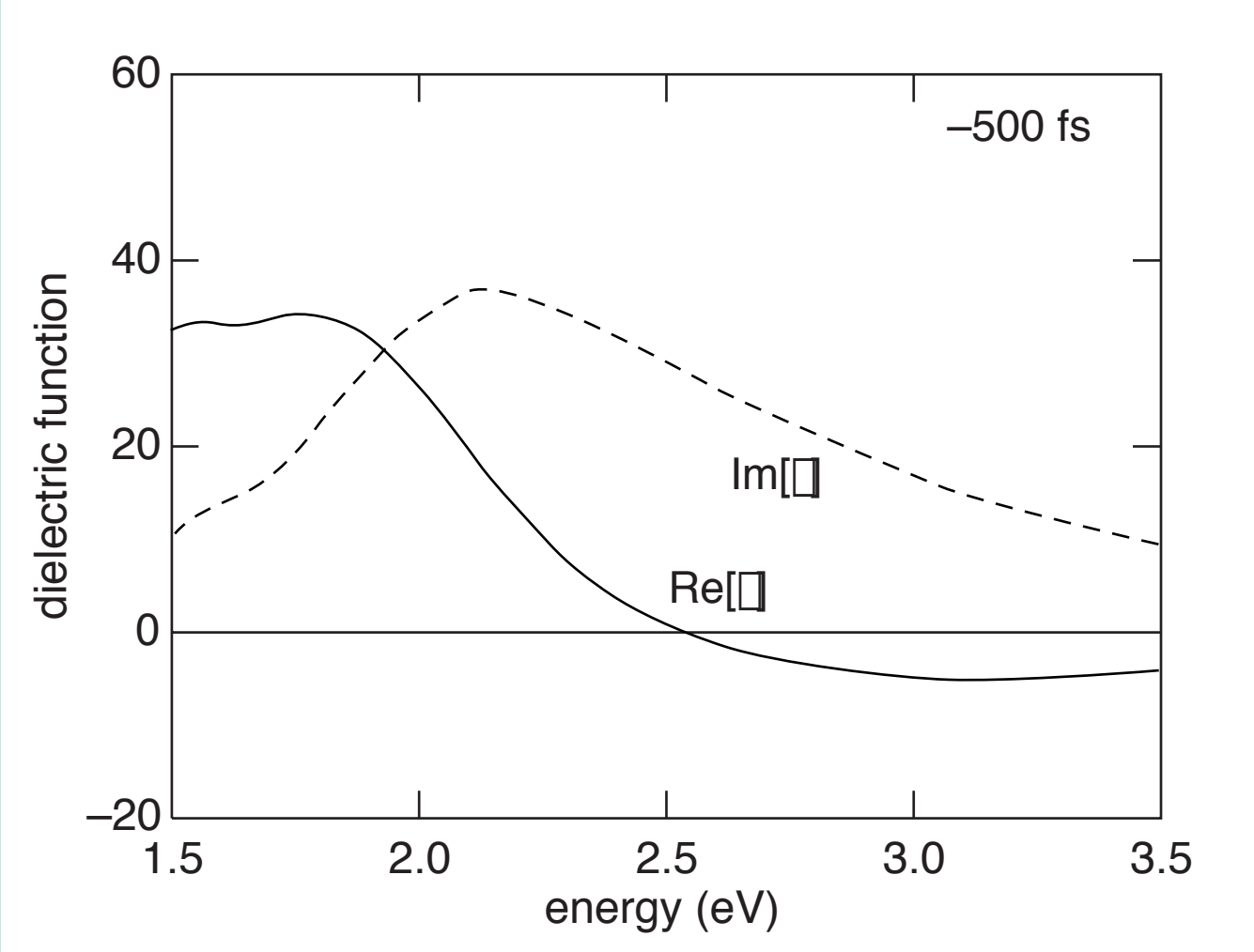
Fresnel equations

Time-resolved ellipsometry

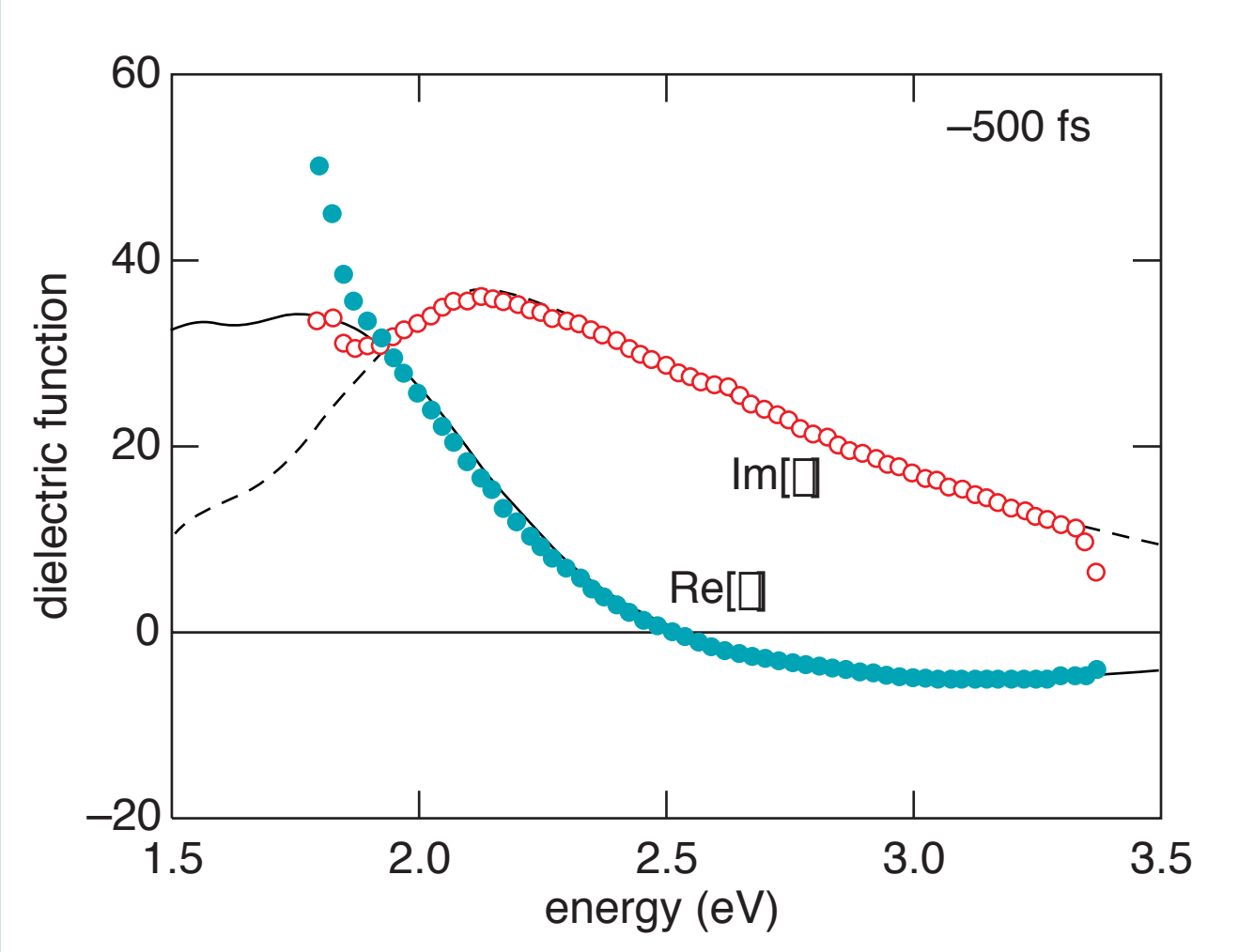


Fresnel equations → Re[□(□)] Im[□(□)]

