Ultrafast reflectivity dynamics of highly excited bulk ZnO

Tina Shih, Tobias Voss* and Eric Mazur

School of Engineering & Applied Sciences - Harvard University, USA

*Institute of Solid State Physics - University of Bremen, Germany

Outline

Motivation

Pump-probe reflectometry

Ultrafast dielectric function dynamics

Summary

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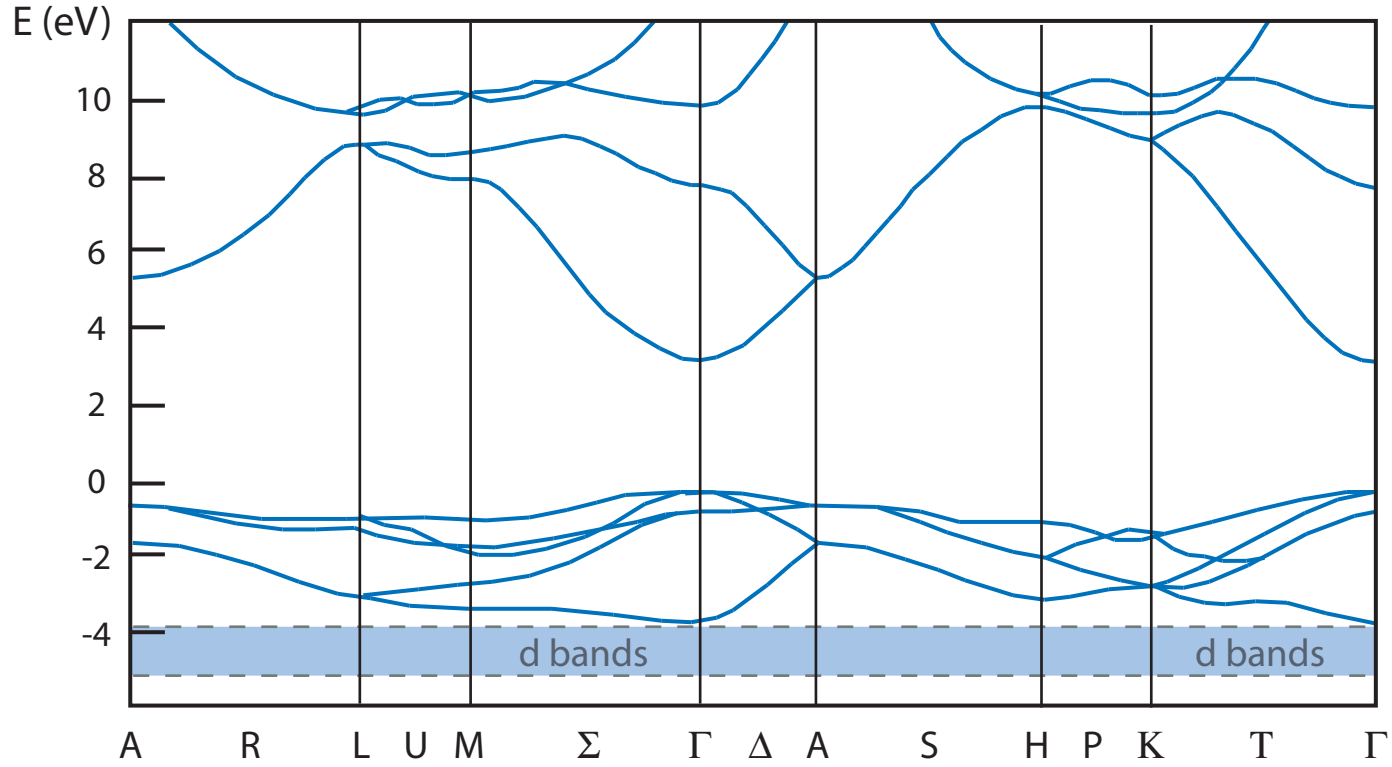
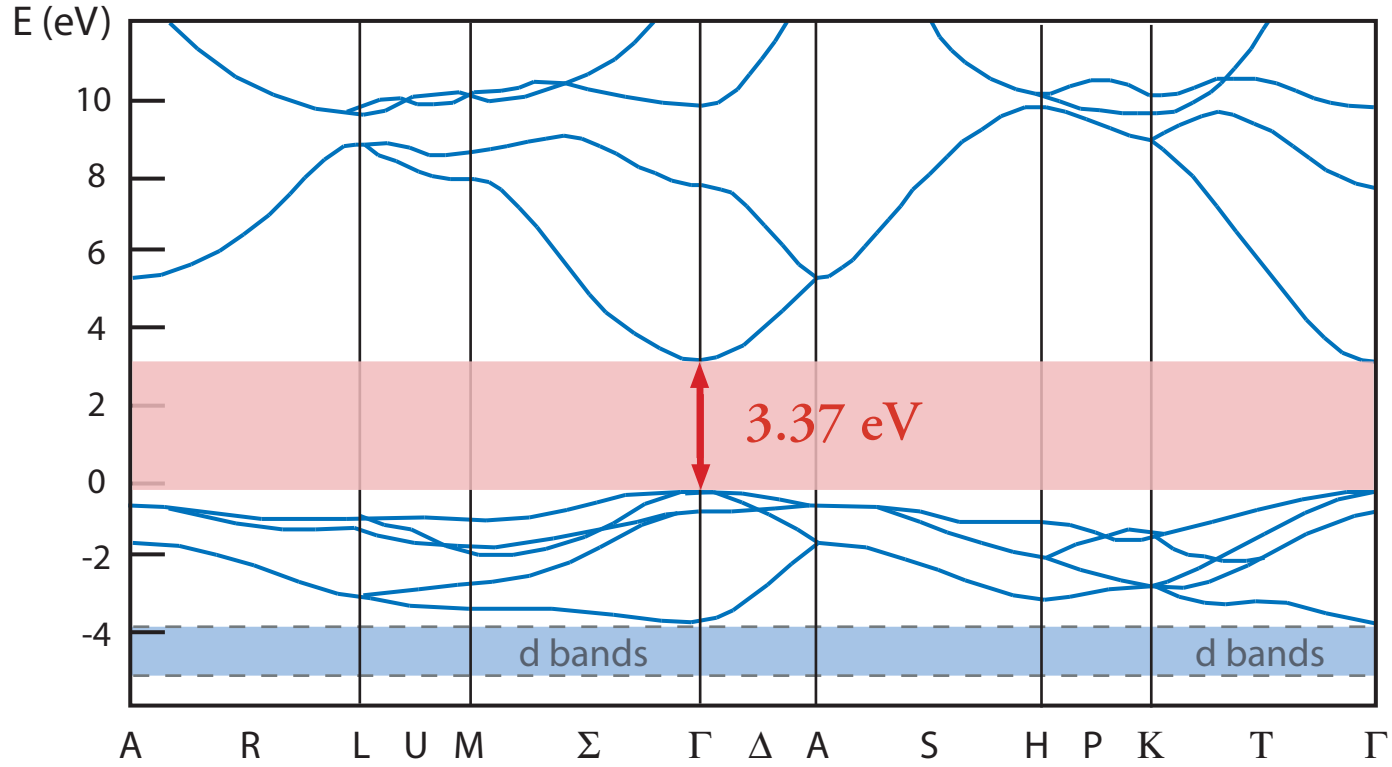
Summary

ZnO Bandstructure



after U. Rossler, *Phys. Rev.* **184**, 733-738 (1969)

ZnO Bandstructure



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Mazur Group

Interaction between laser light and solid materials

Motivation

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Femtosecond laser pulses

Semiconducting materials

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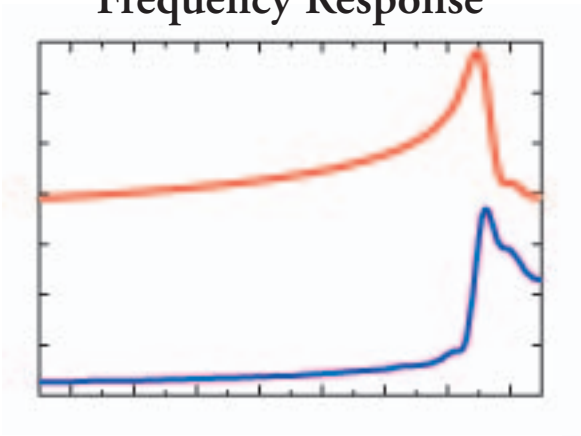
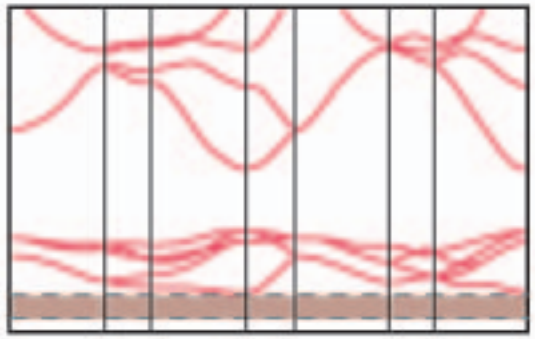
Dielectric Function

Motivation

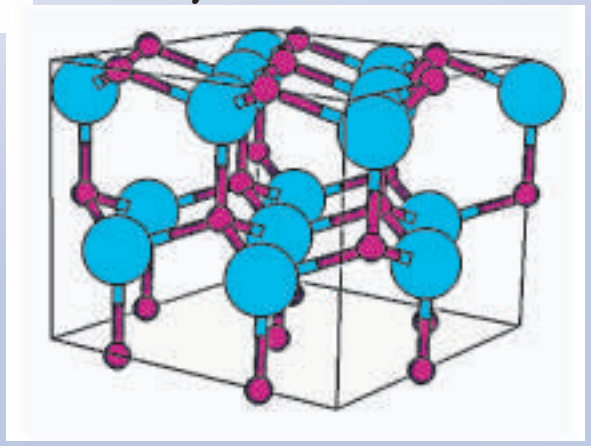
Frequency Response



Bandstructure

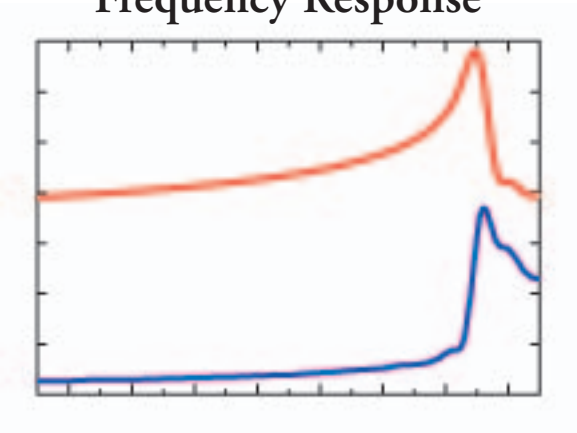


Crystal Structure



Motivation

Frequency Response



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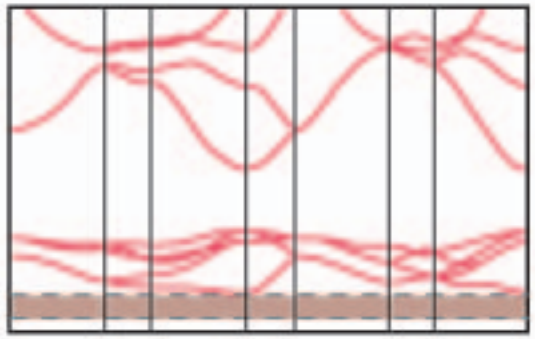

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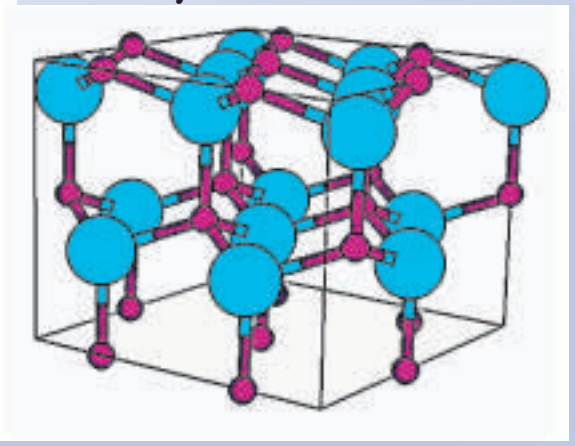
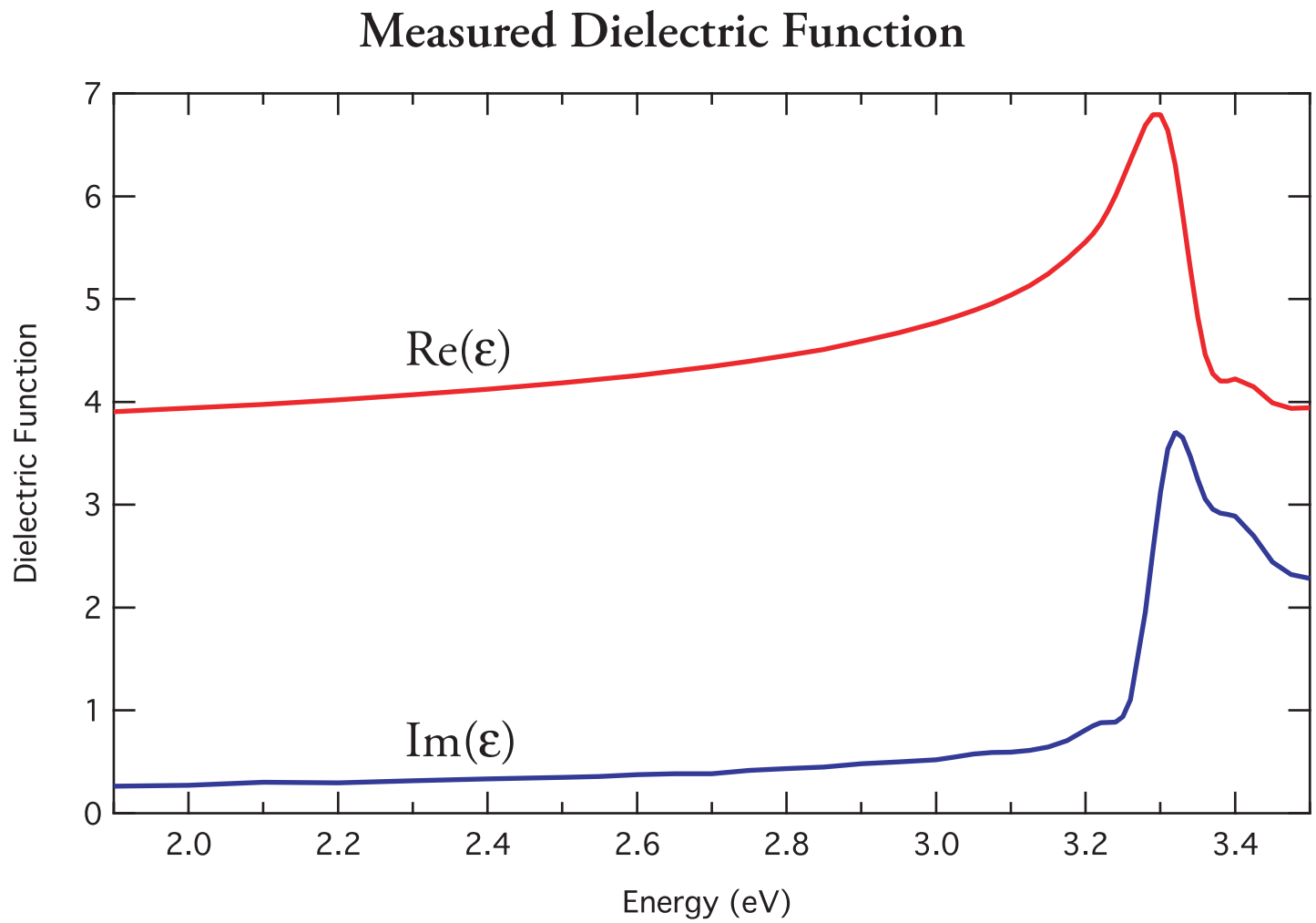


Motivation


Measured Dielectric Function
Dielectric Function
Energy (eV)
Re(ε)
Im(ε)
0
1
2
3
4
5
6
7
2.0
2.2
2.4
2.6
2.8
3.0
3.2
3.4

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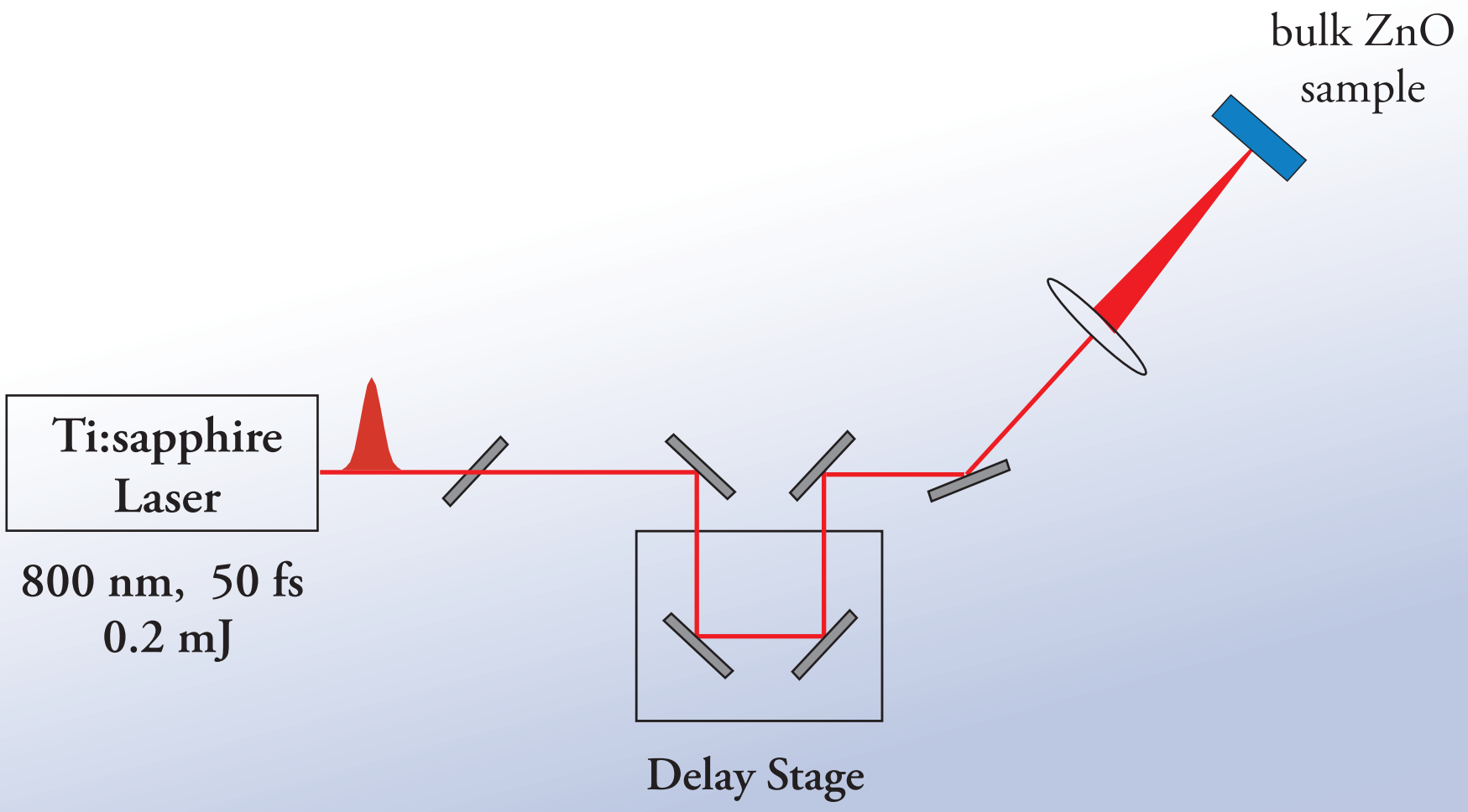
Pump-Probe Reflectometry