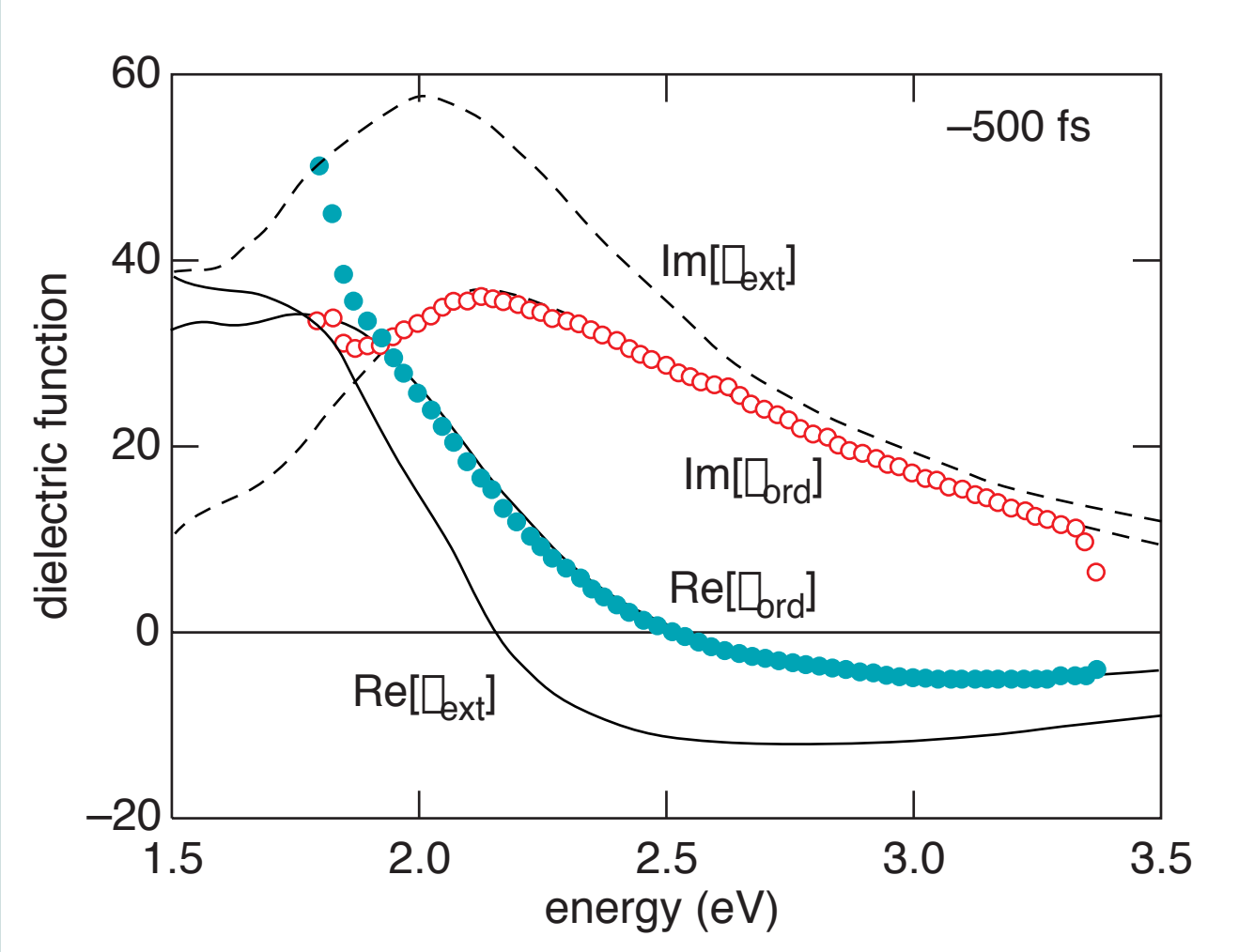
RESULTS


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

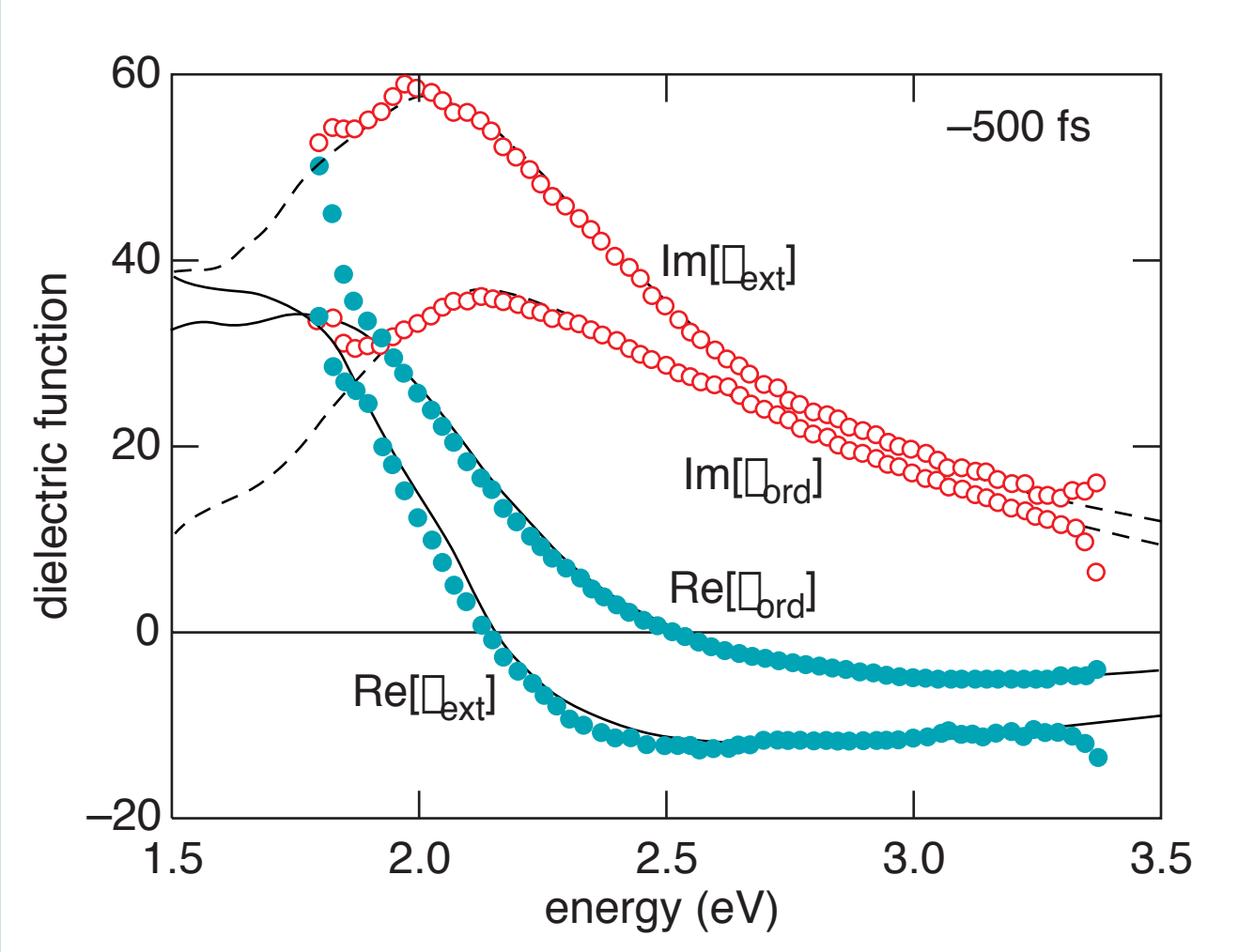
RESULTS


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

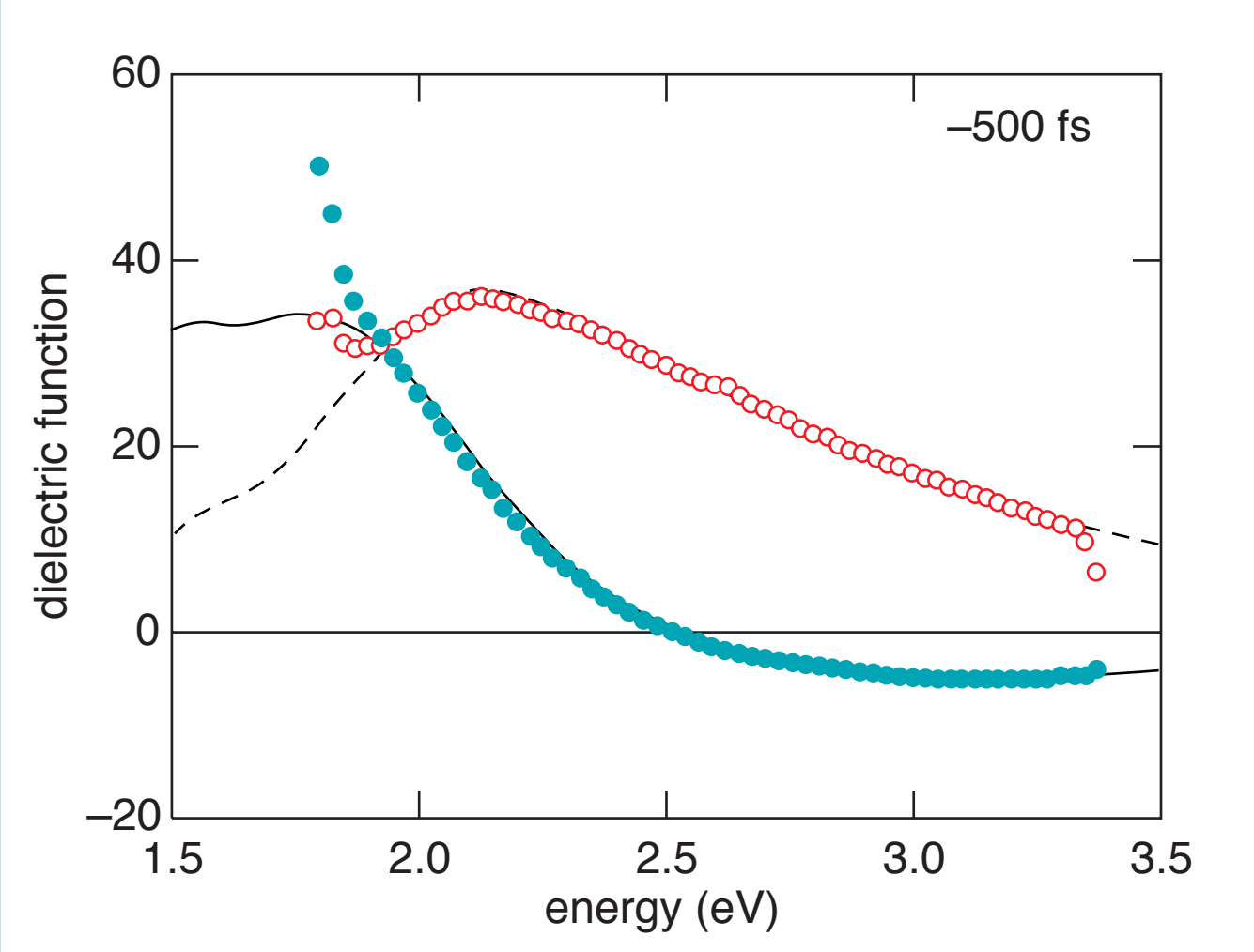
RESULTS


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

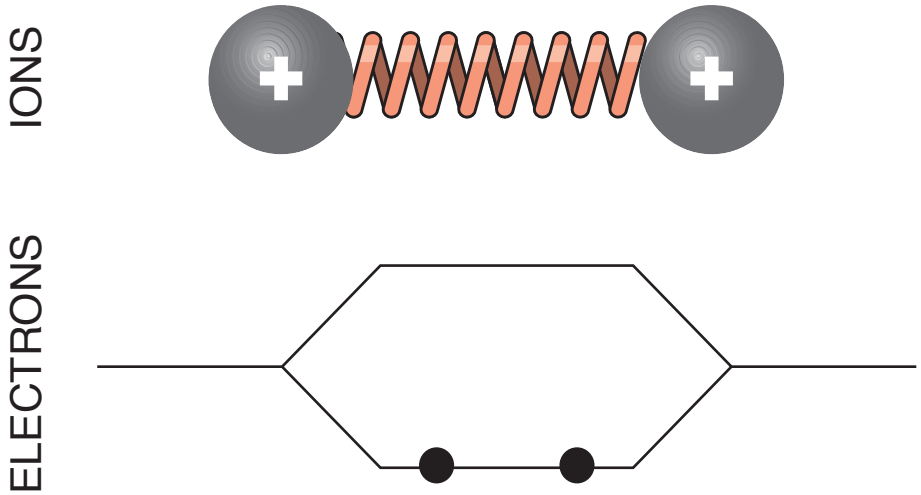
RESULTS


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

RESULTS


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

"Two-atom" model



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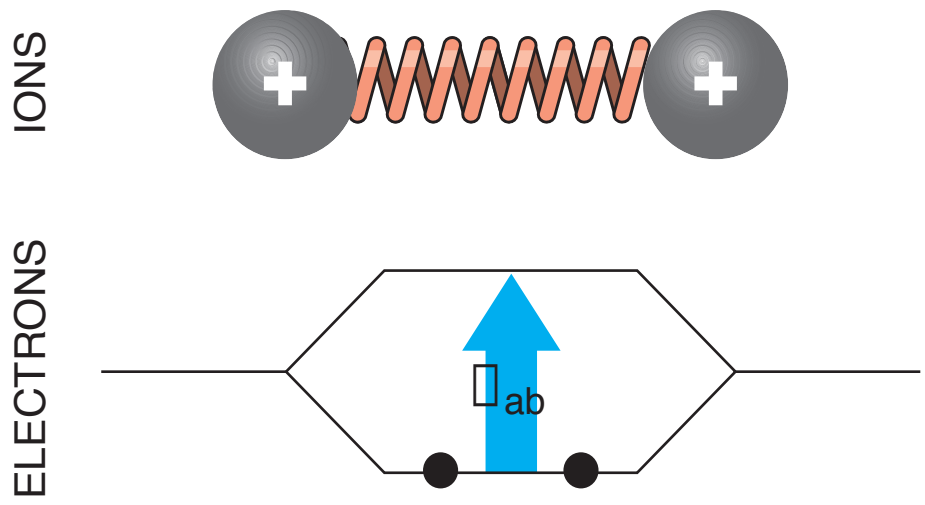
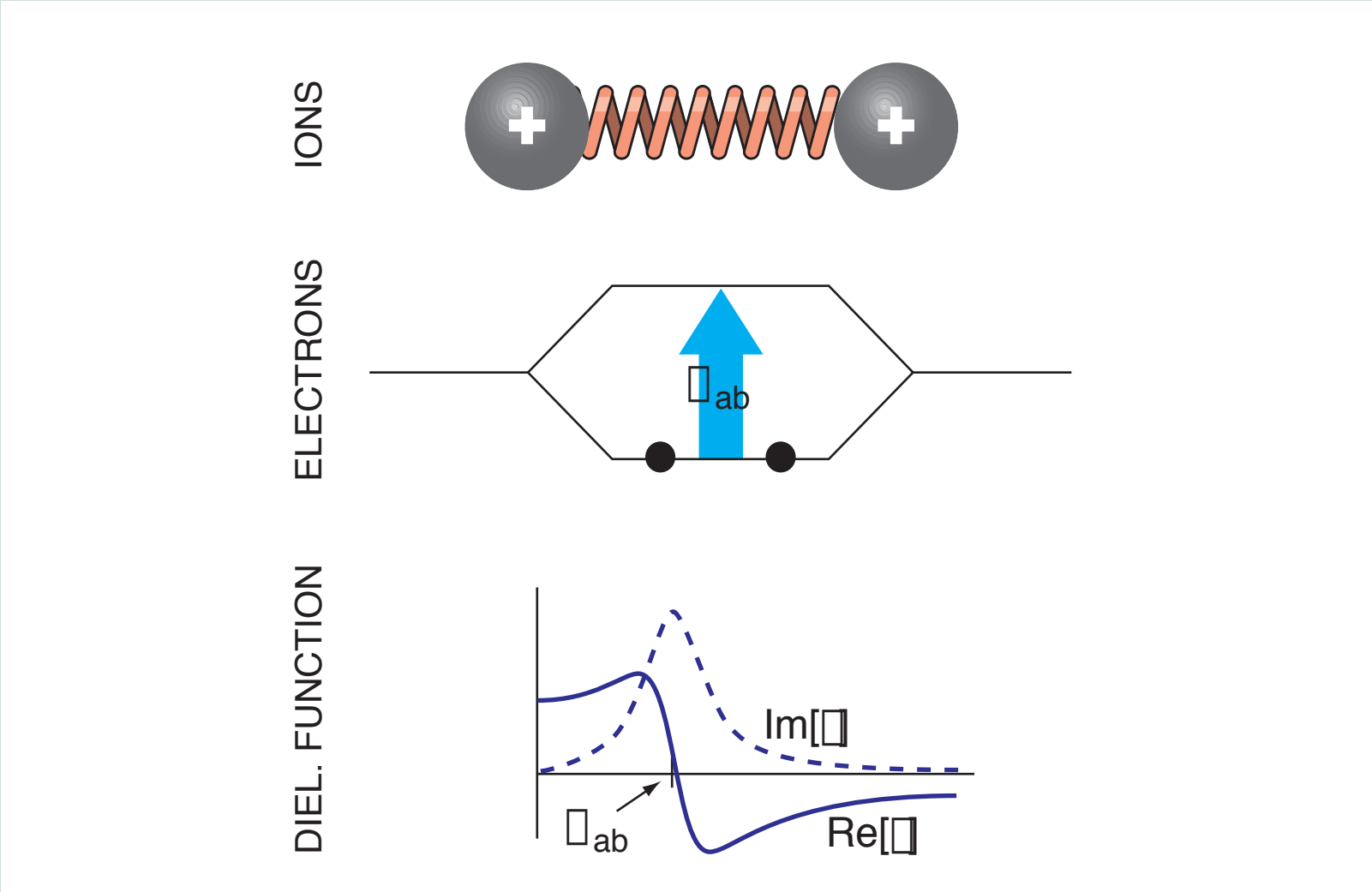


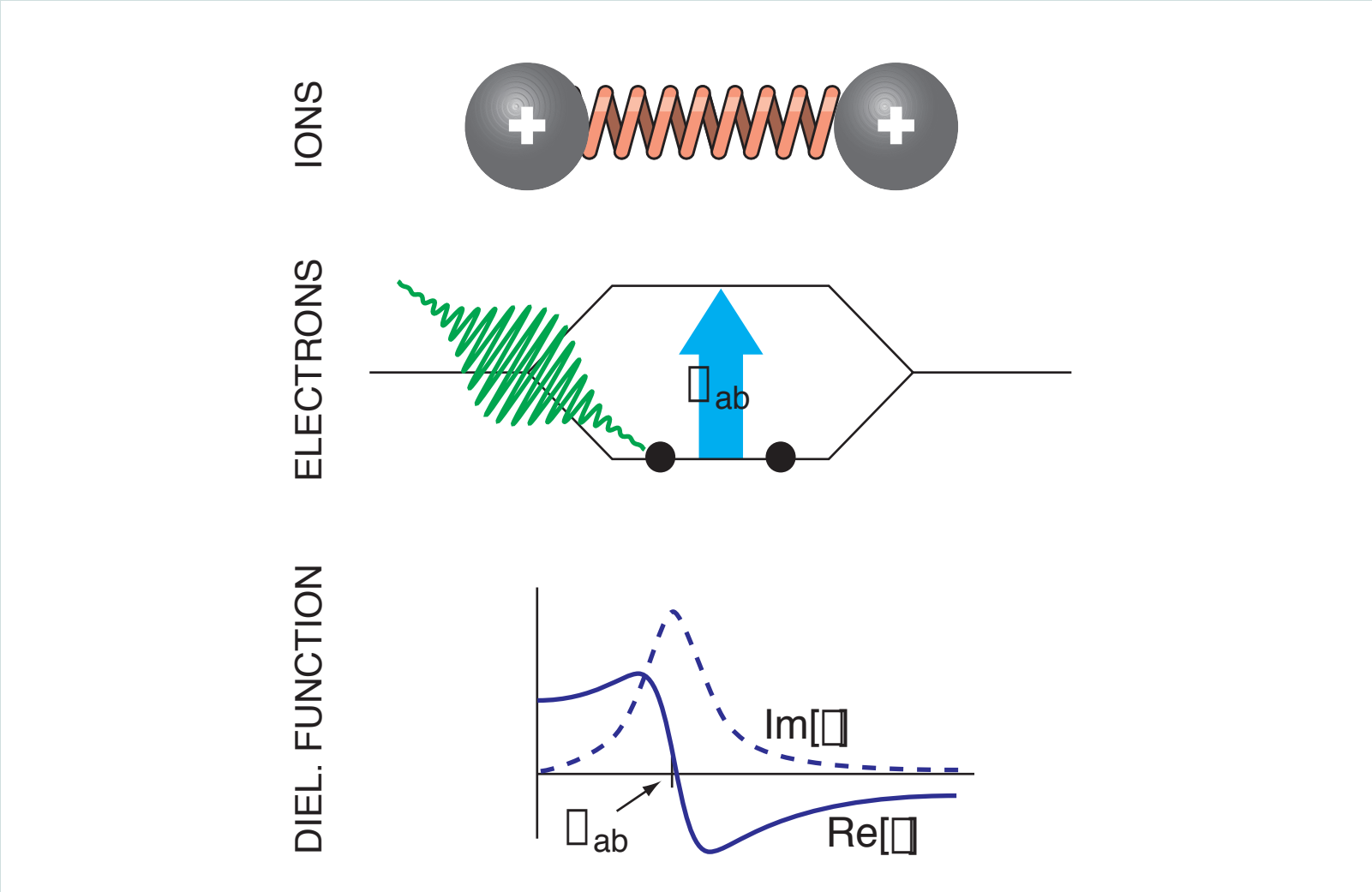
Bonding-antibonding splitting

"Two-atom" model



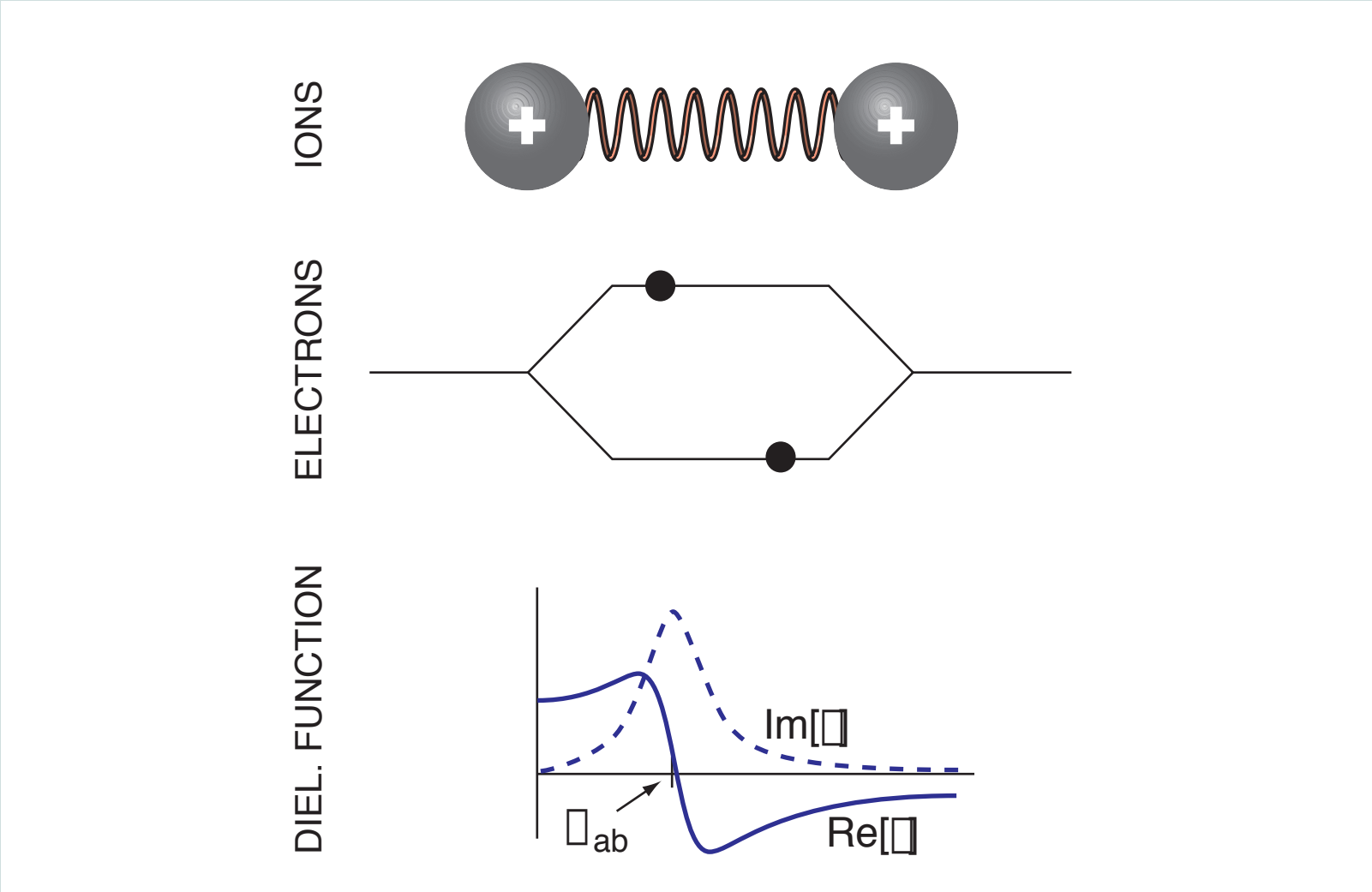
Lorentz oscillator model

"Two-atom" model



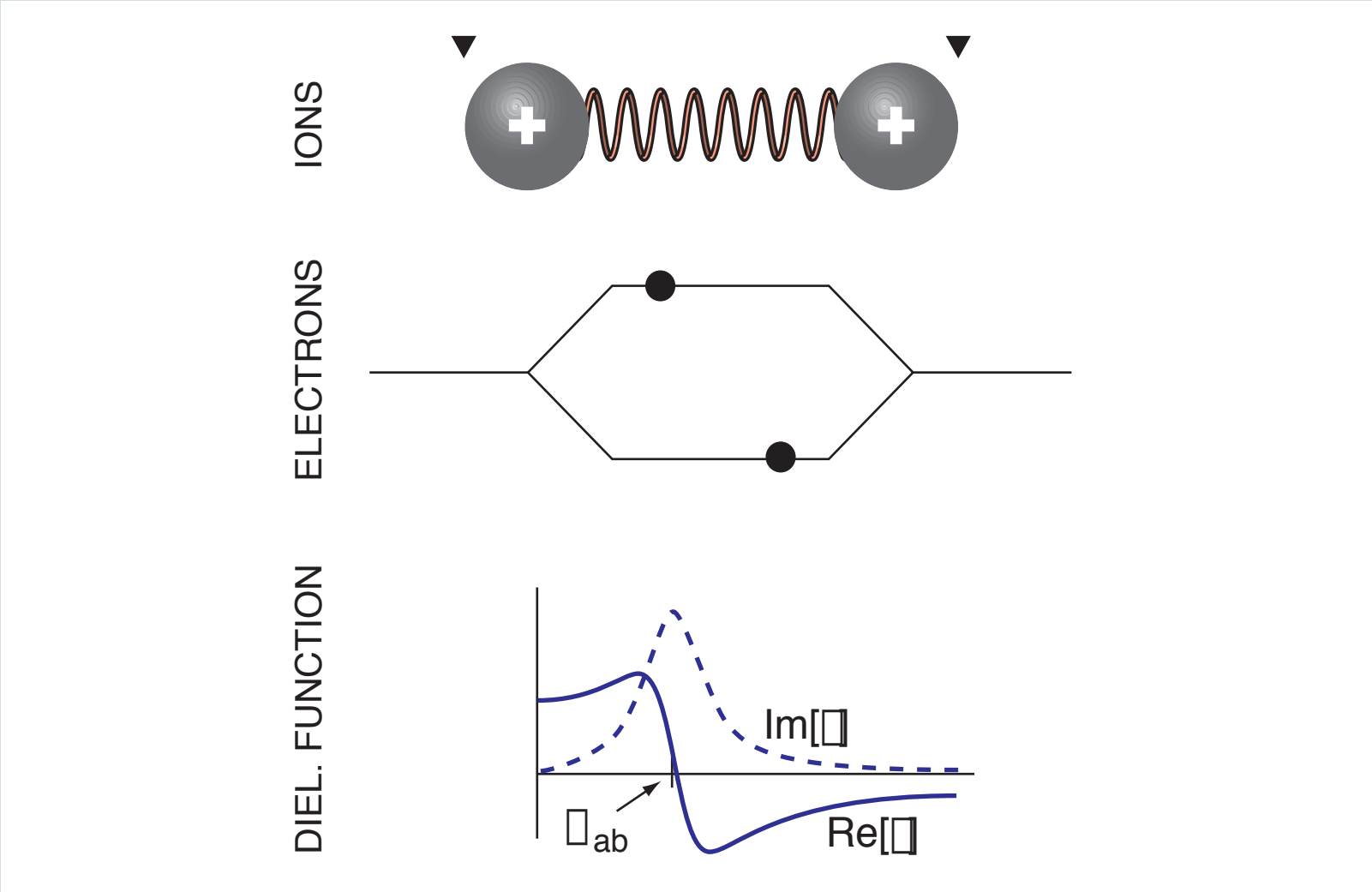
photon promotes an electron...

"Two-atom" model



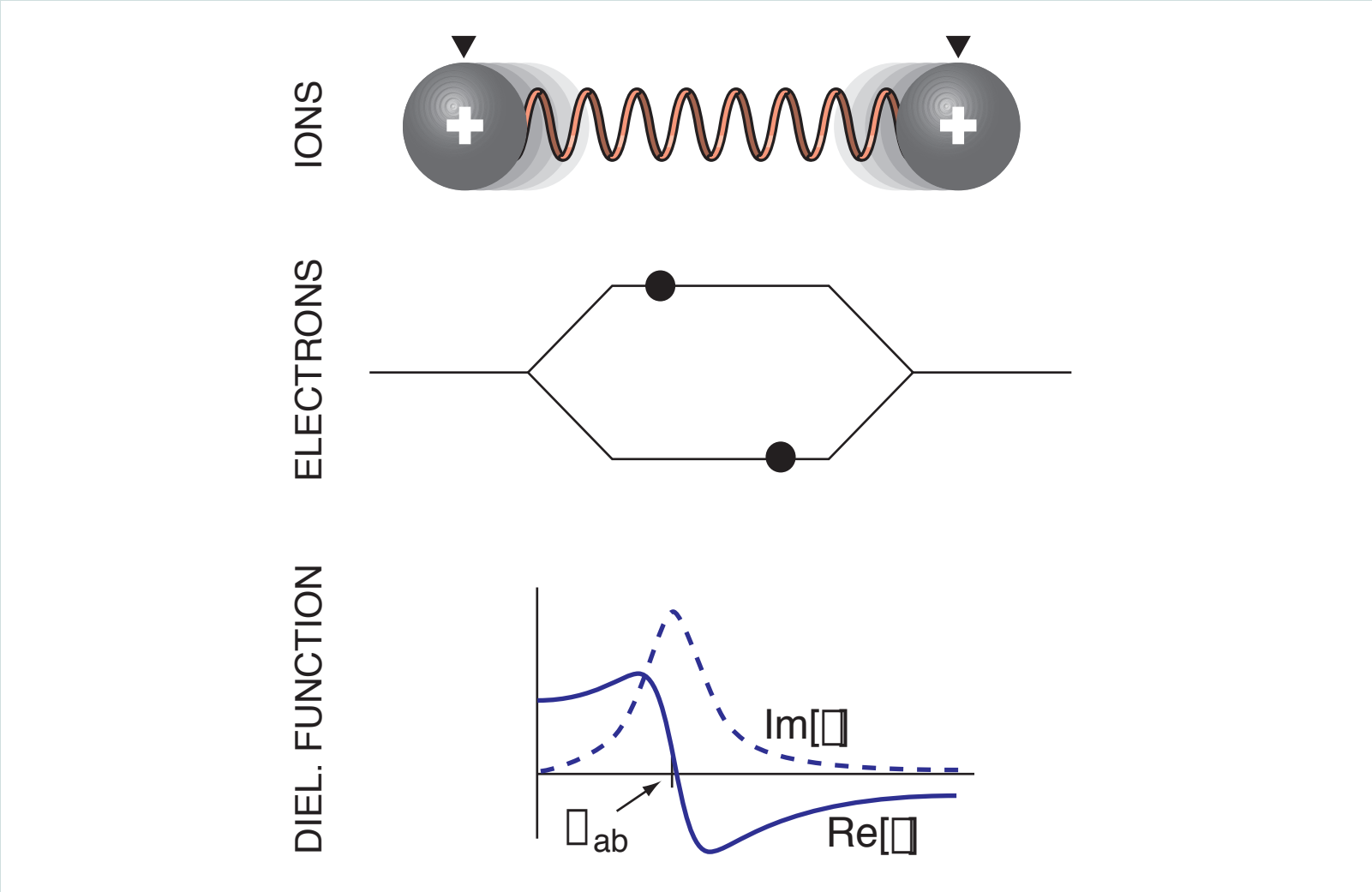
... weakening the bond...

"Two-atom" model



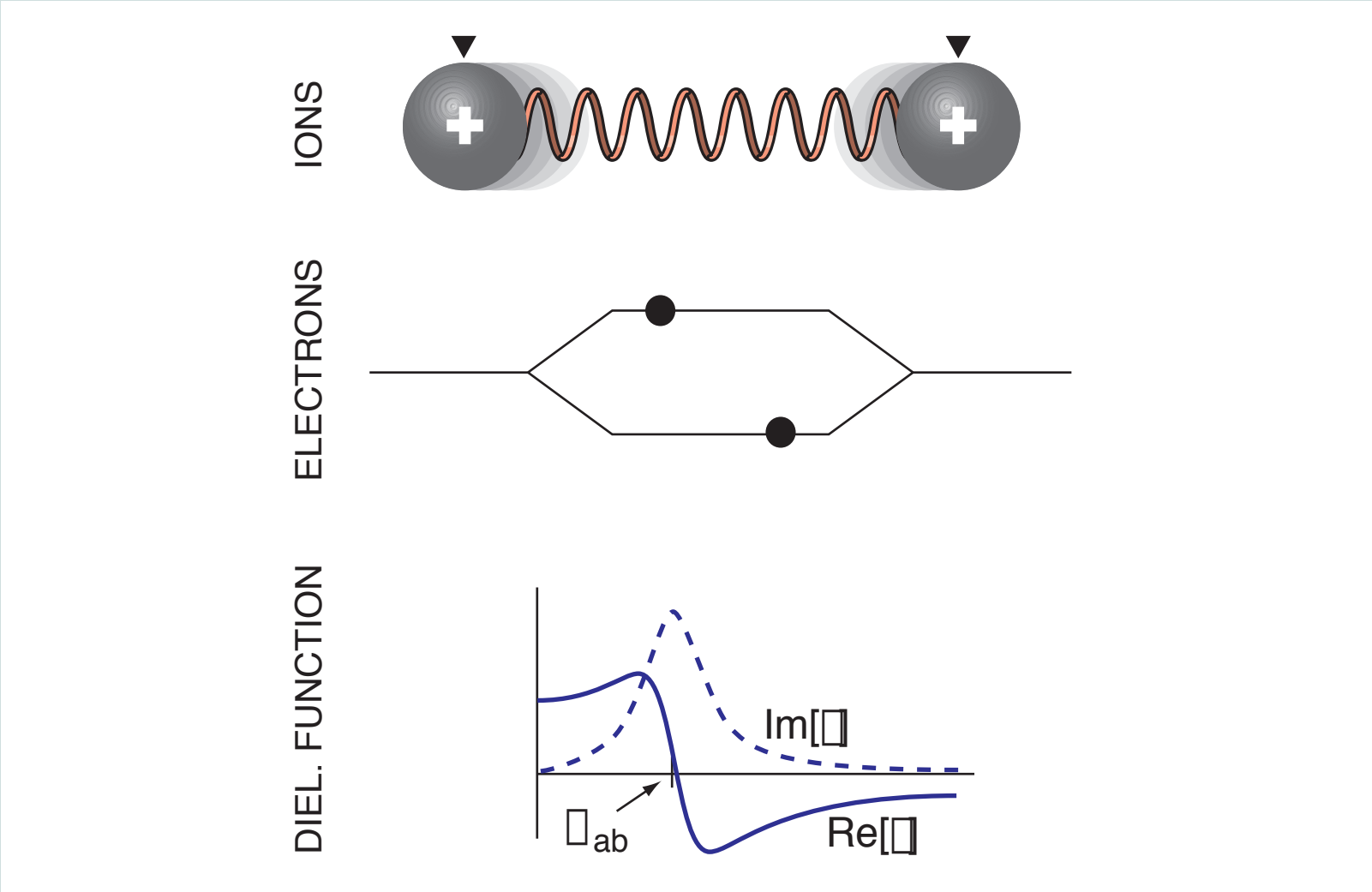
... establishing new equilibrium positions

"Two-atom" model



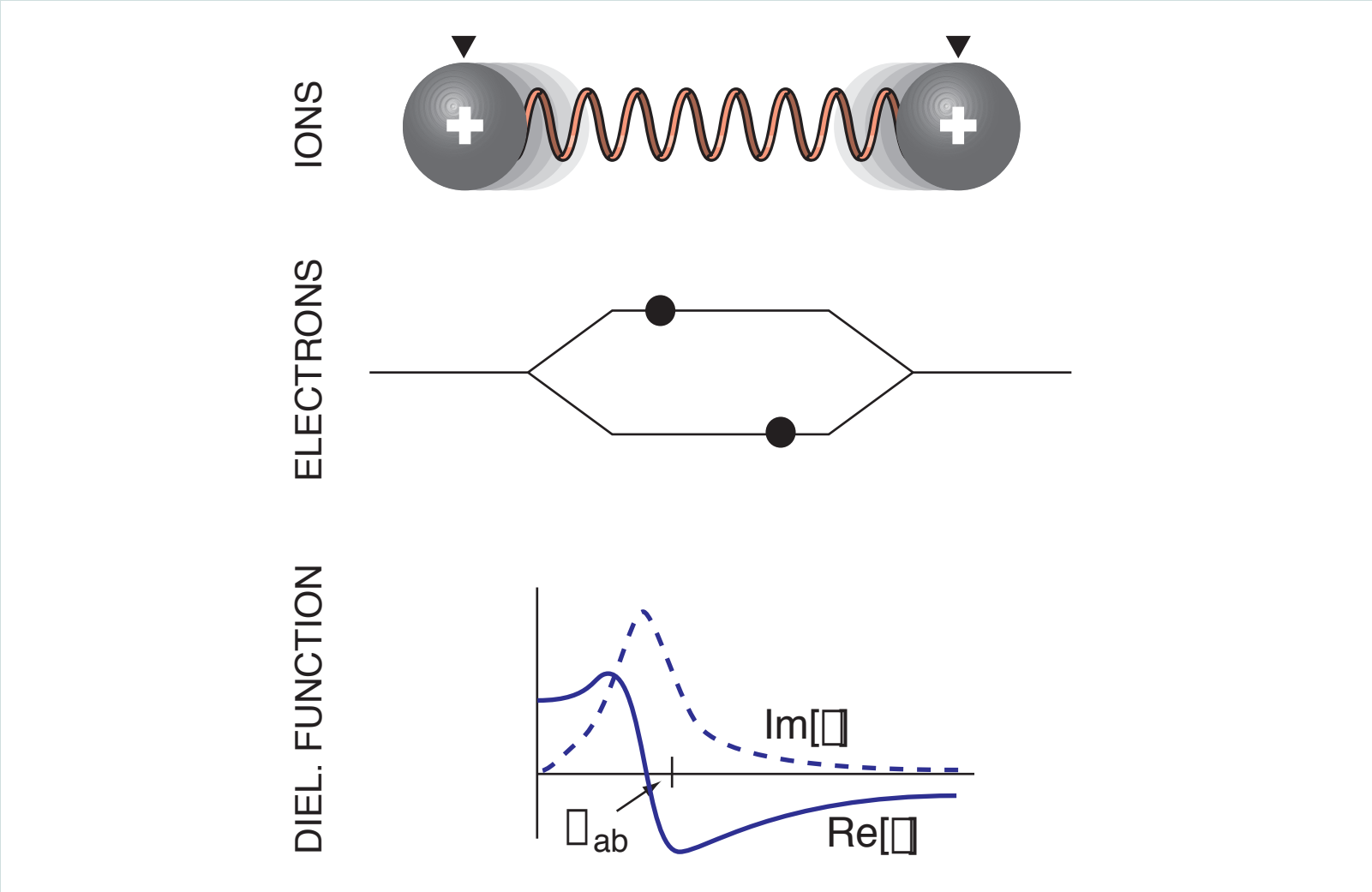
ions move to new equilibrium positions...