Ti:sapphire
Laser

800 nm, 50 fs
0.2 mJ

Pump-Probe Reflectometry



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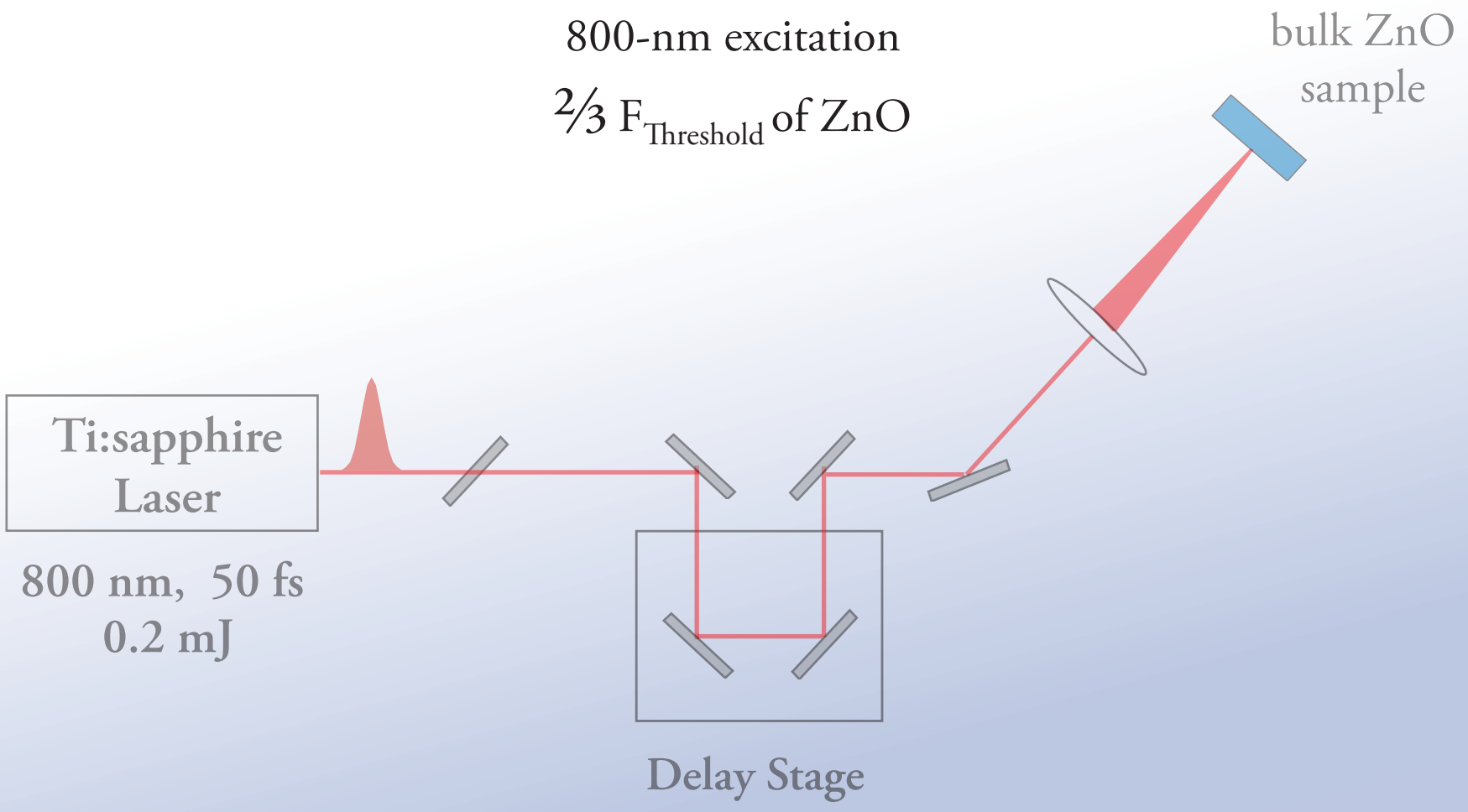
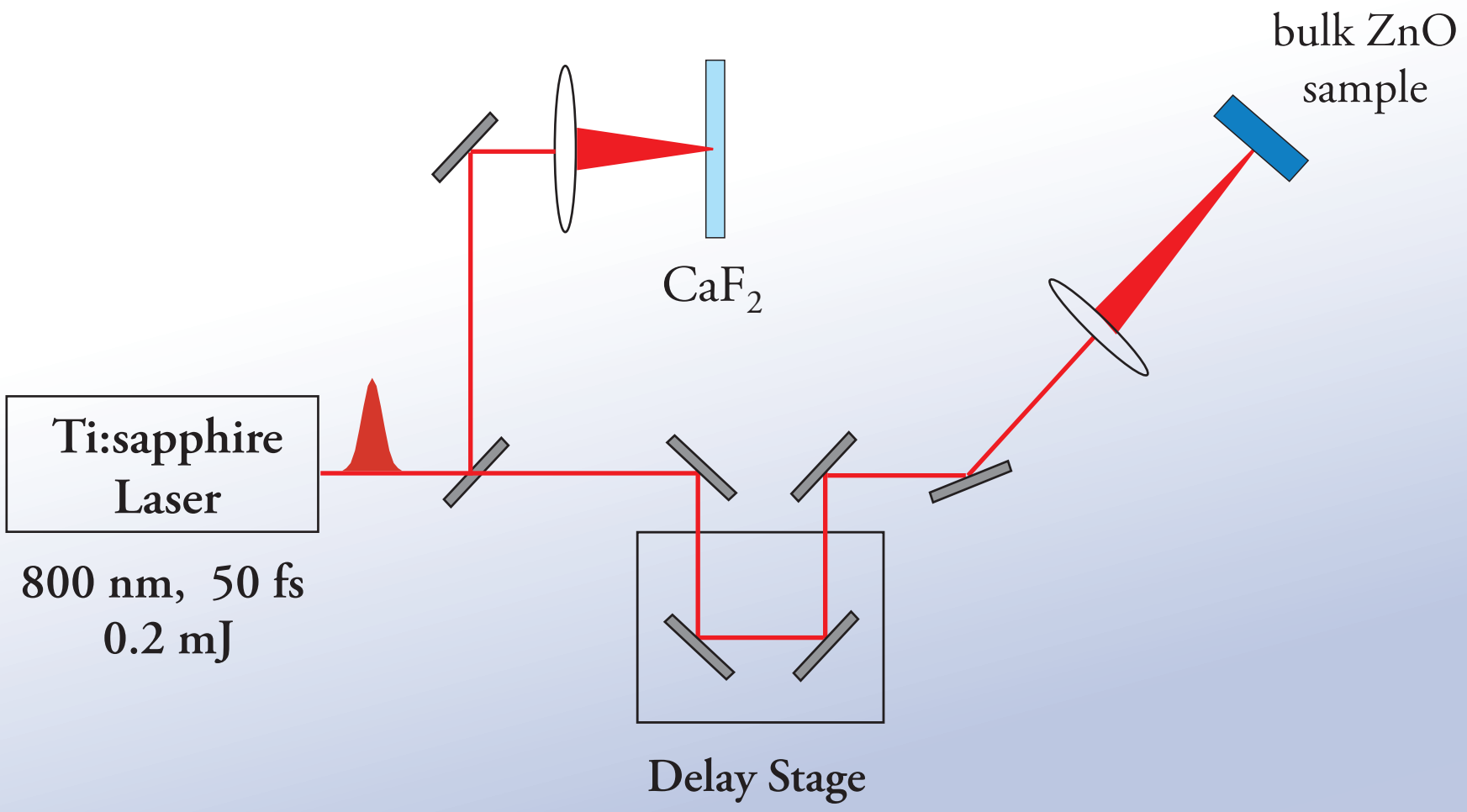
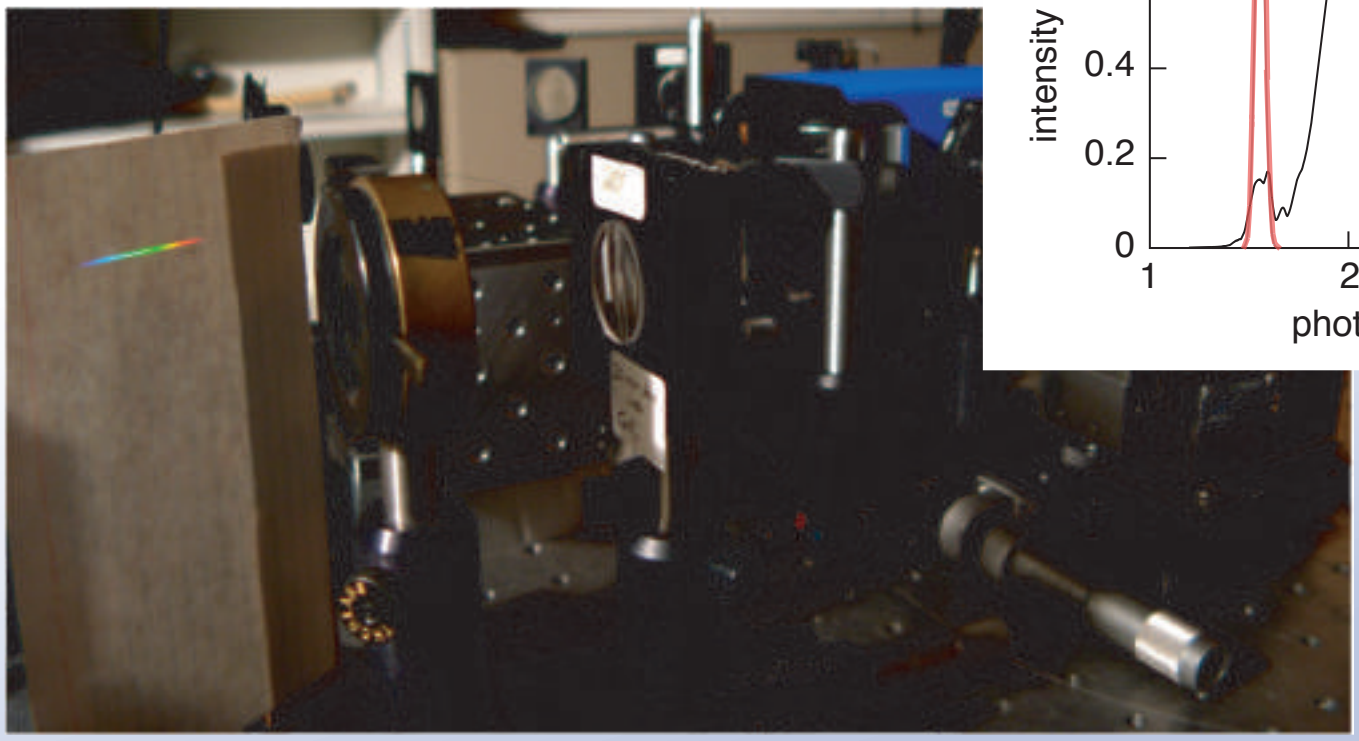


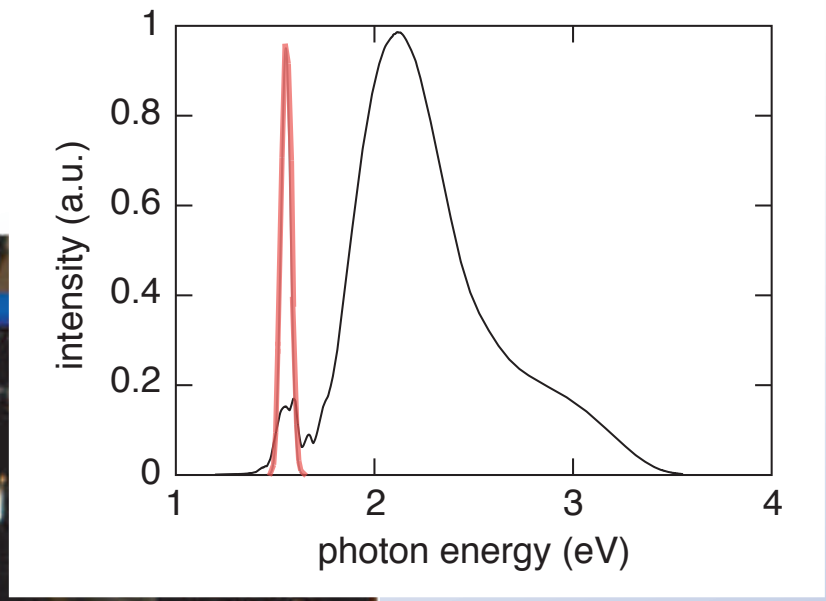
Pump-Probe Reflectometry



Pump-Probe Reflectometry

Femtosecond white light generation

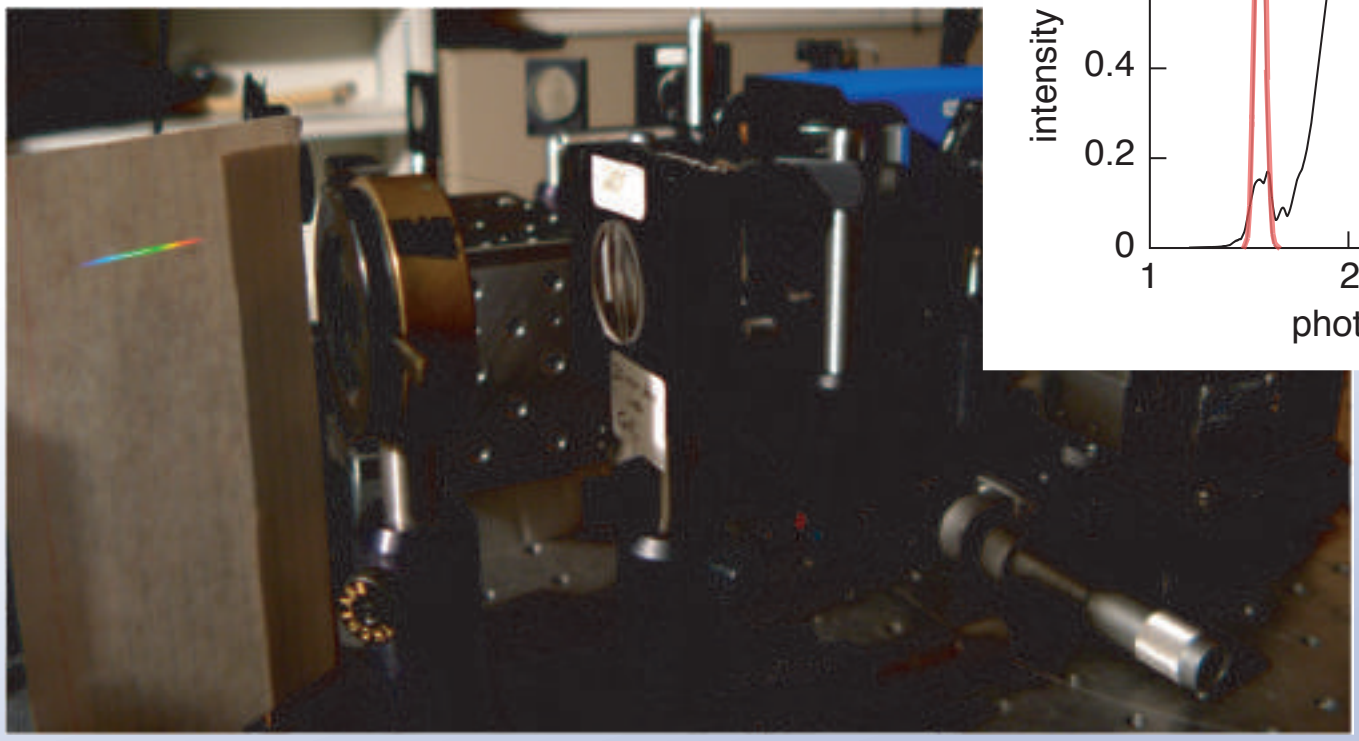


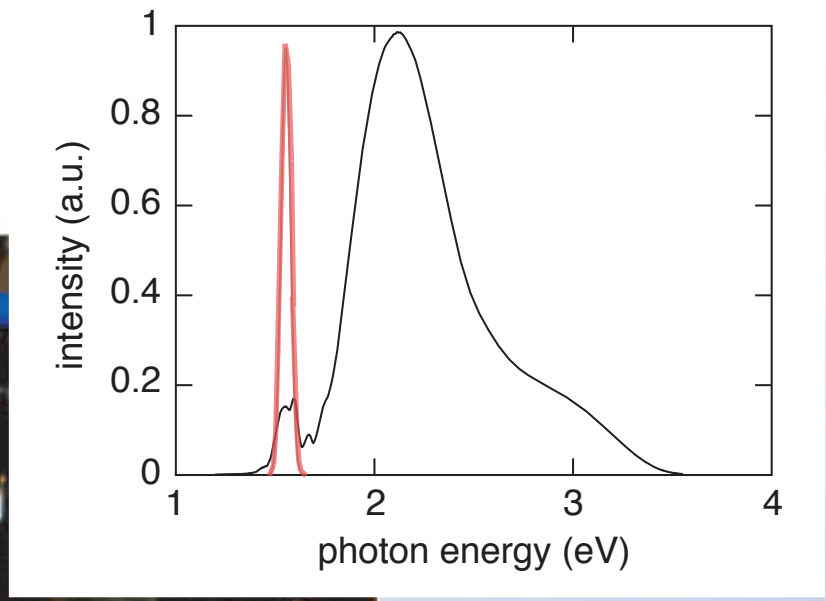


Pump-Probe Reflectometry

Femtosecond white light generation

broadband pulse duration = 150 fs





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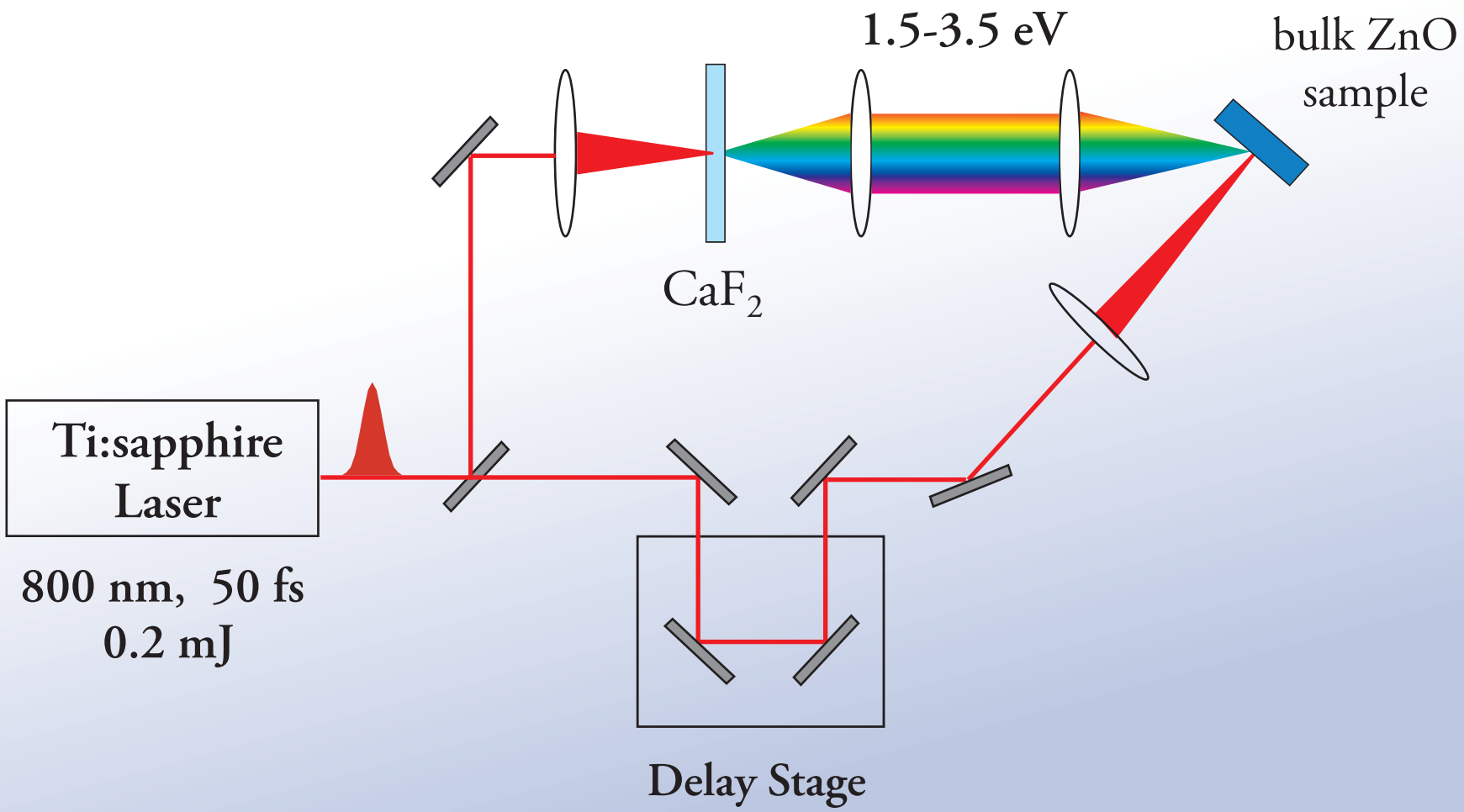

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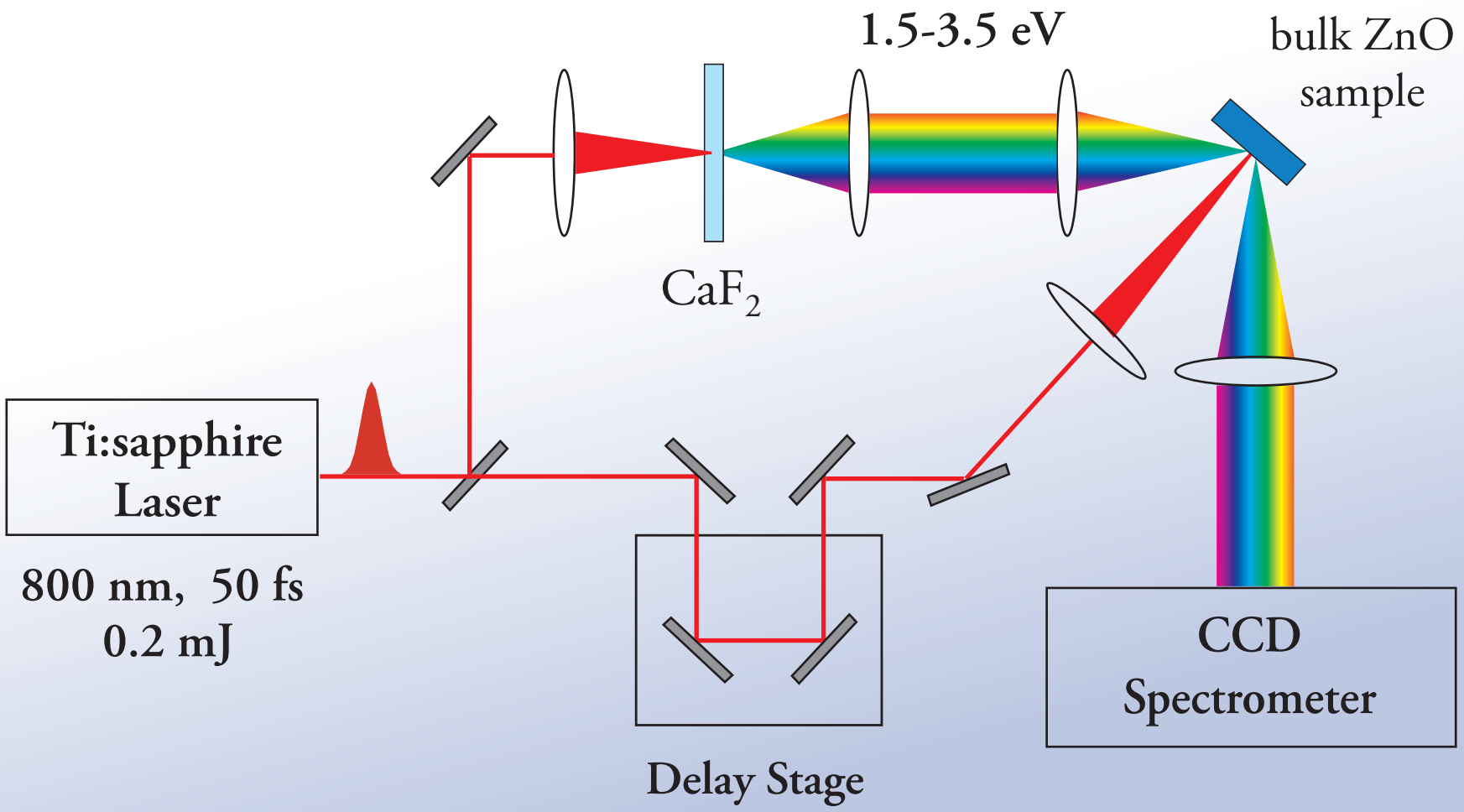