"Two-atom" model



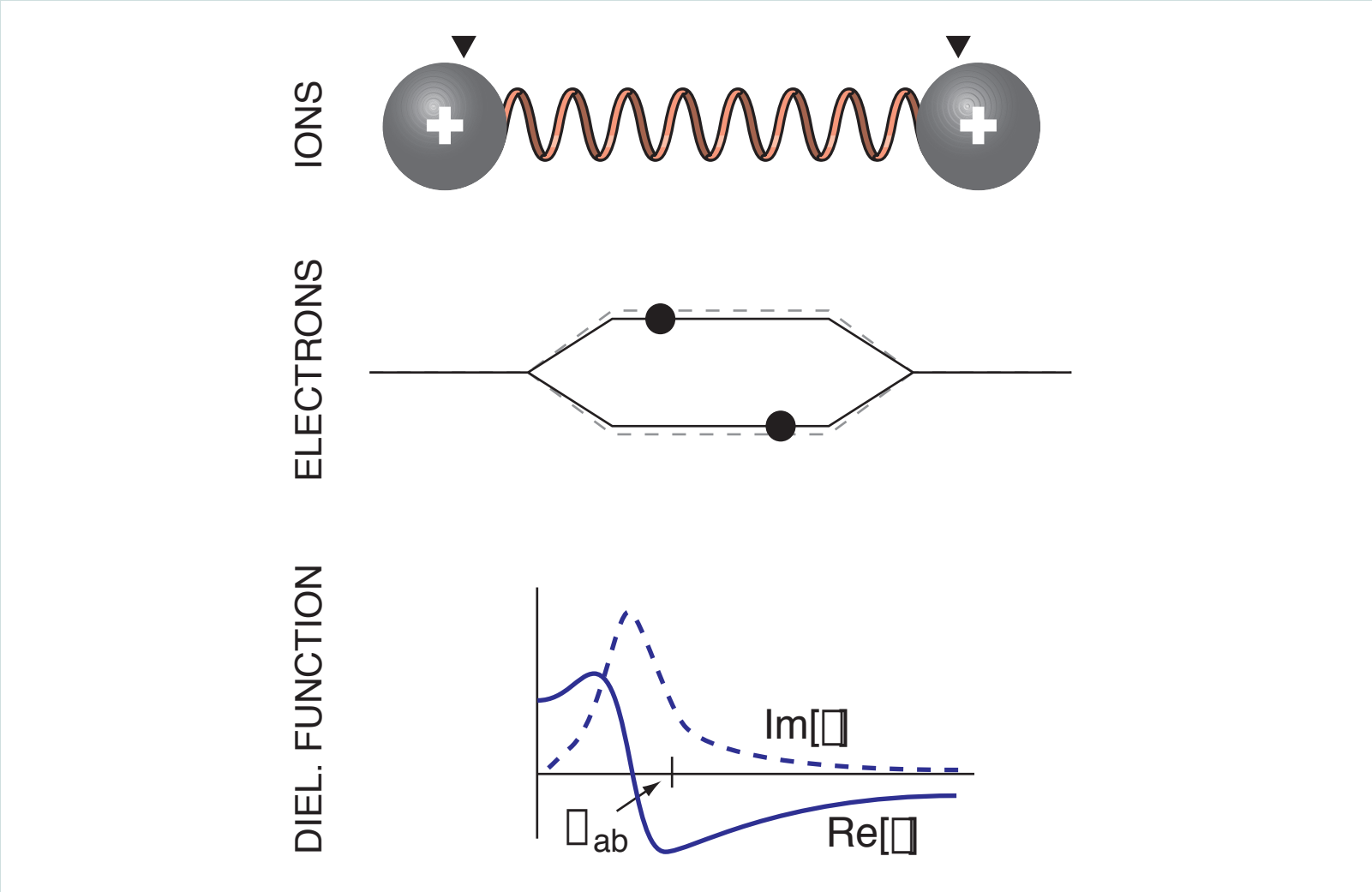
... decreasing the splitting...

"Two-atom" model



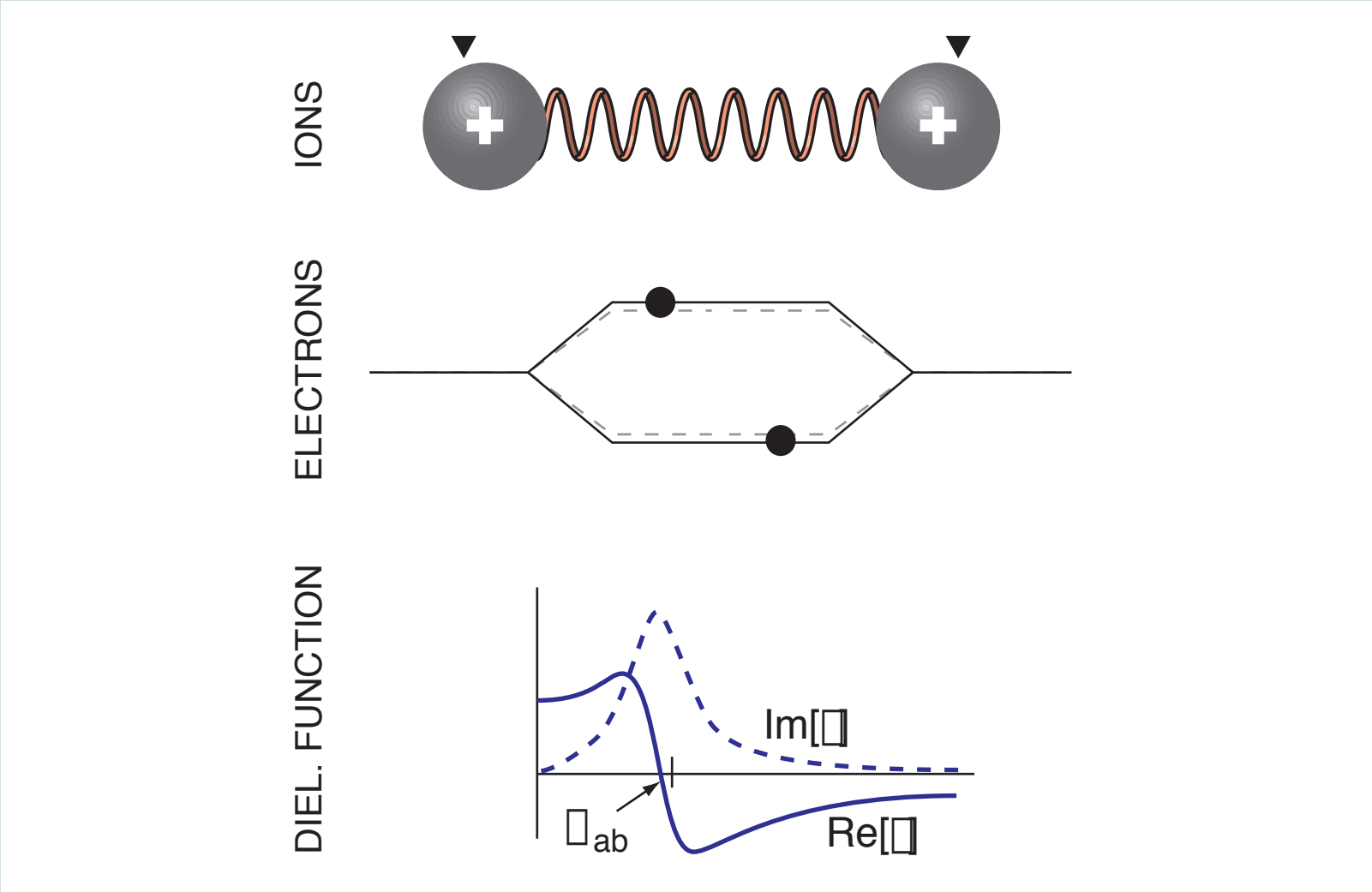
... and redshifting the dielectric function

"Two-atom" model



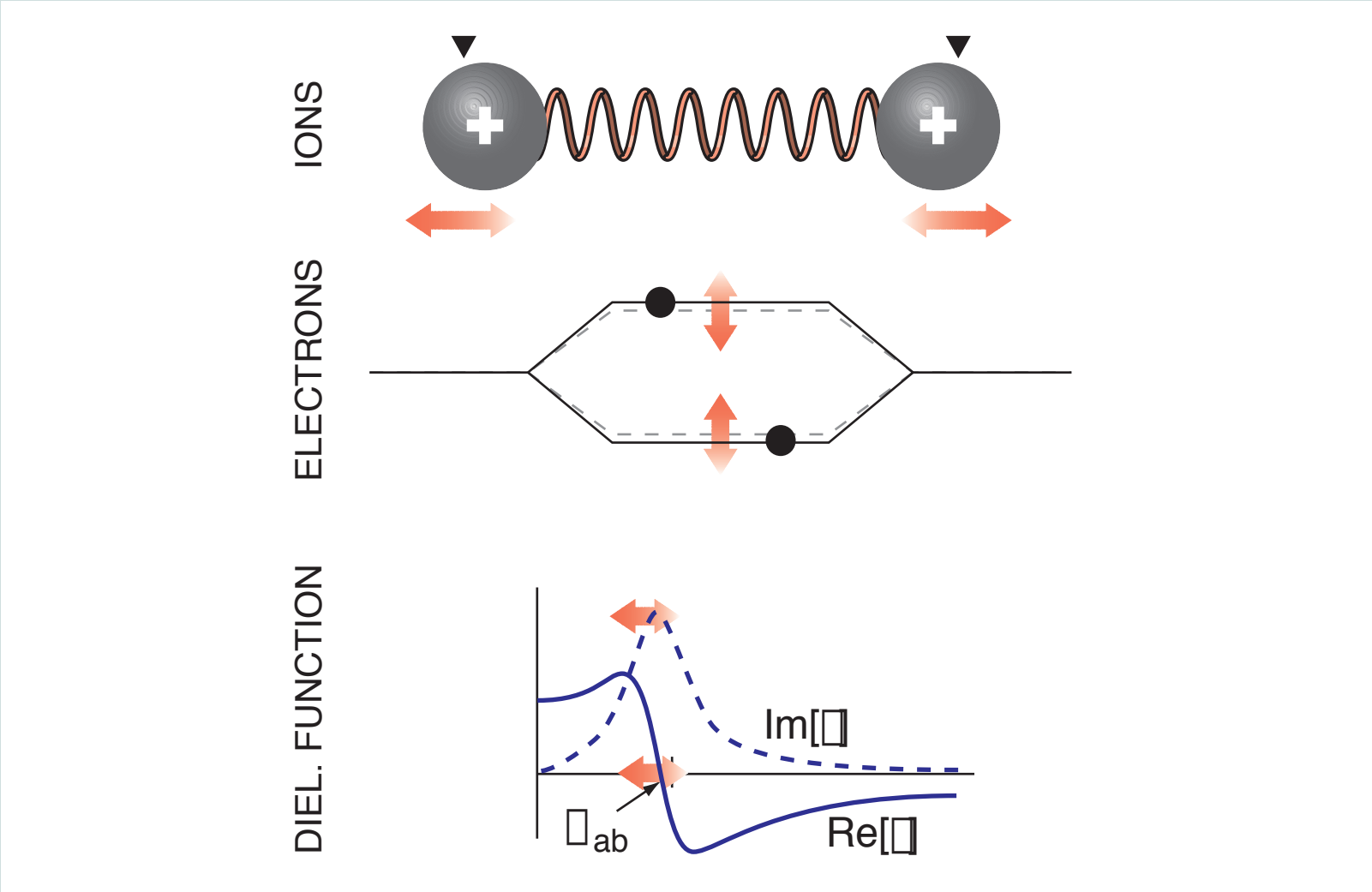
ions overshoot equilibrium positions...

"Two-atom" model



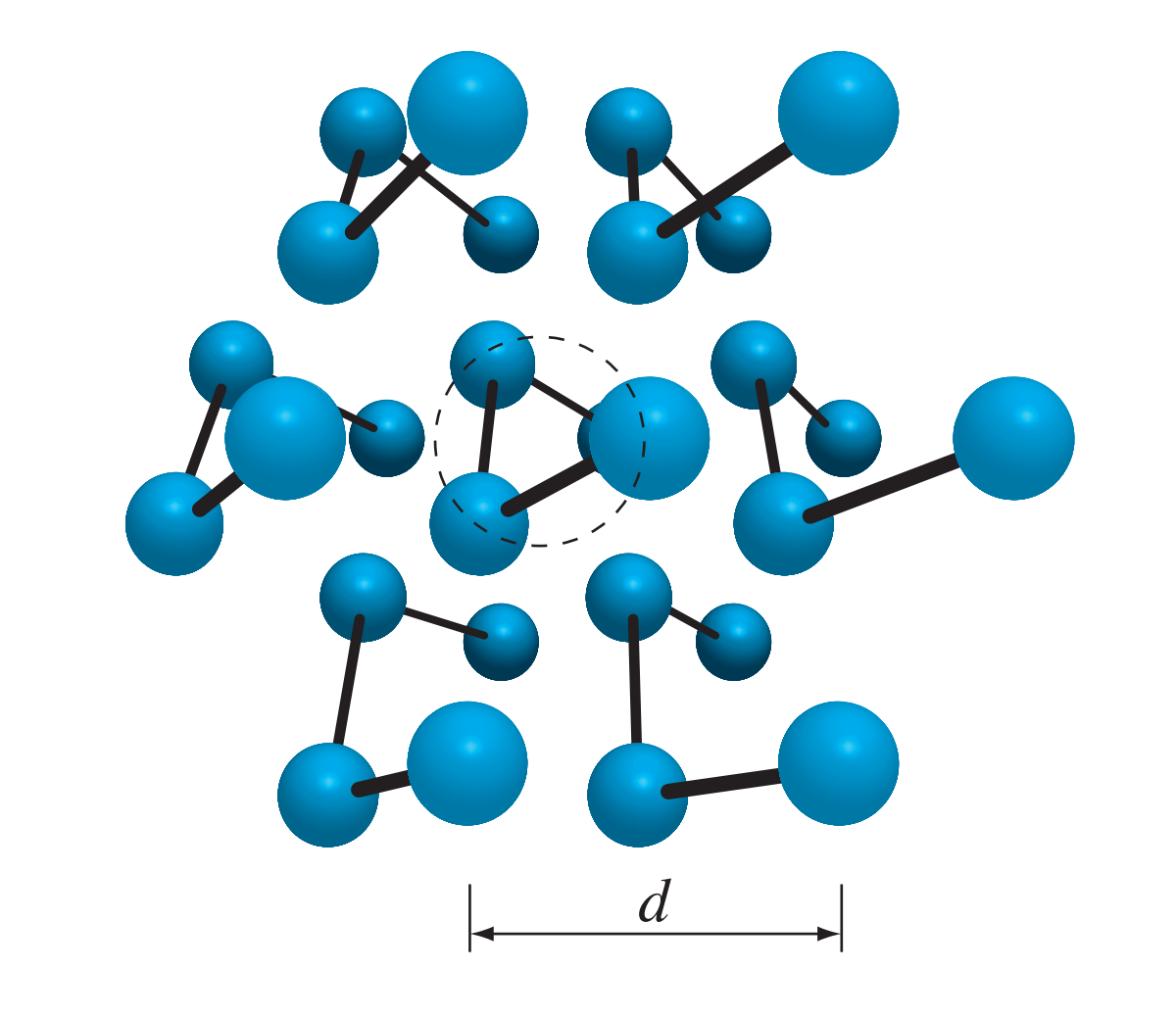
... reversing travel and overshooting again

"Two-atom" model



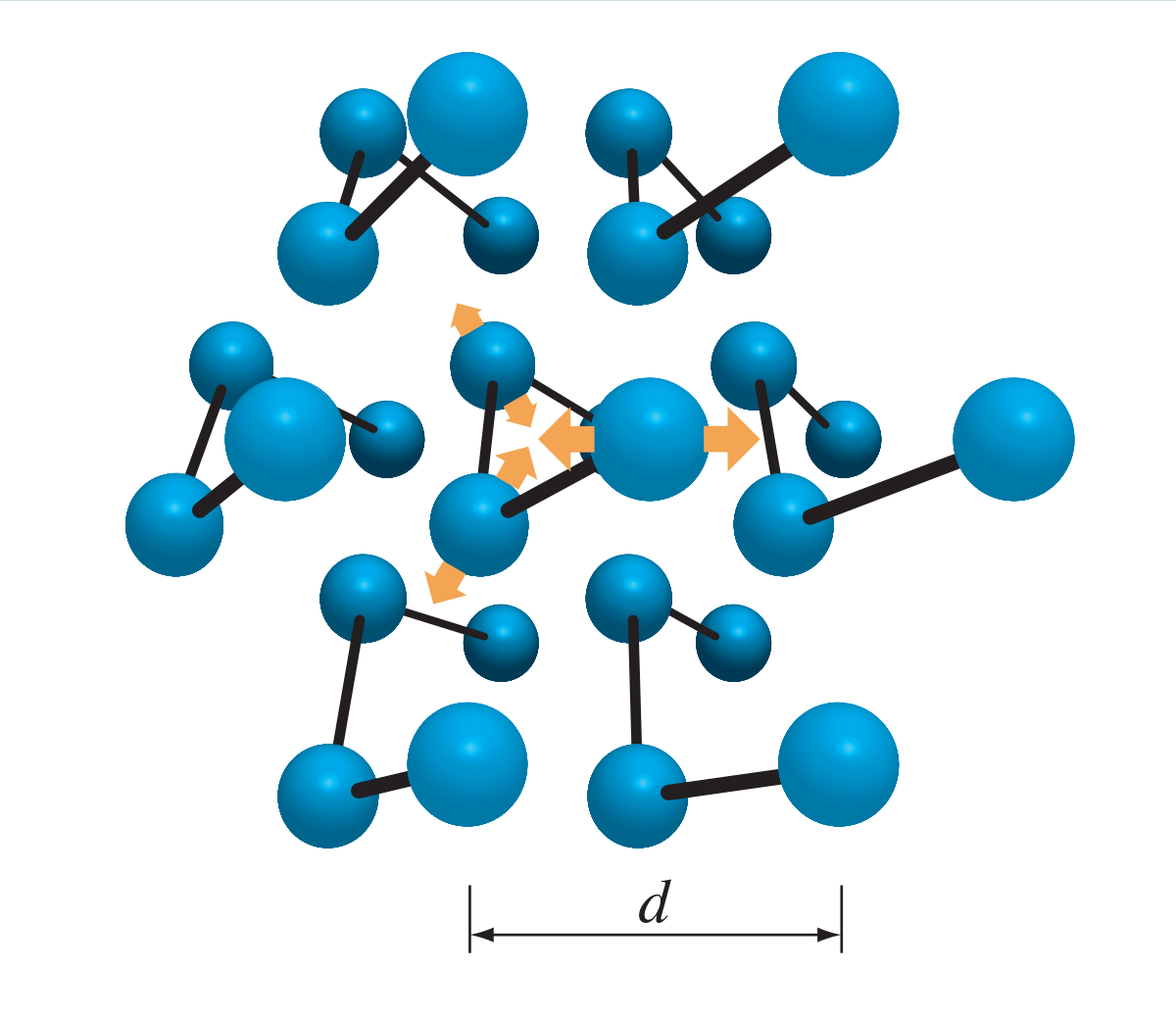
oscillation around displaced equilibrium position