Pump-Probe Reflectometry

Room temperature measurements

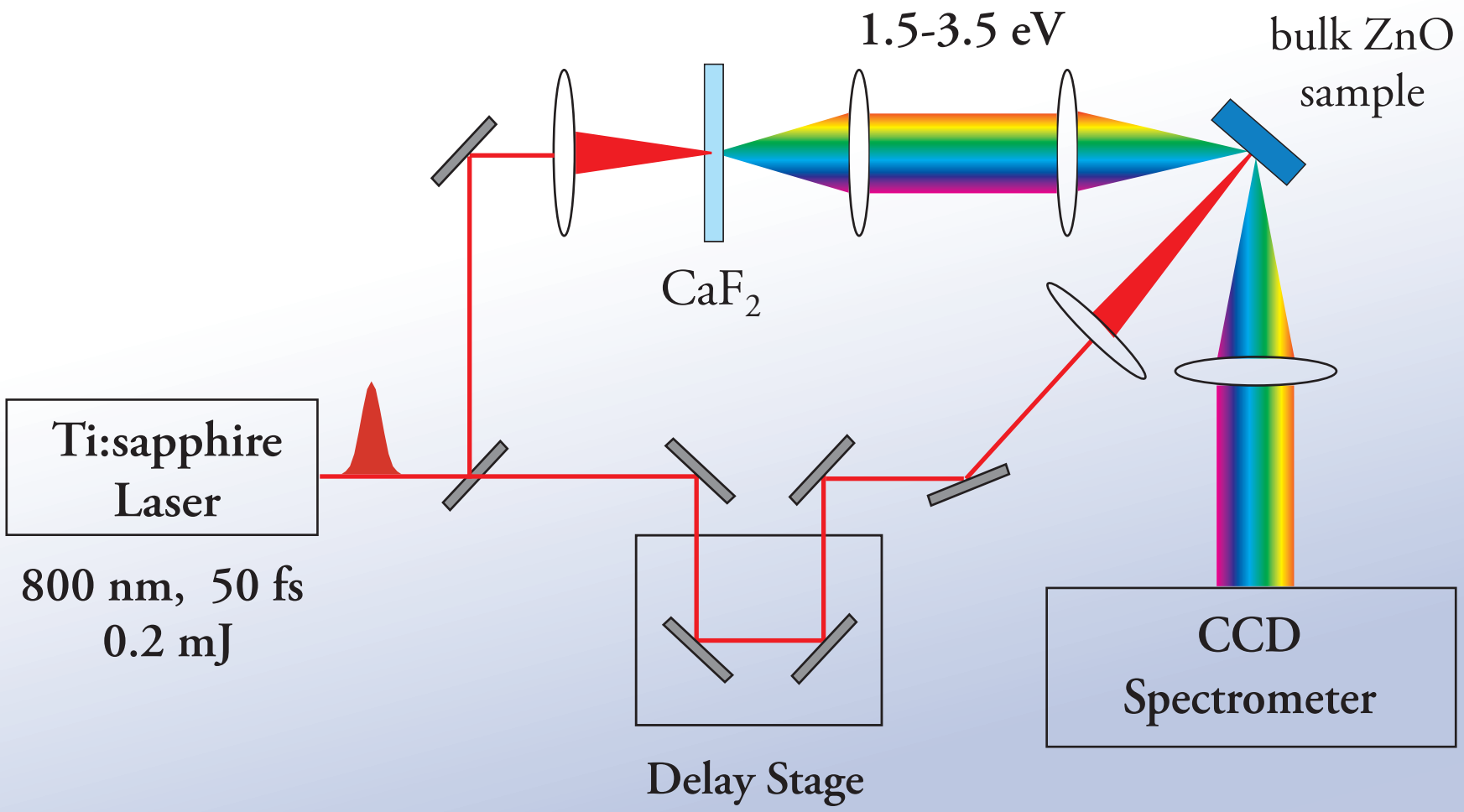
800-nm pump excitation

Femtosecond broadband probe

Ultrafast time resolution

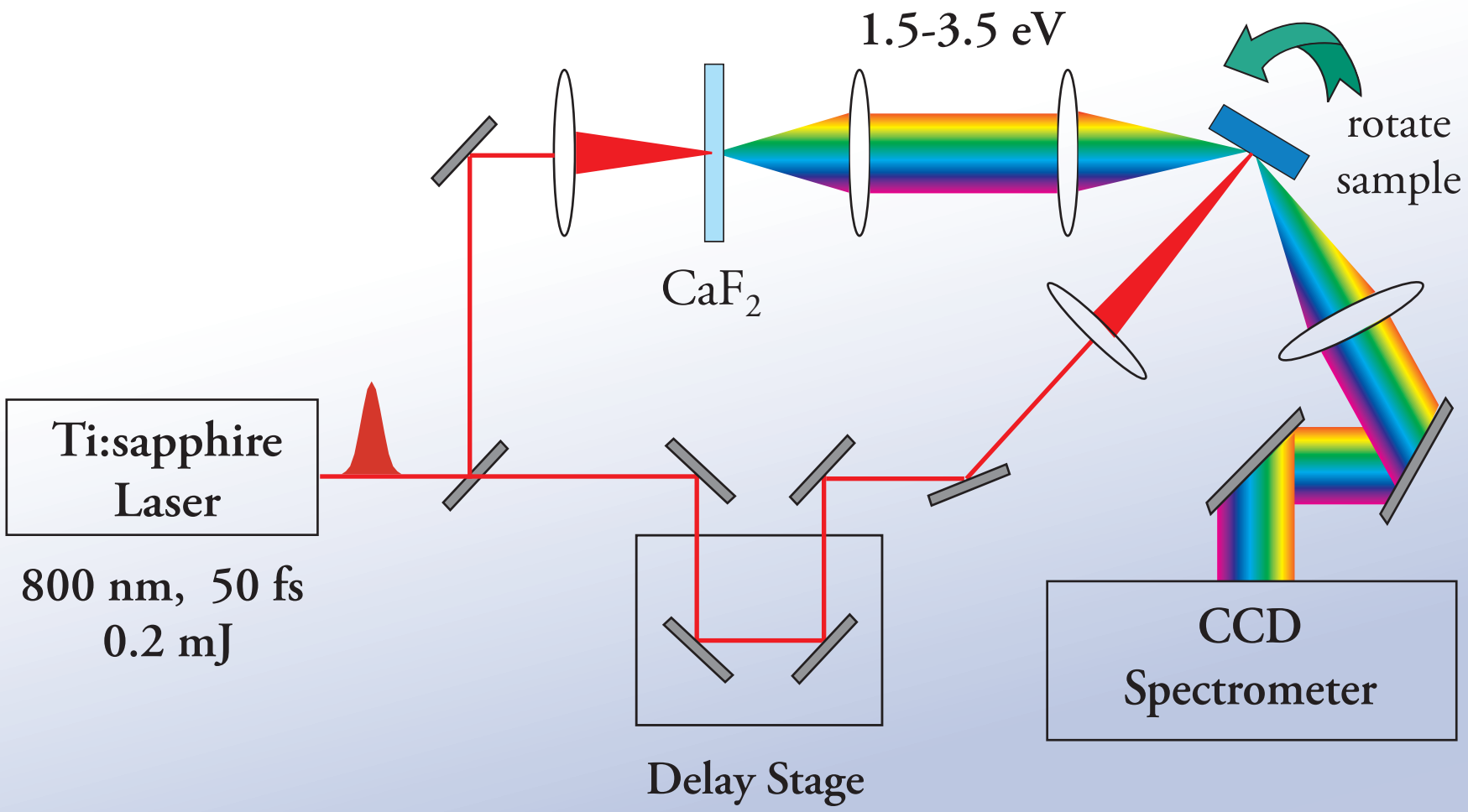
Dual-angle reflectivity measurements

Pump-Probe Reflectometry



Pump-Probe Reflectometry

Broadband dual-angle reflectometry



Fresnel Equations

$$r_p = \frac{\epsilon \cos\theta - \sqrt{\epsilon_0}\sqrt{\epsilon - \epsilon_0 \sin^2\theta}}{\epsilon \cos\theta + \sqrt{\epsilon_0}\sqrt{\epsilon - \epsilon_0 \sin^2\theta}}$$

$$r_s = \frac{\sqrt{\epsilon_0}\cos\theta - \sqrt{\epsilon - \epsilon_0 \sin^2\theta}}{\sqrt{\epsilon_0}\cos\theta + \sqrt{\epsilon - \epsilon_0 \sin^2\theta}}$$

Fresnel Equations

$$\{ R(\omega, \theta_1), R(\omega, \theta_2) \}$$

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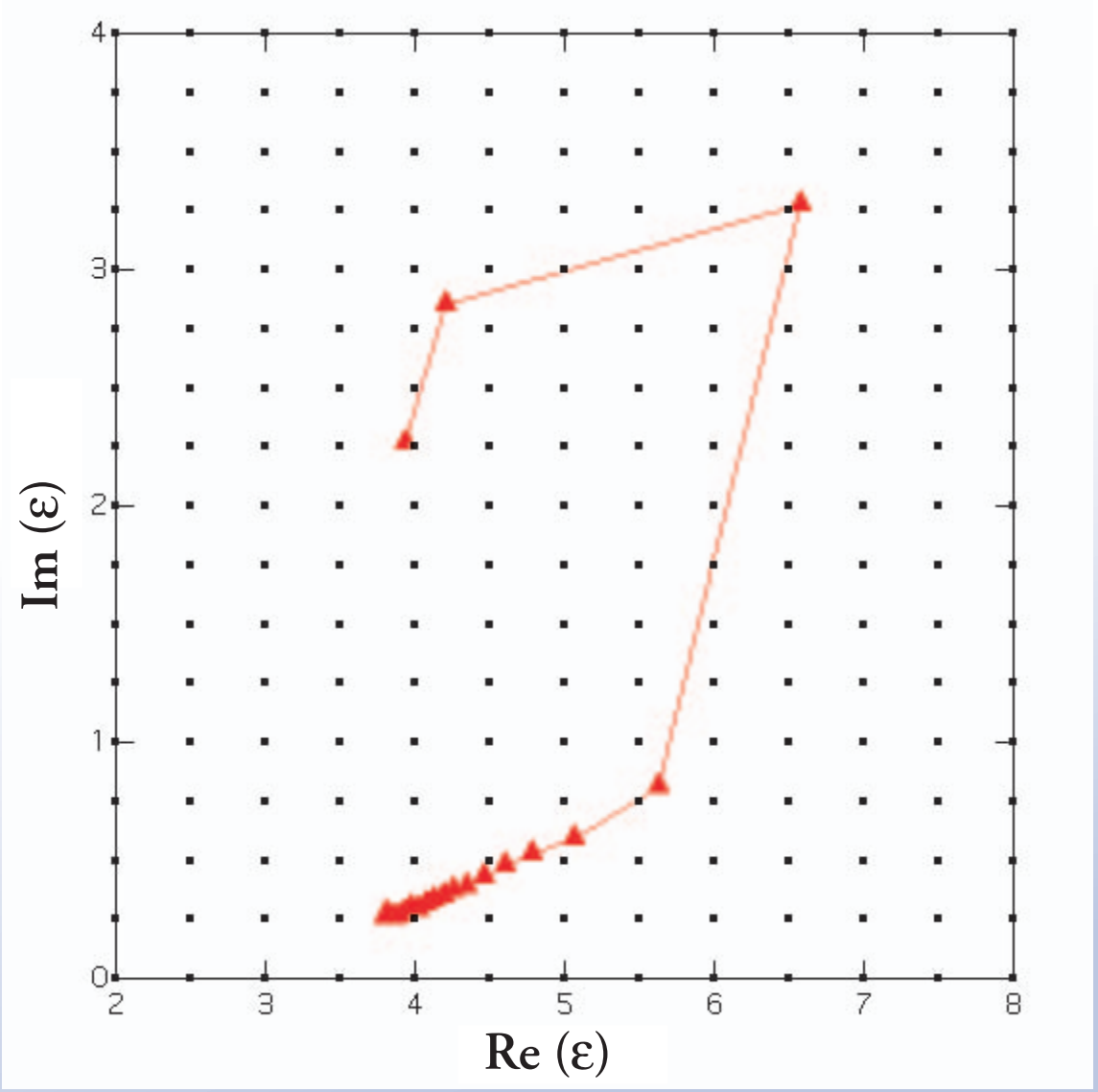
$$r_s = \frac{\sqrt{\epsilon_0}\cos\theta - \sqrt{\epsilon - \epsilon_0 \sin^2\theta}}{\sqrt{\epsilon_0}\cos\theta + \sqrt{\epsilon - \epsilon_0 \sin^2\theta}}$$