Tellurium lattice



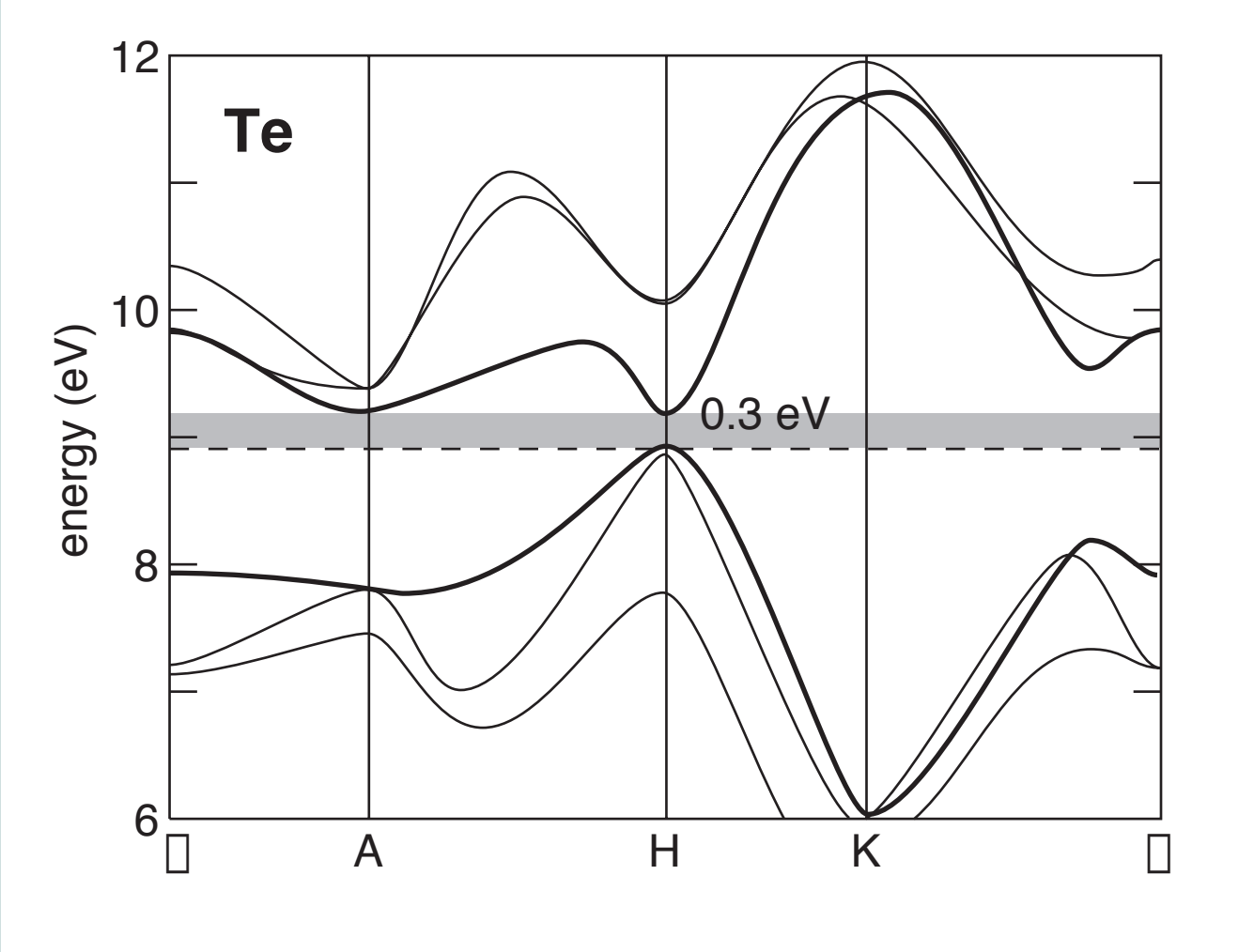
helical radius x=0.26d

Tellurium lattice



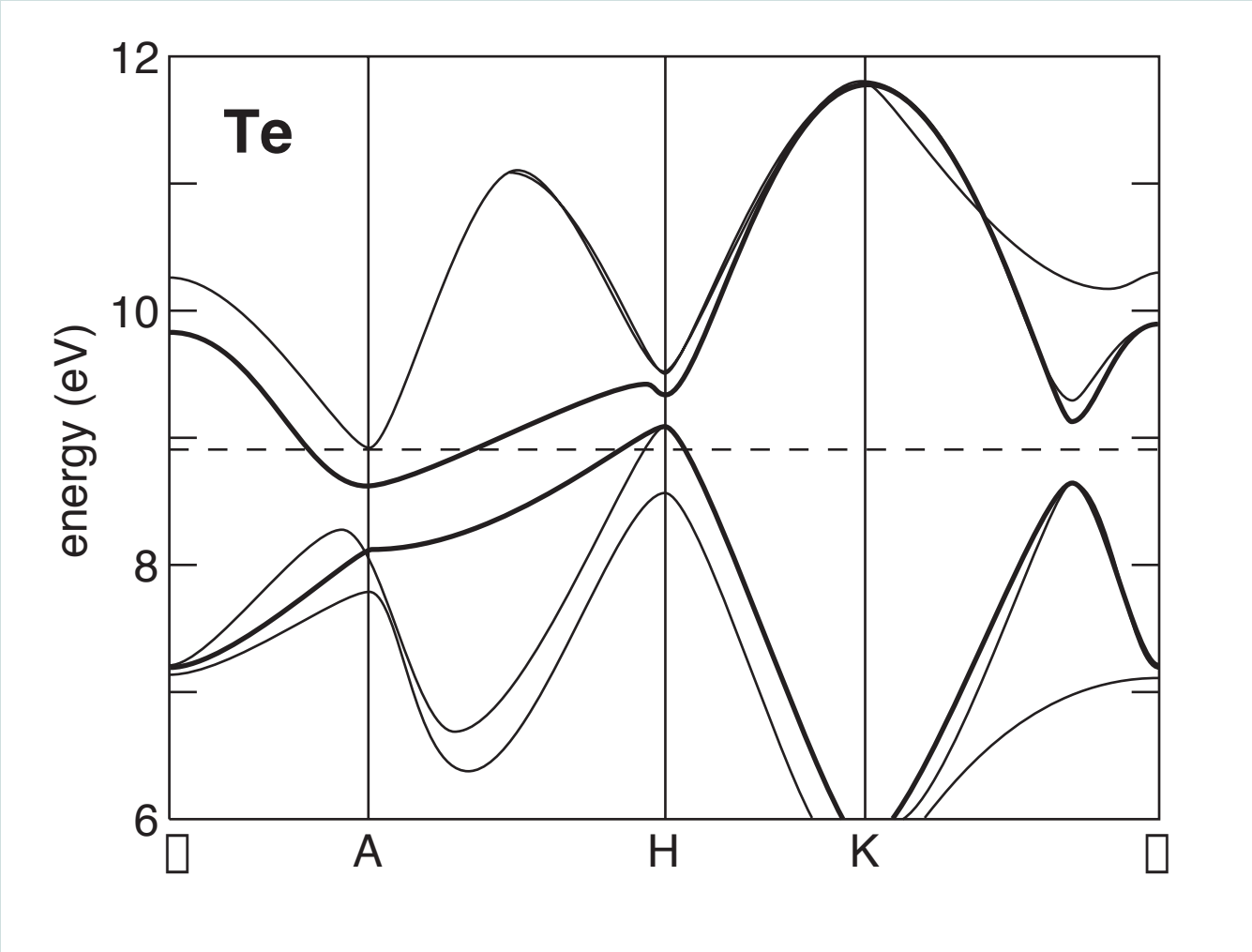
A_1 mode modulates x

Band structure is sensitive to x



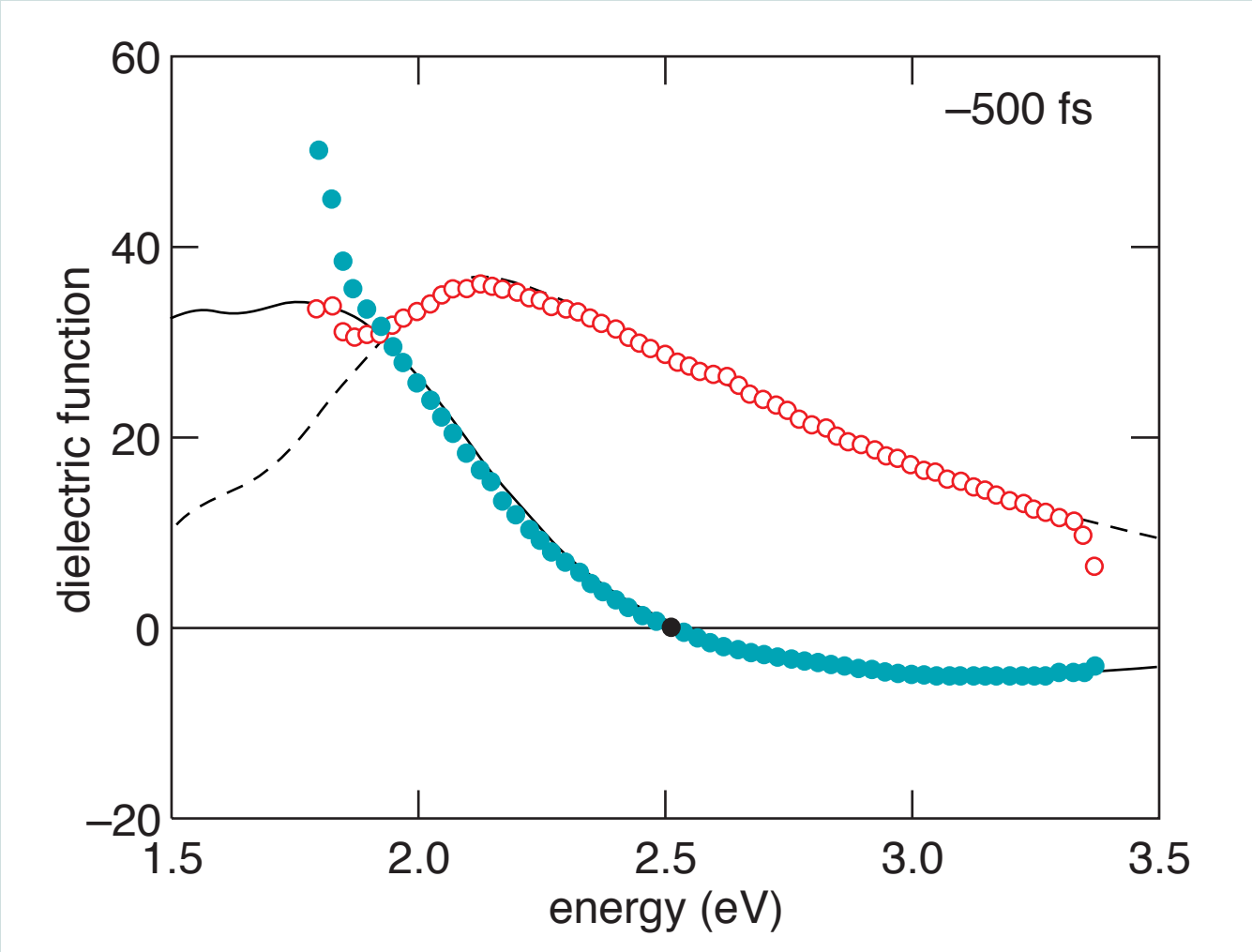
P. Tangney (Princeton) and S. Fahy (Cork), *private communication*