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

Fresnel Equations



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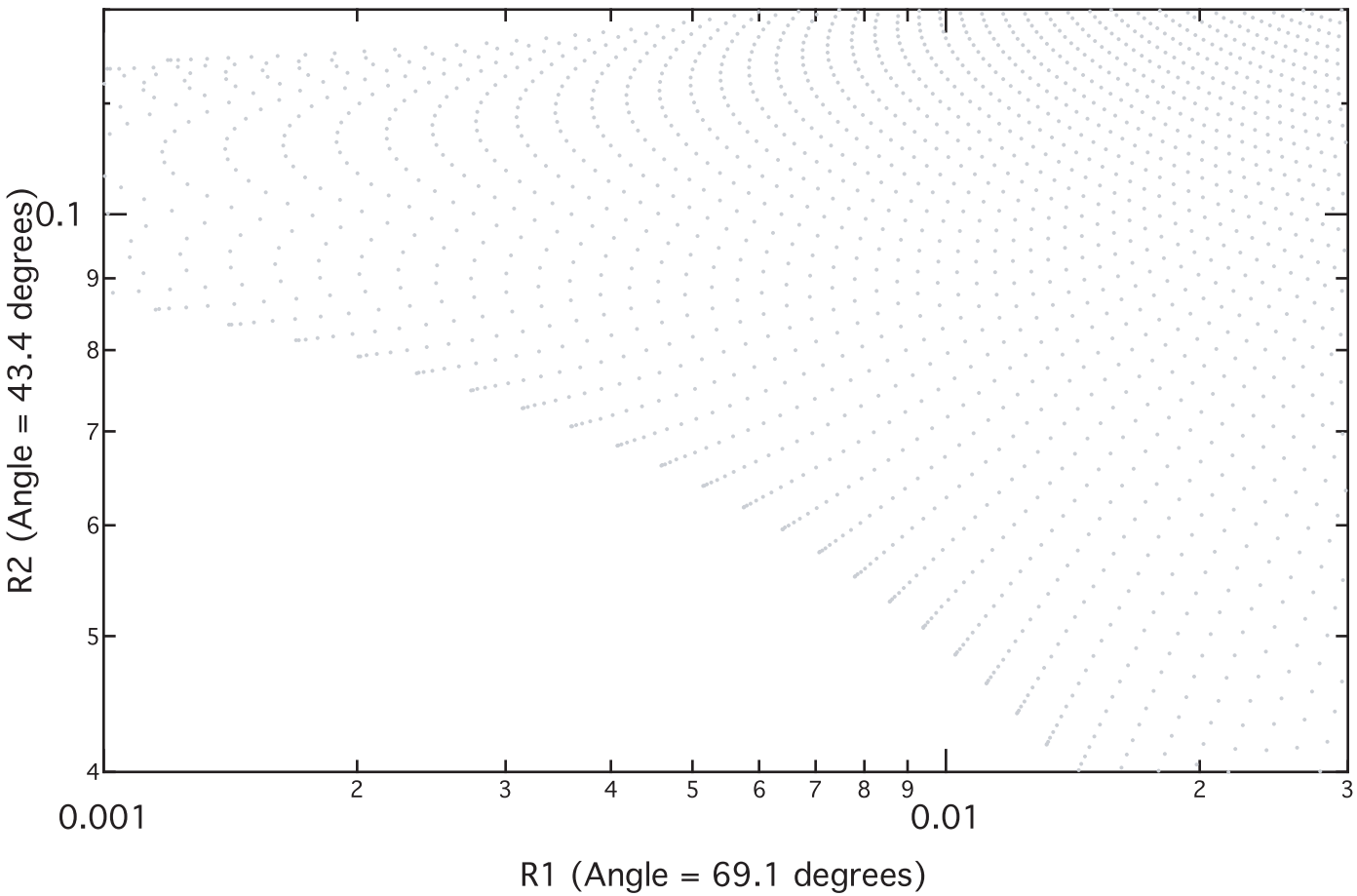
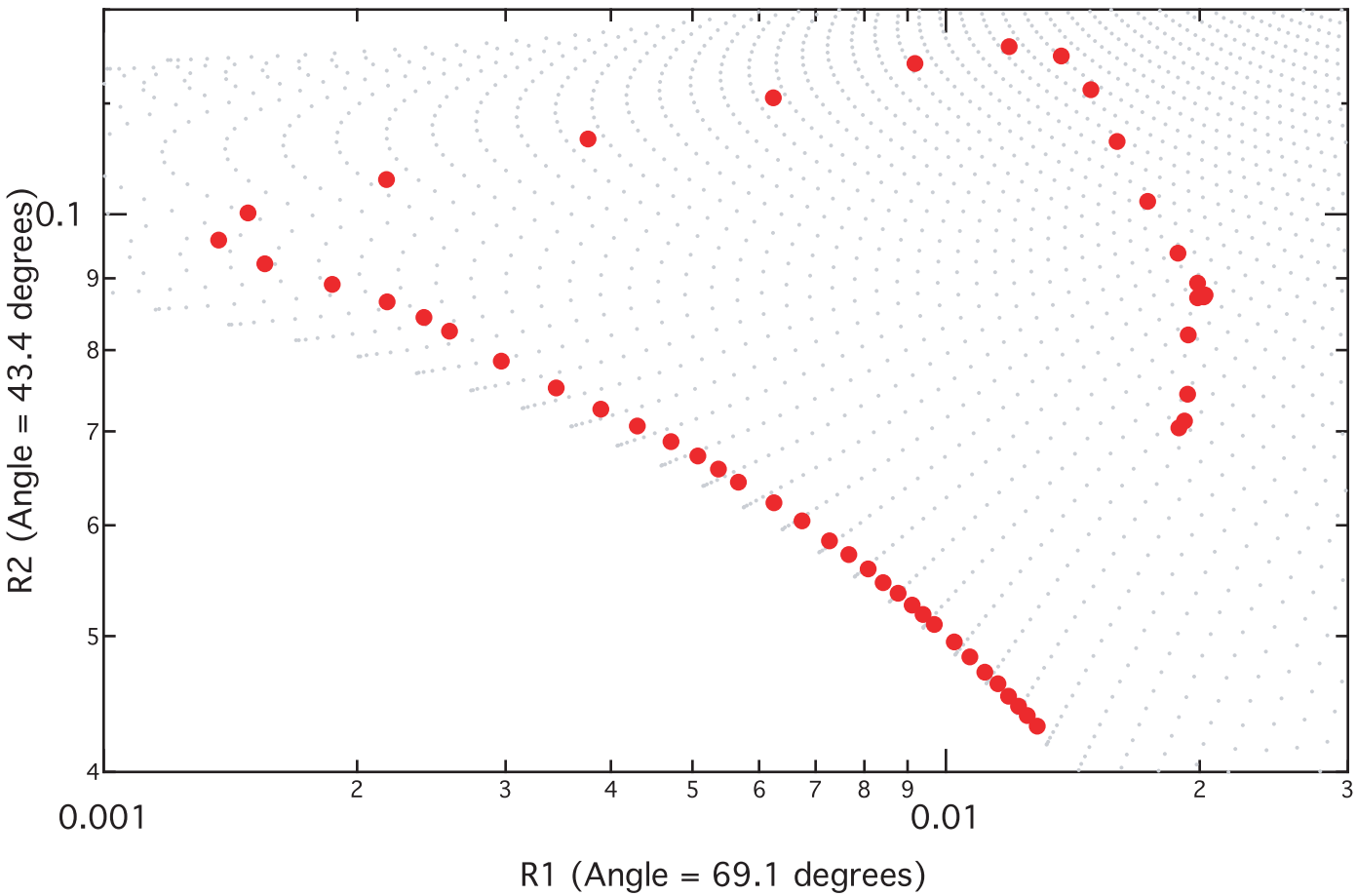
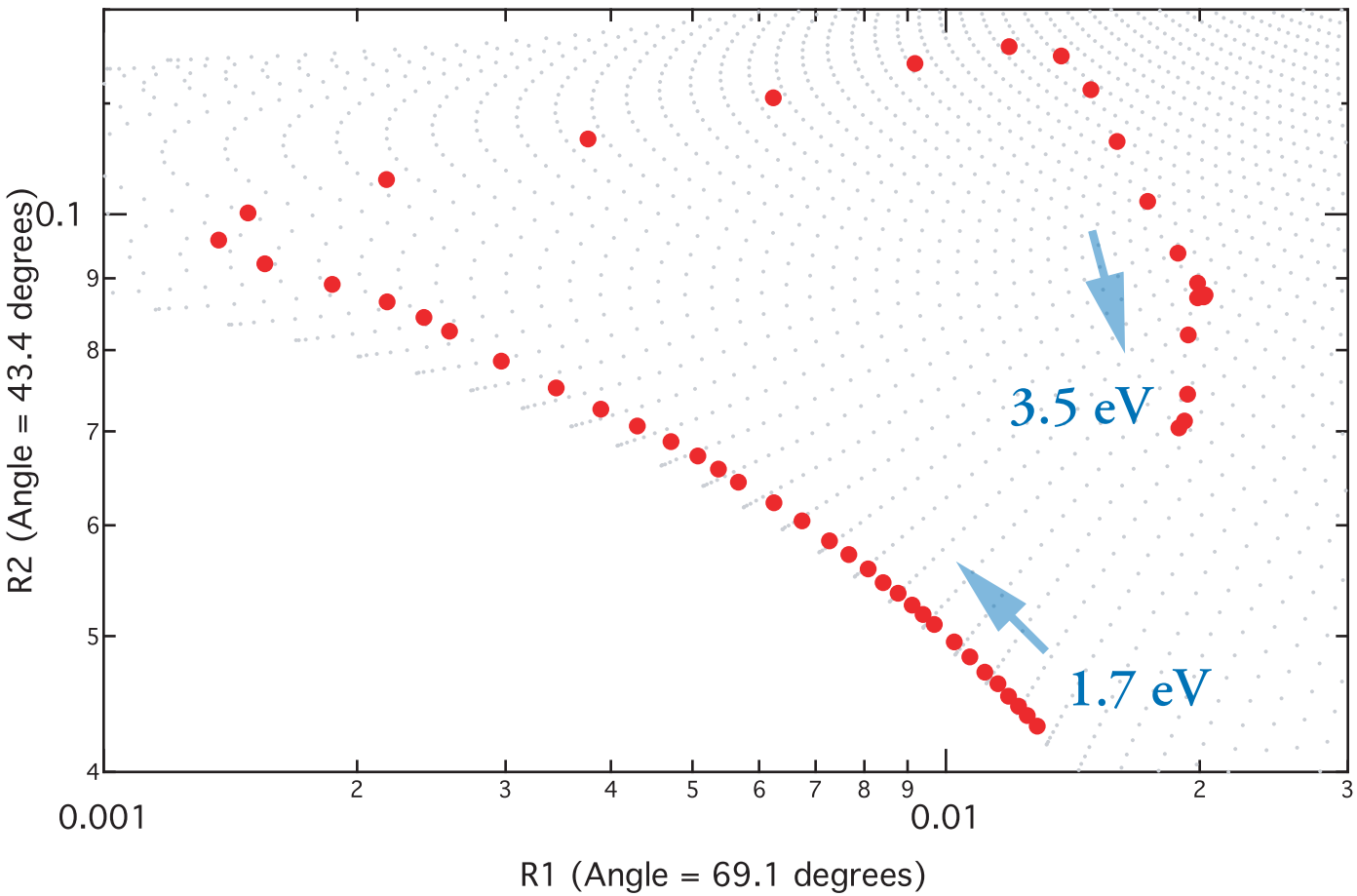