Bands cross when x changes by 6%

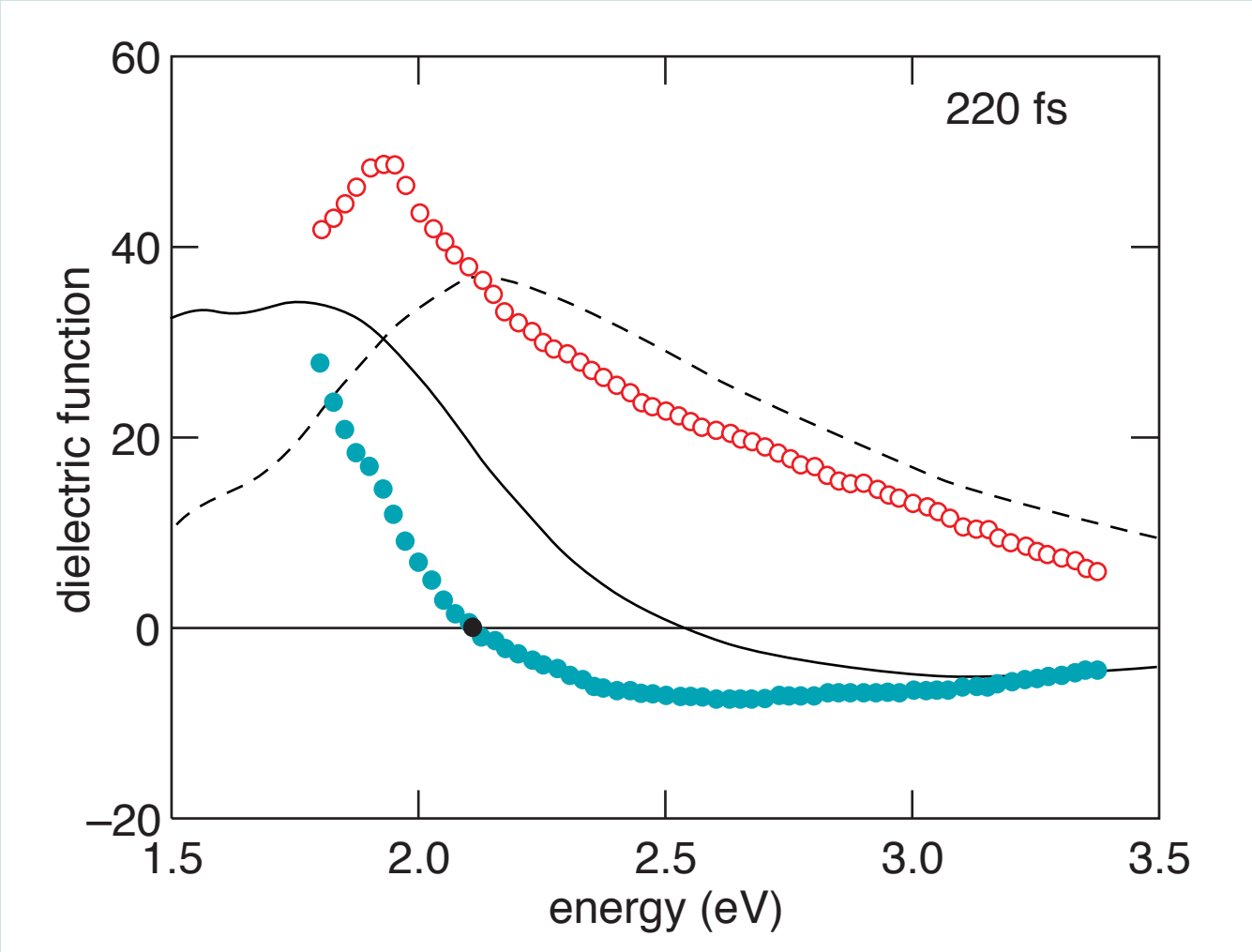


P. Tangney (Princeton) and S. Fahy (Cork), *private communication*

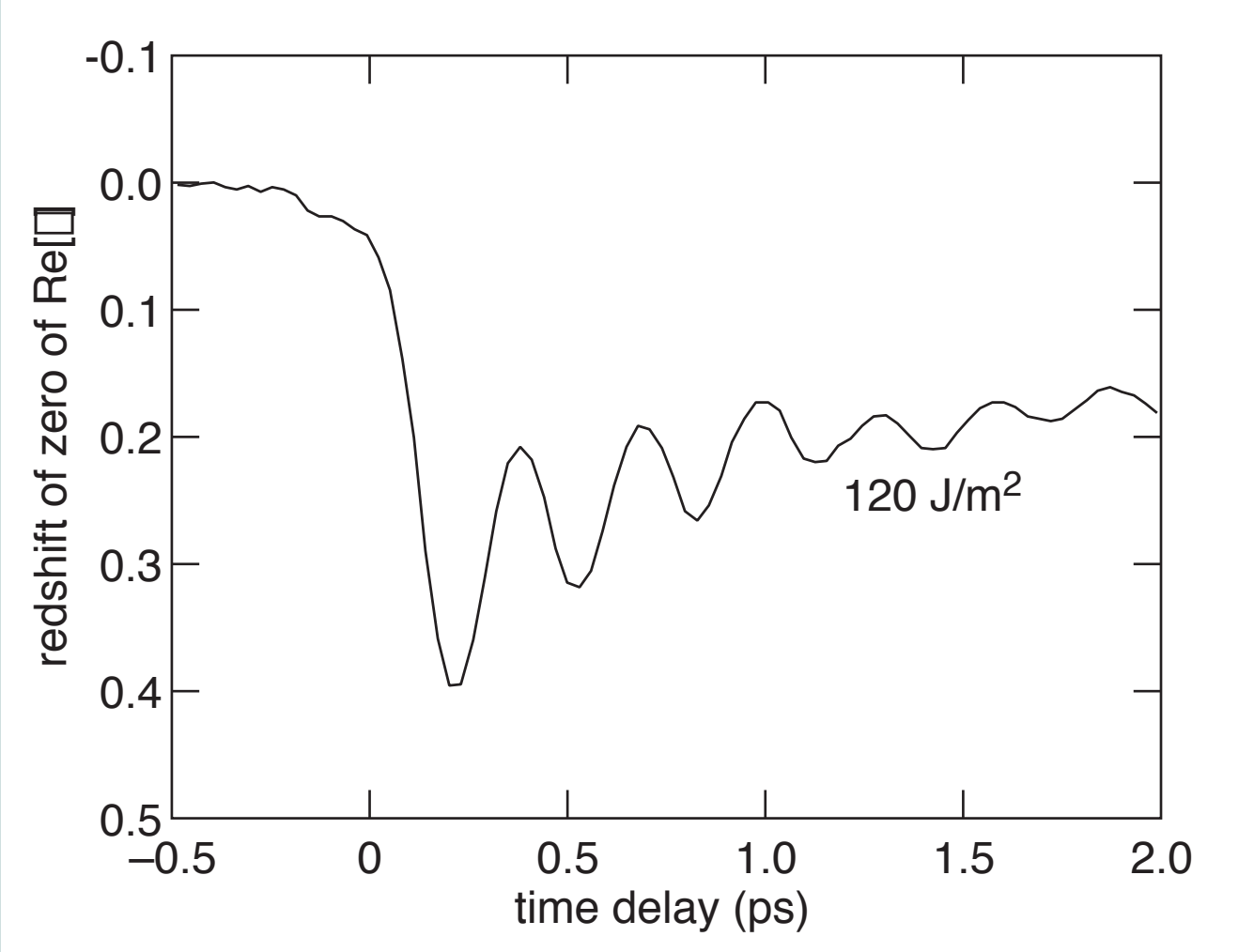
Track zero of real part



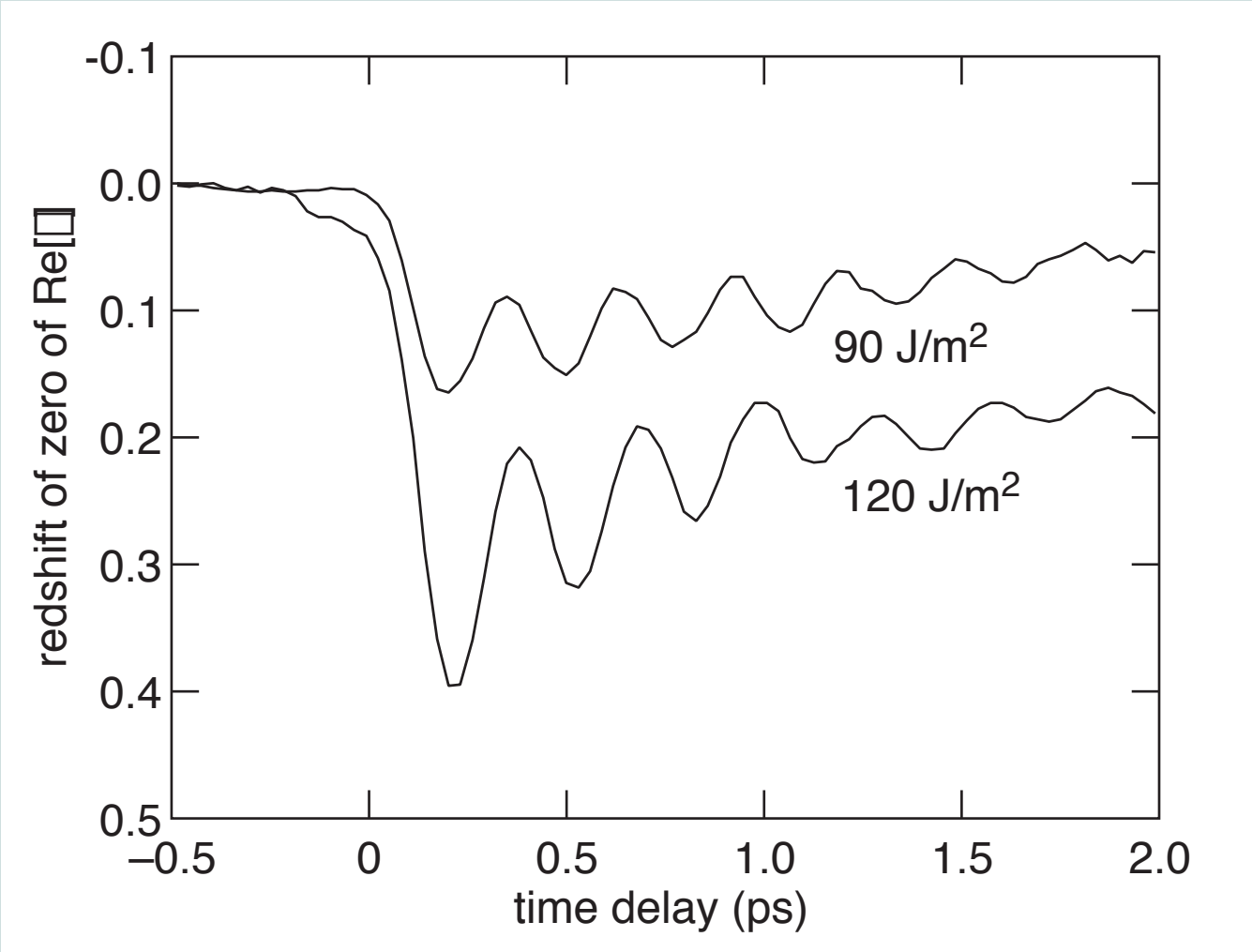
Track zero of real part



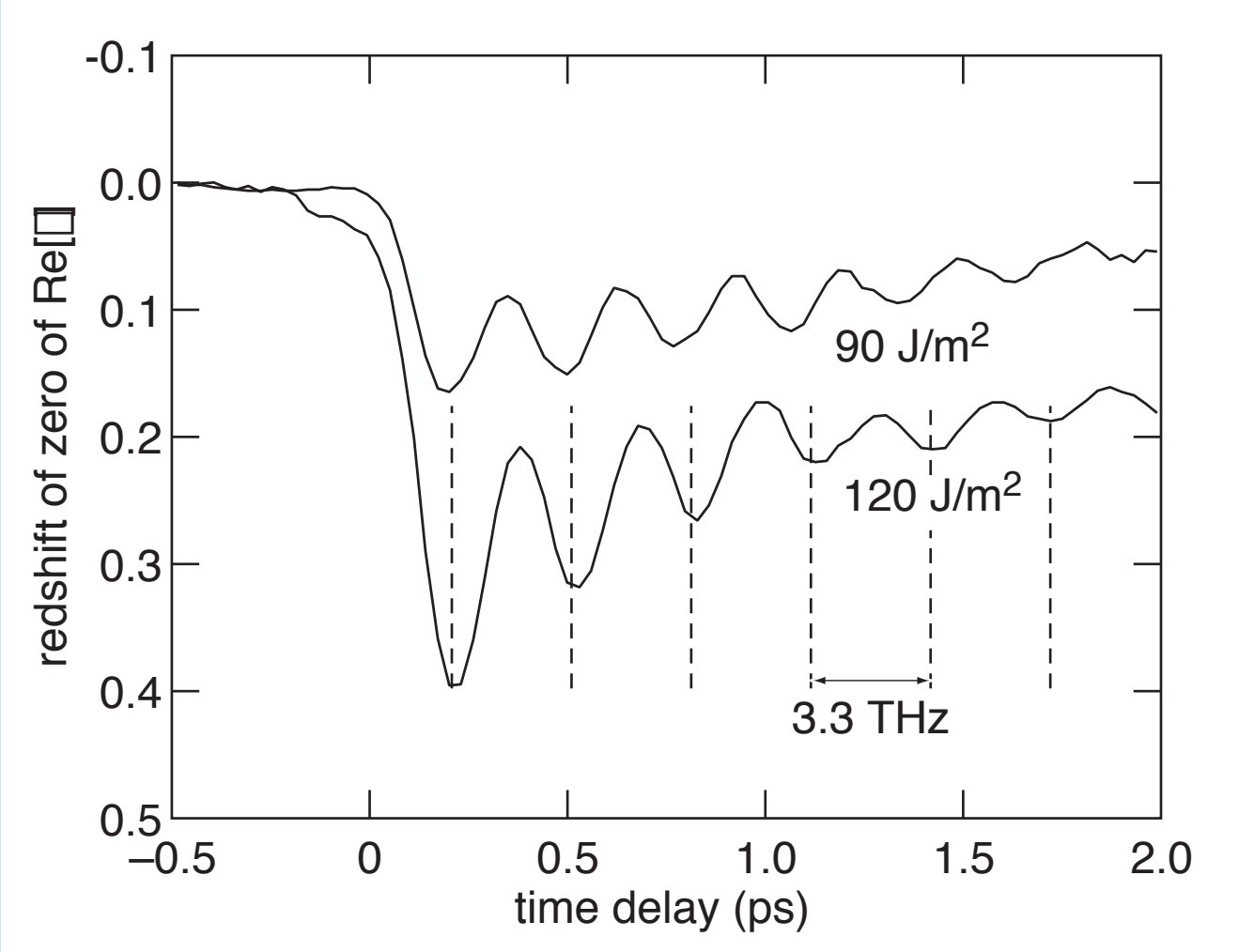
Track zero of real part



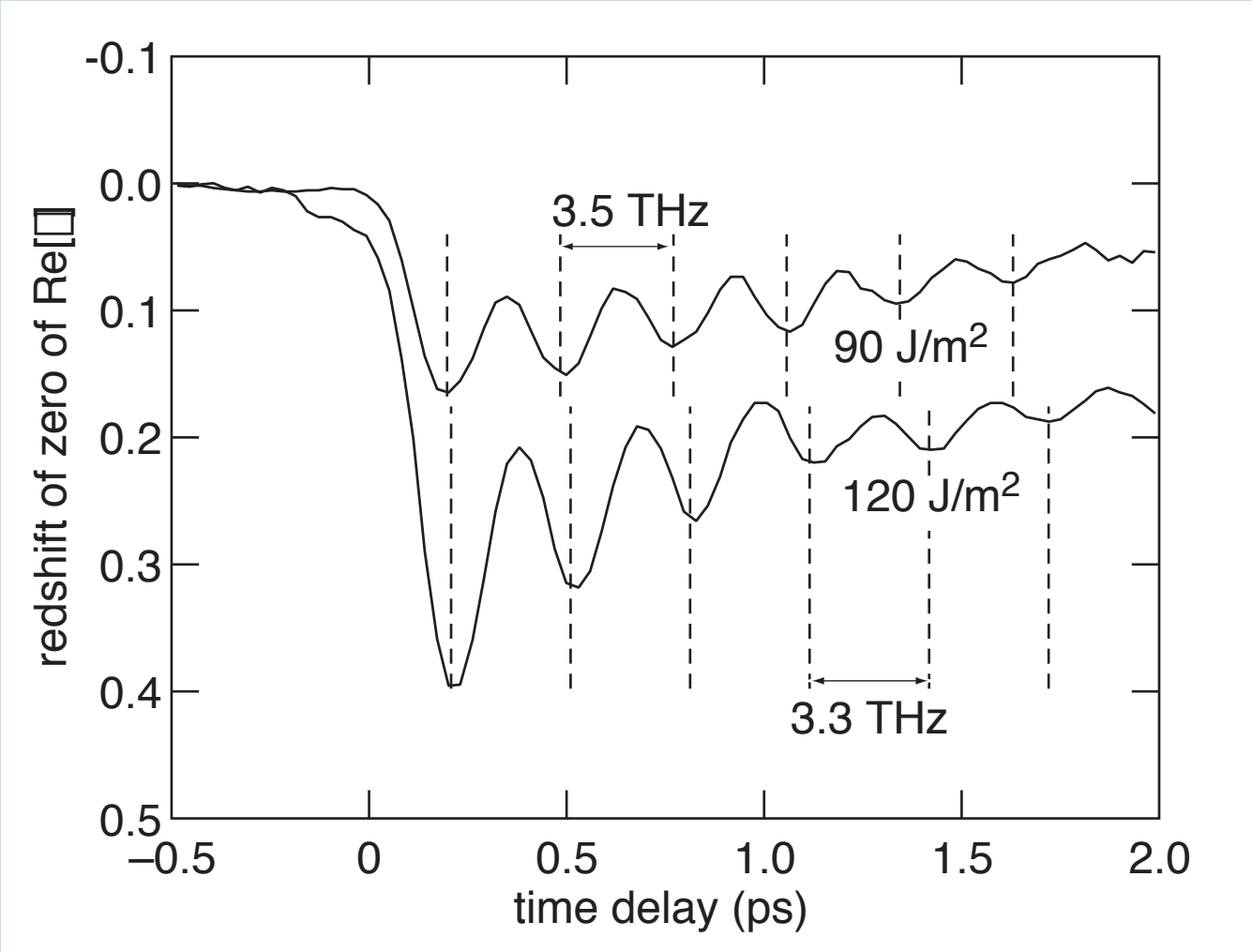
Higher fluence: larger amplitude phonons



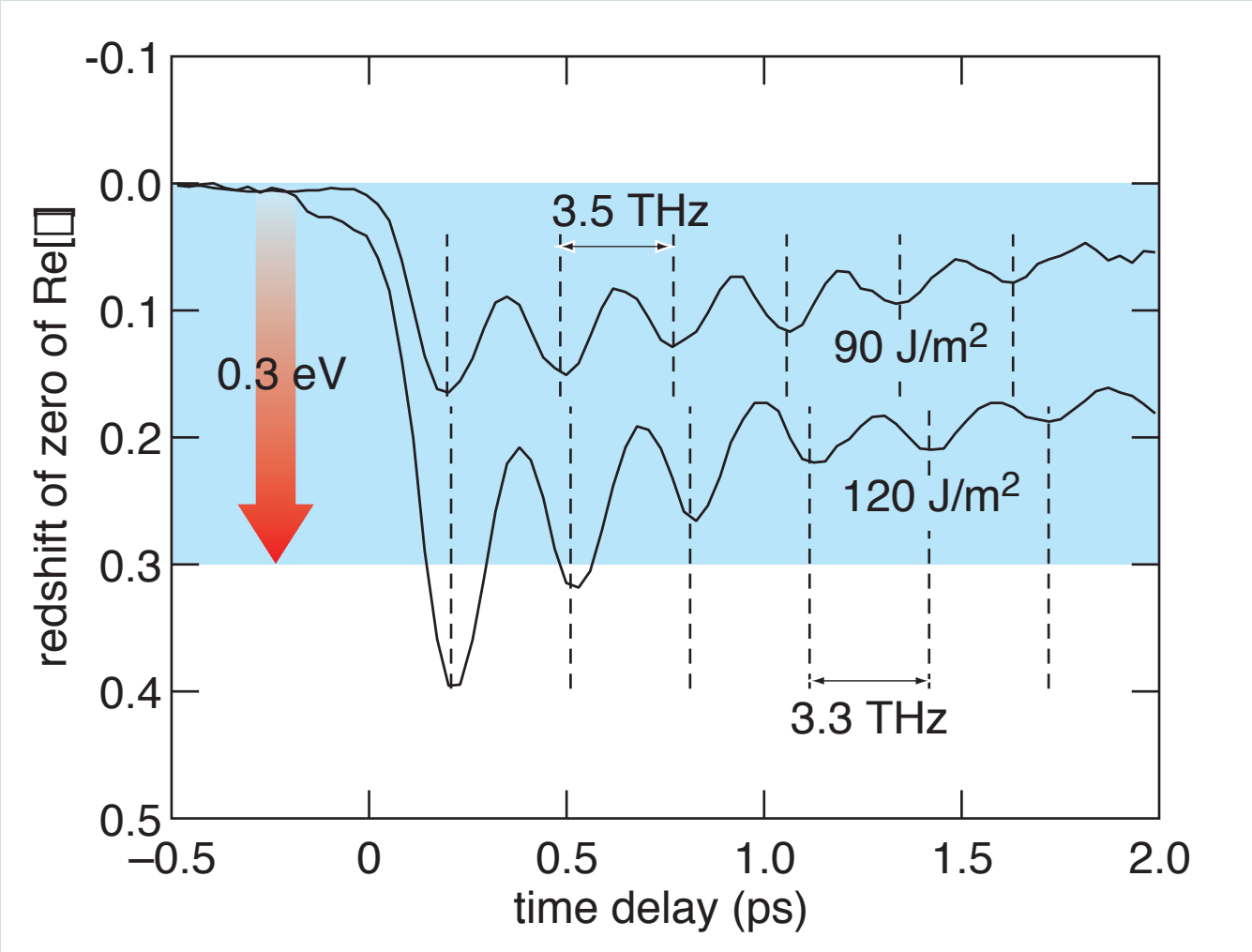
Frequency less than 3.6 THz



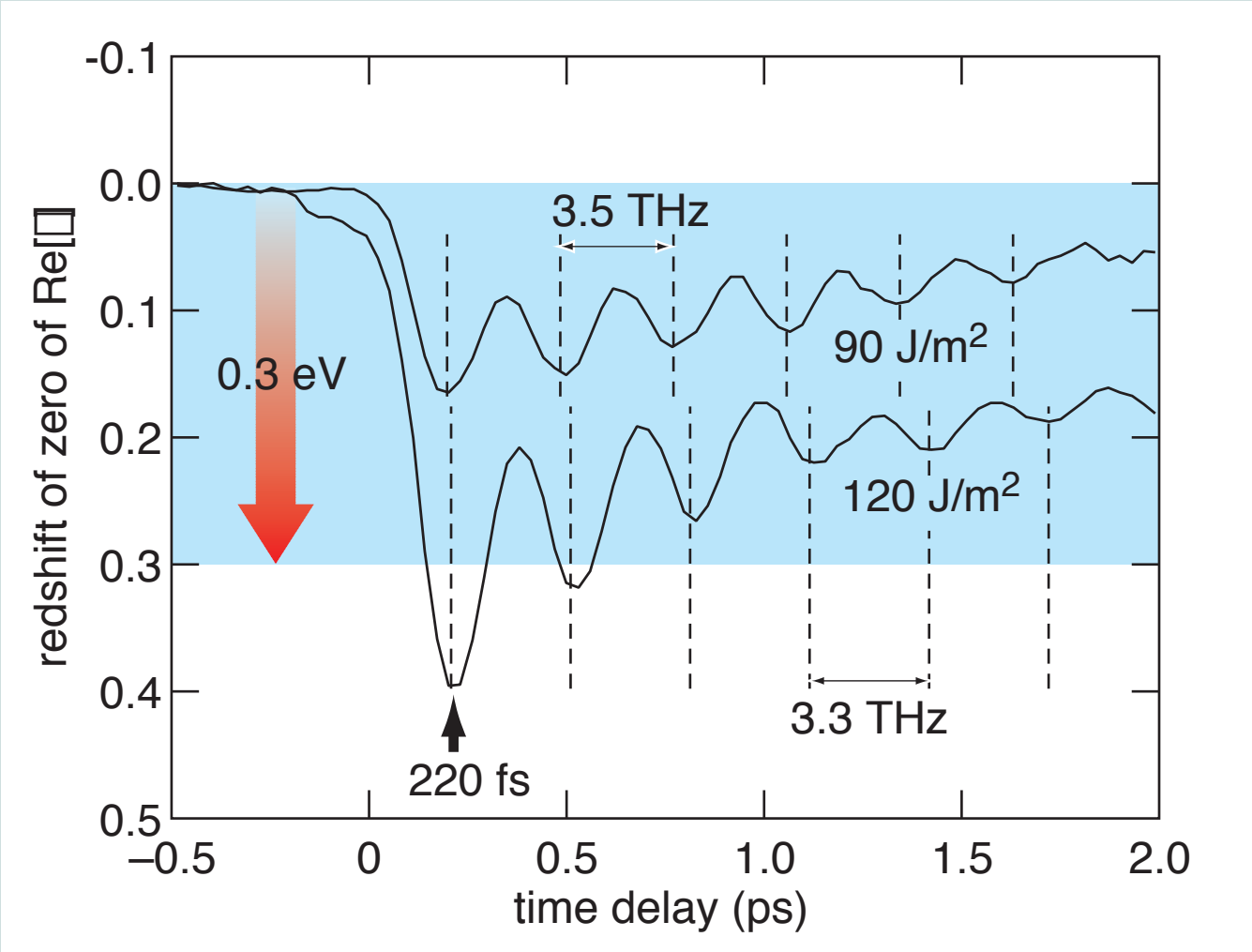
Phonon mode softens



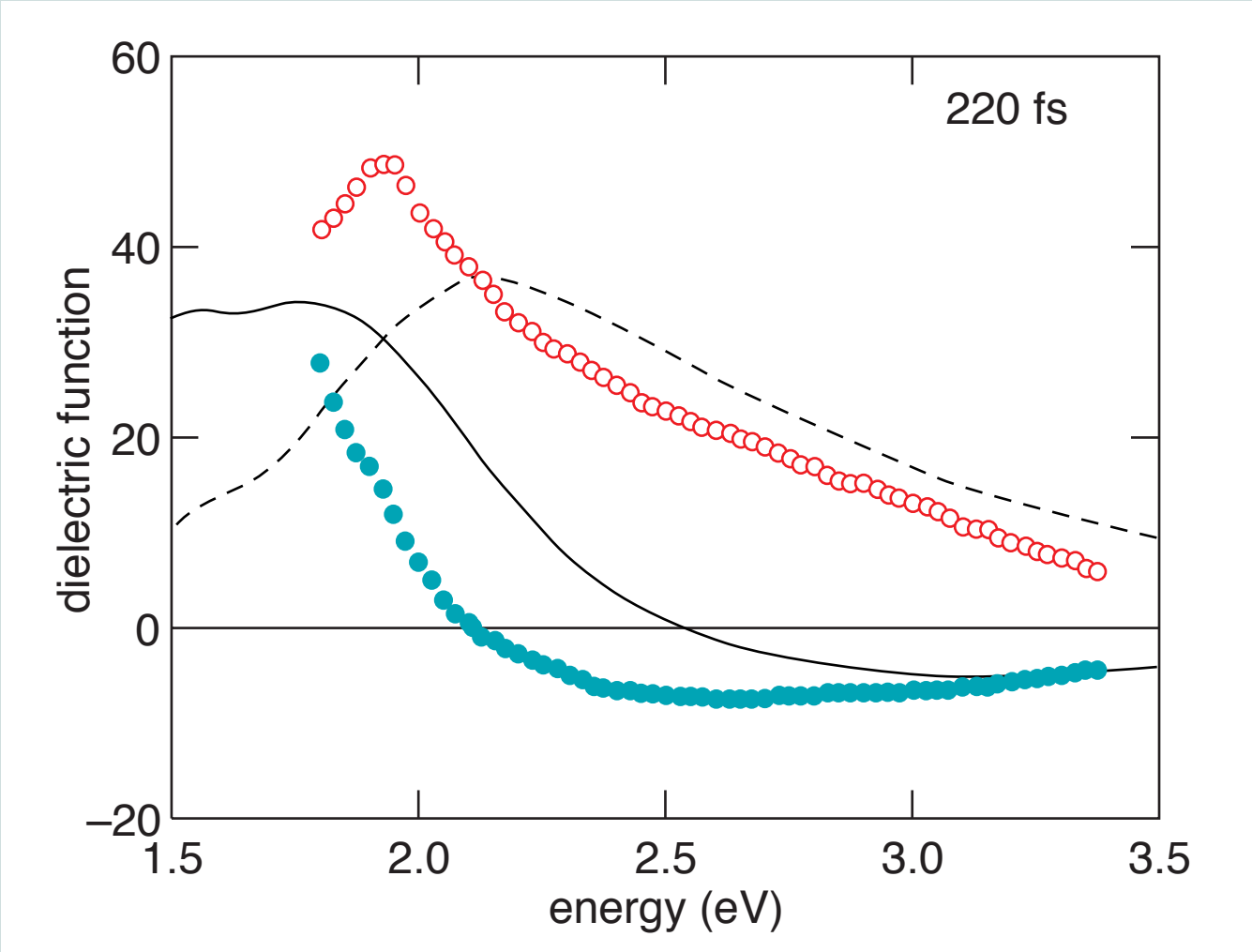
Compare shift to band gap



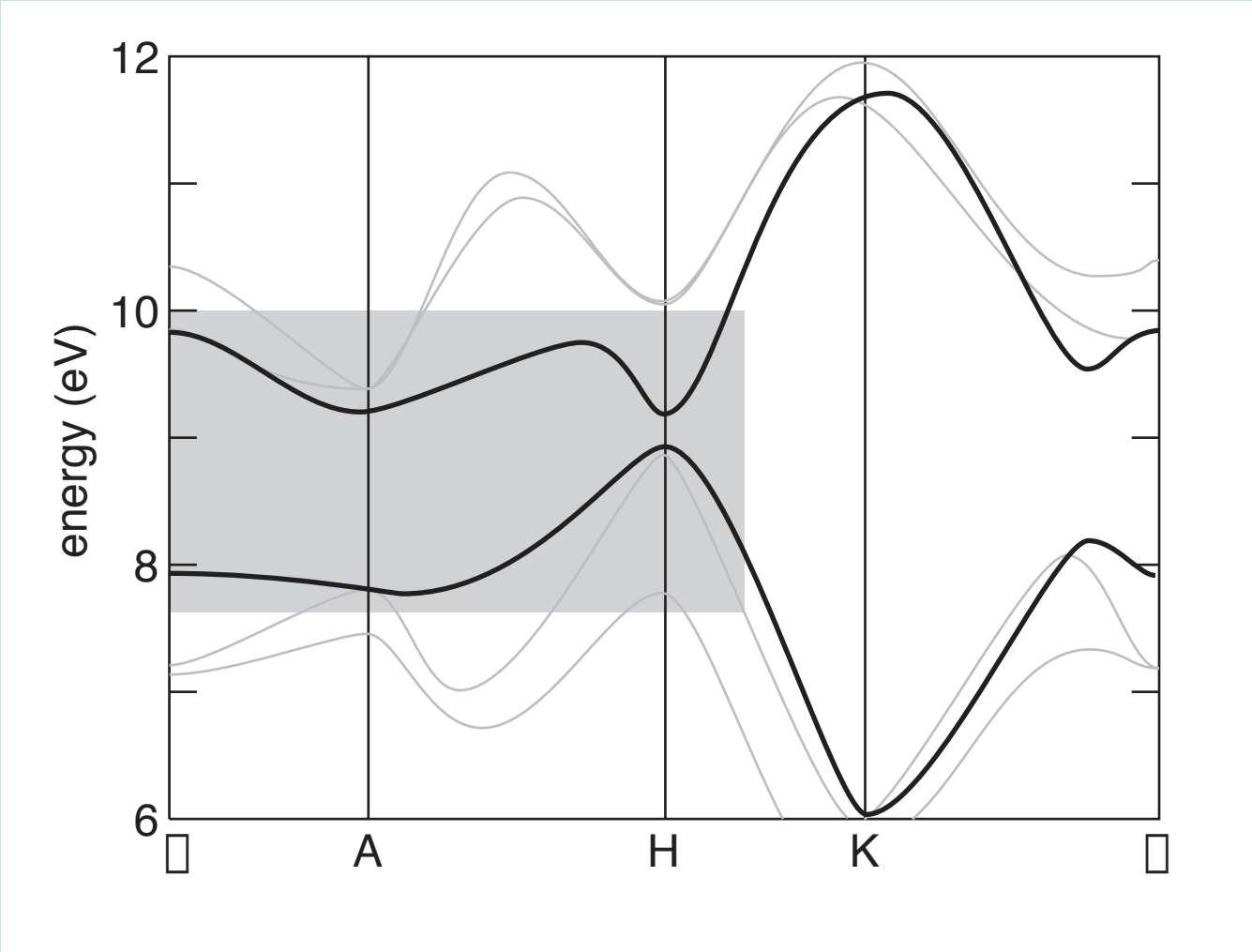
Compare shift to band gap



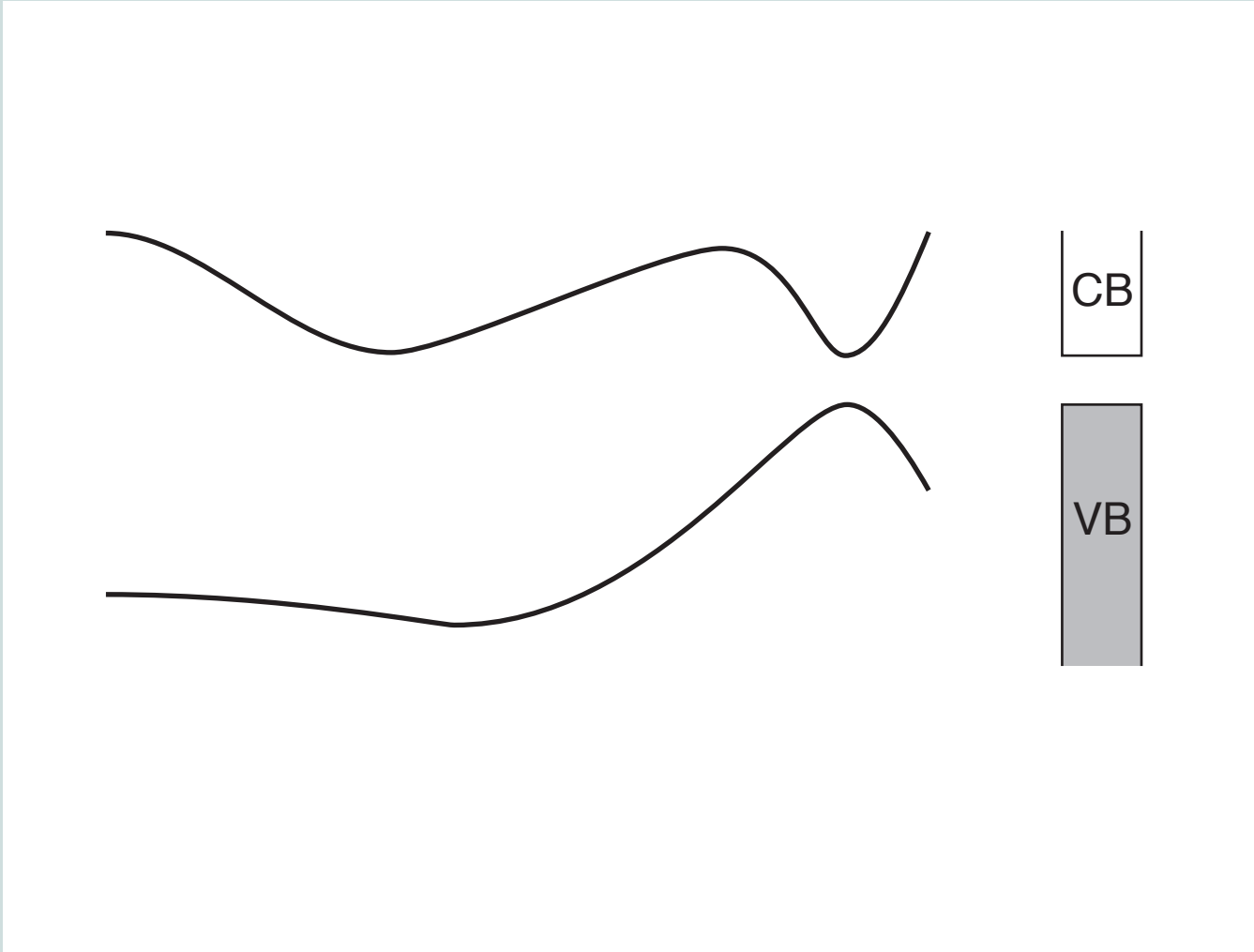
□(□) is not metallic



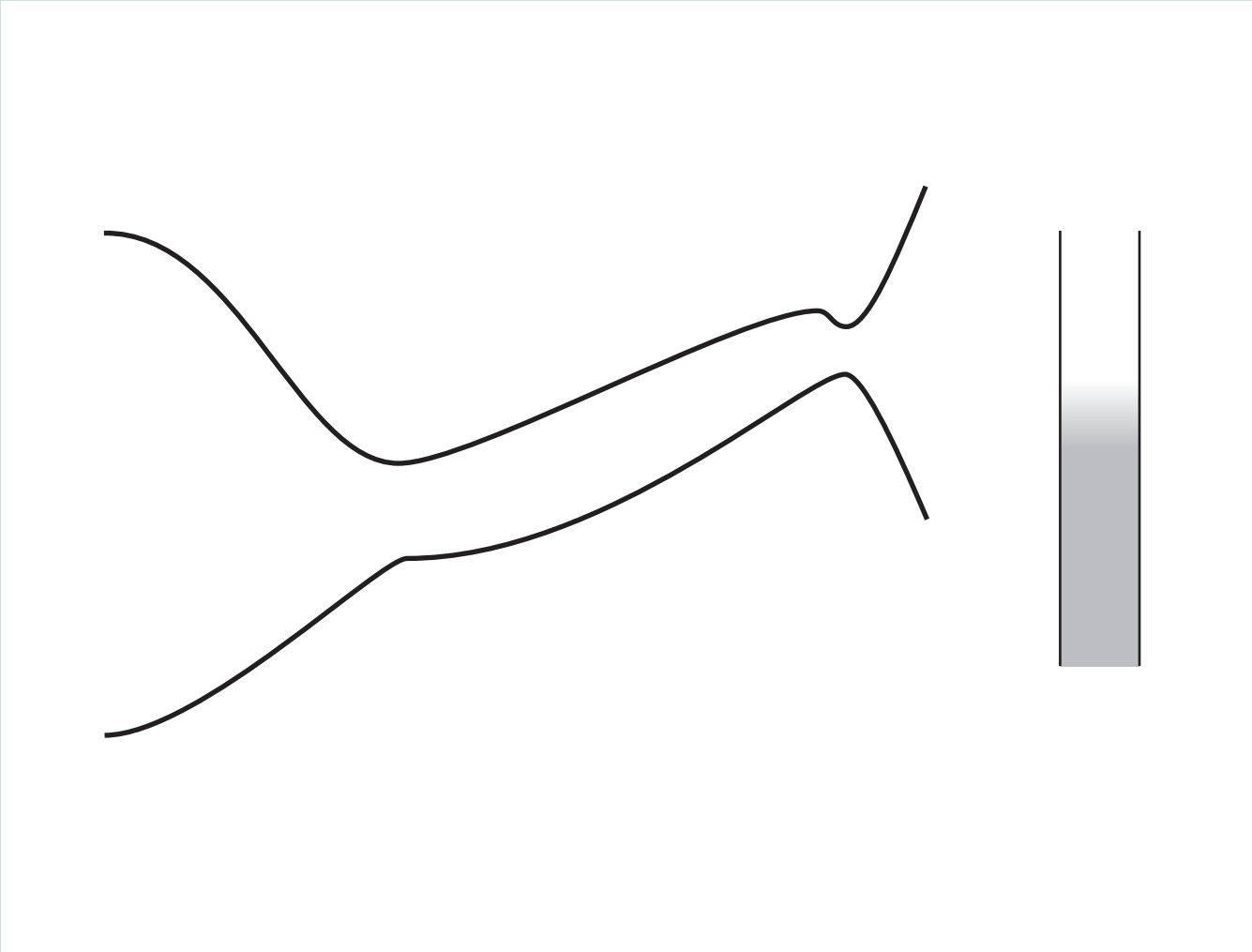
DISCUSSION



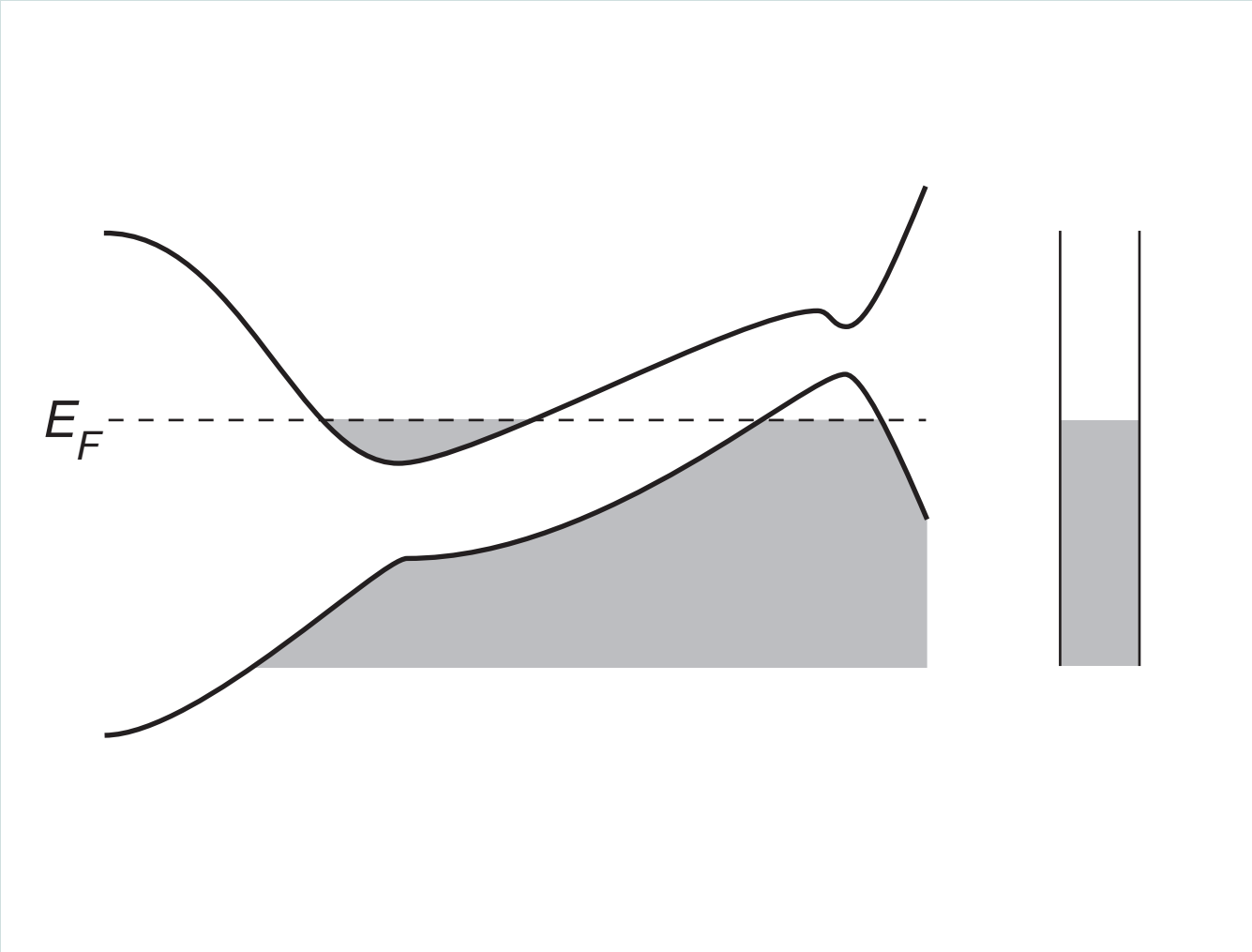
Semiconducting because of 0.3-eV gap



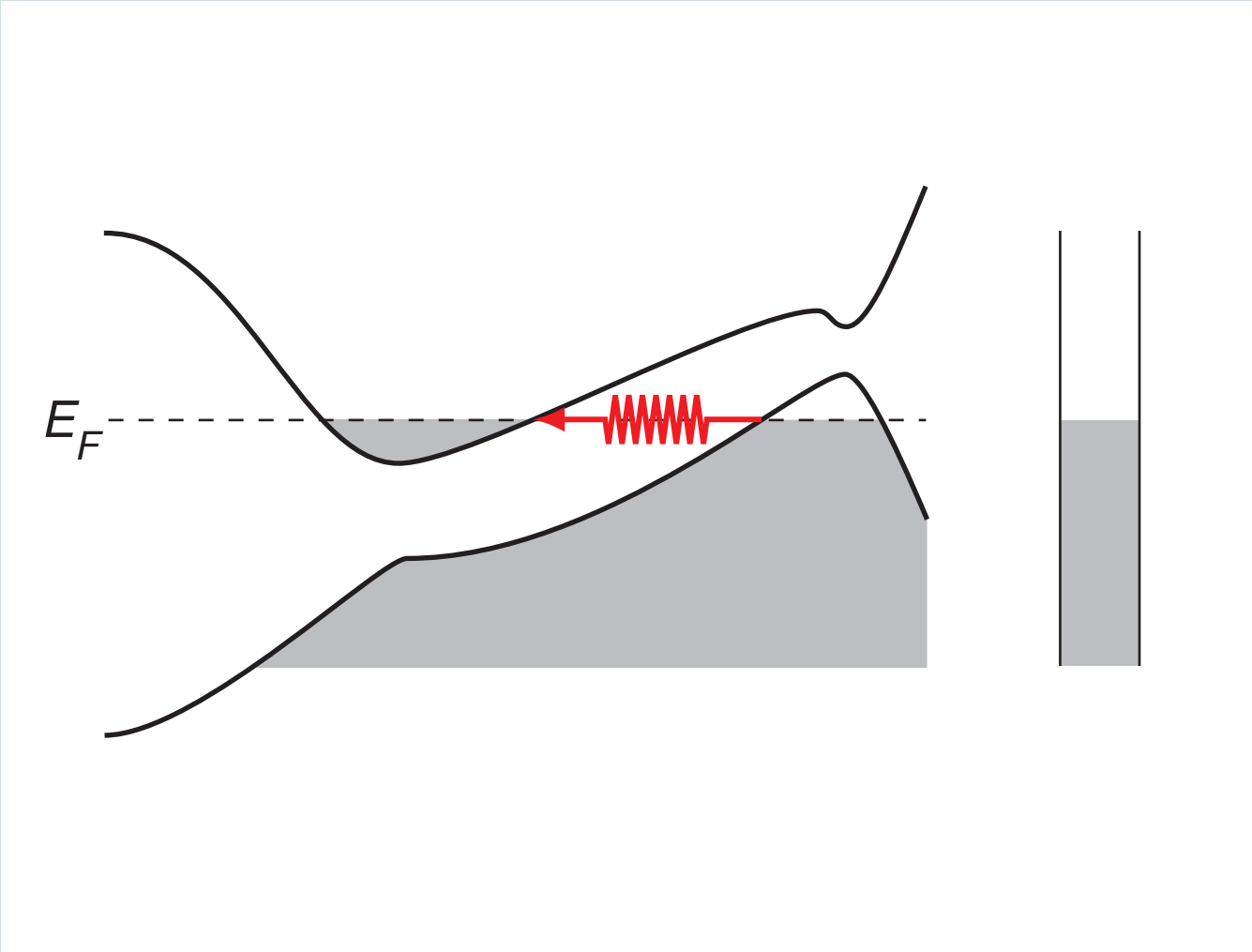
After bands cross...



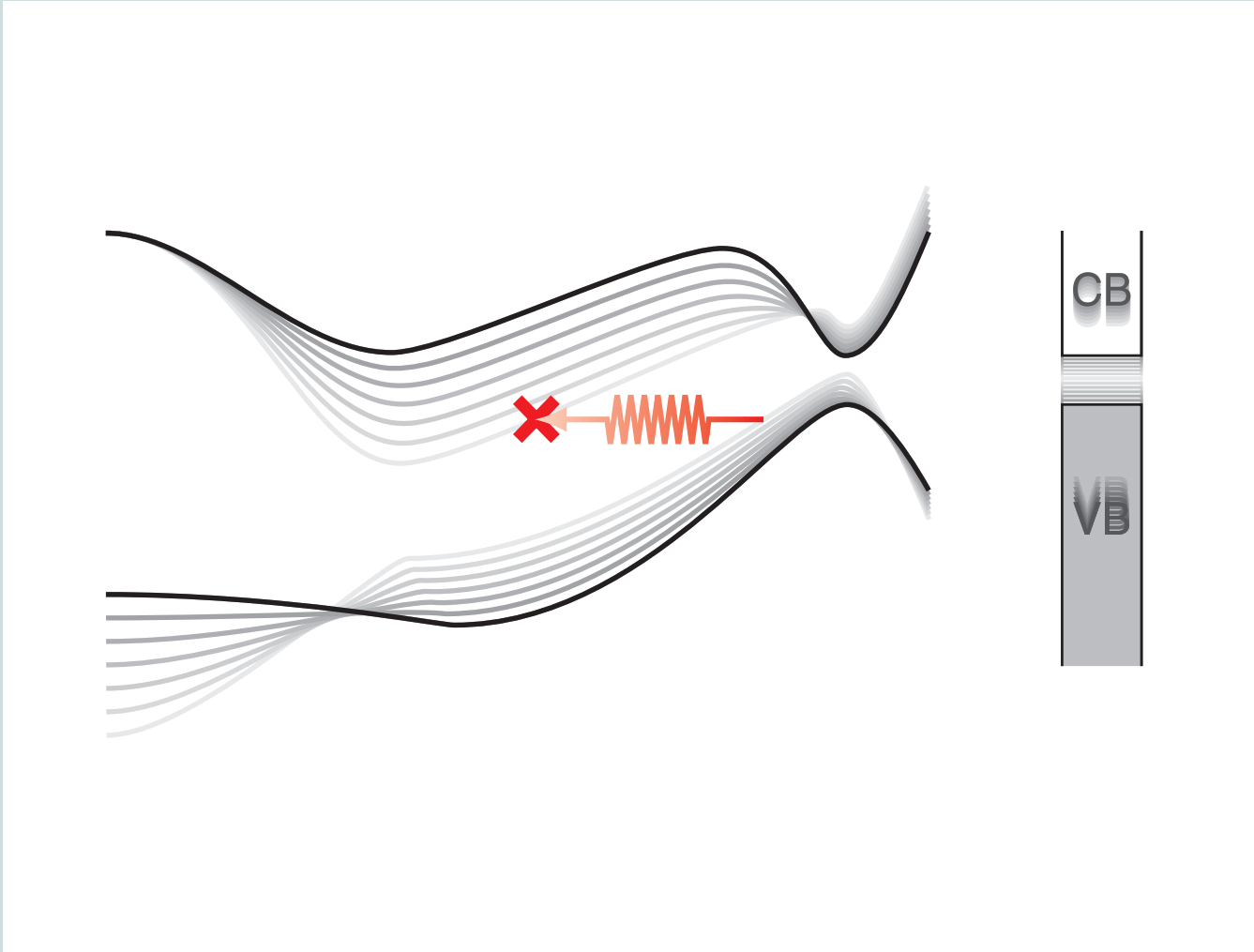
... material can become metallic...



... provided phonons scatter electrons



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



SUMMARY

Coherent phonons modulate dielectric function

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Coherent phonons modulate dielectric function

Evidence for transient band-crossing

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Coherent phonons modulate dielectric function

Evidence for transient band-crossing

... but no metal

Funding: National Science Foundation

Acknowledgments:

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Nick Choly (Harvard University)
Prof. Tim Kaxiras (Harvard University)

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

http://mazur-www.harvard.edu