Fresnel Equations



Fresnel Equations



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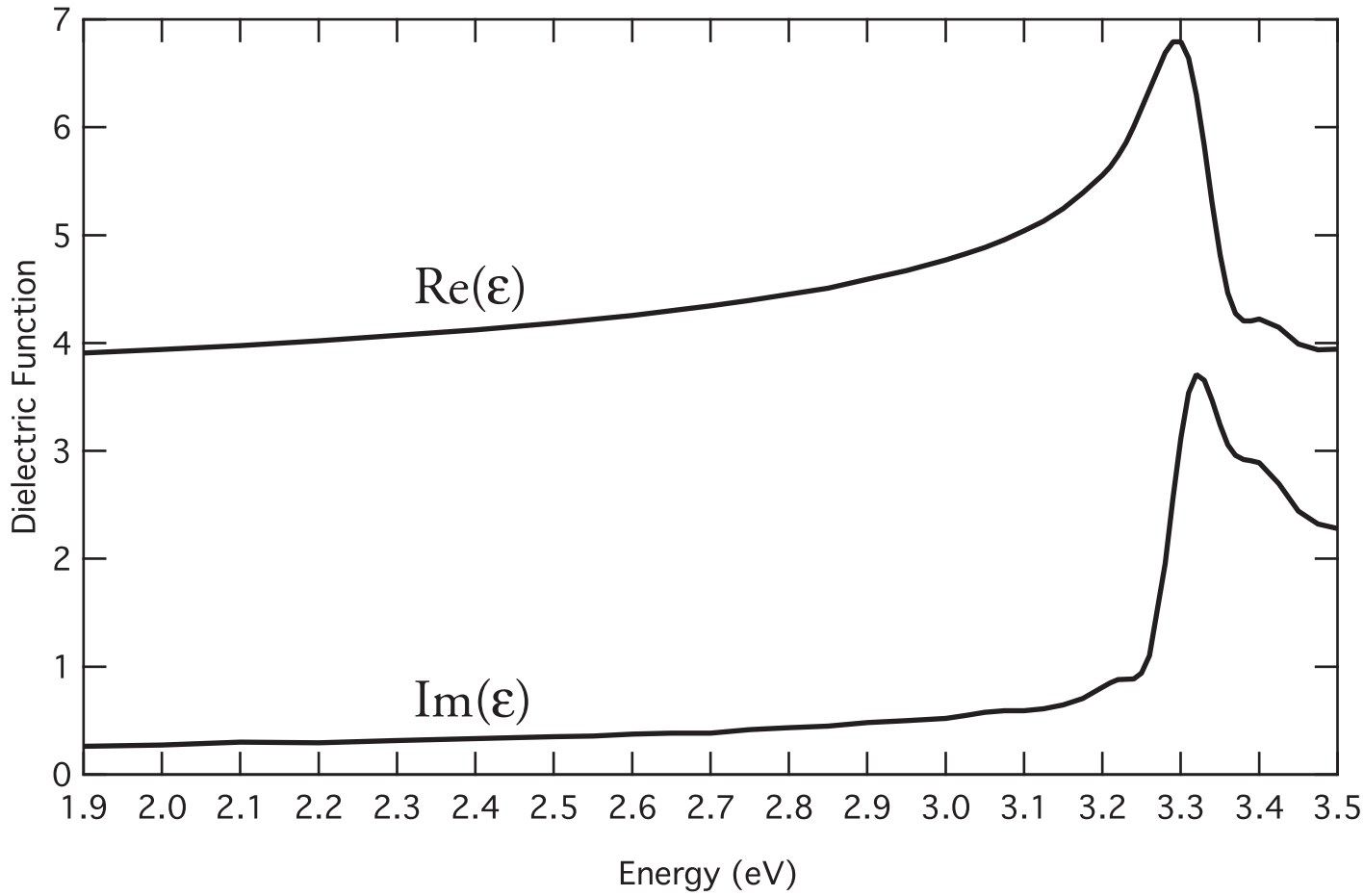
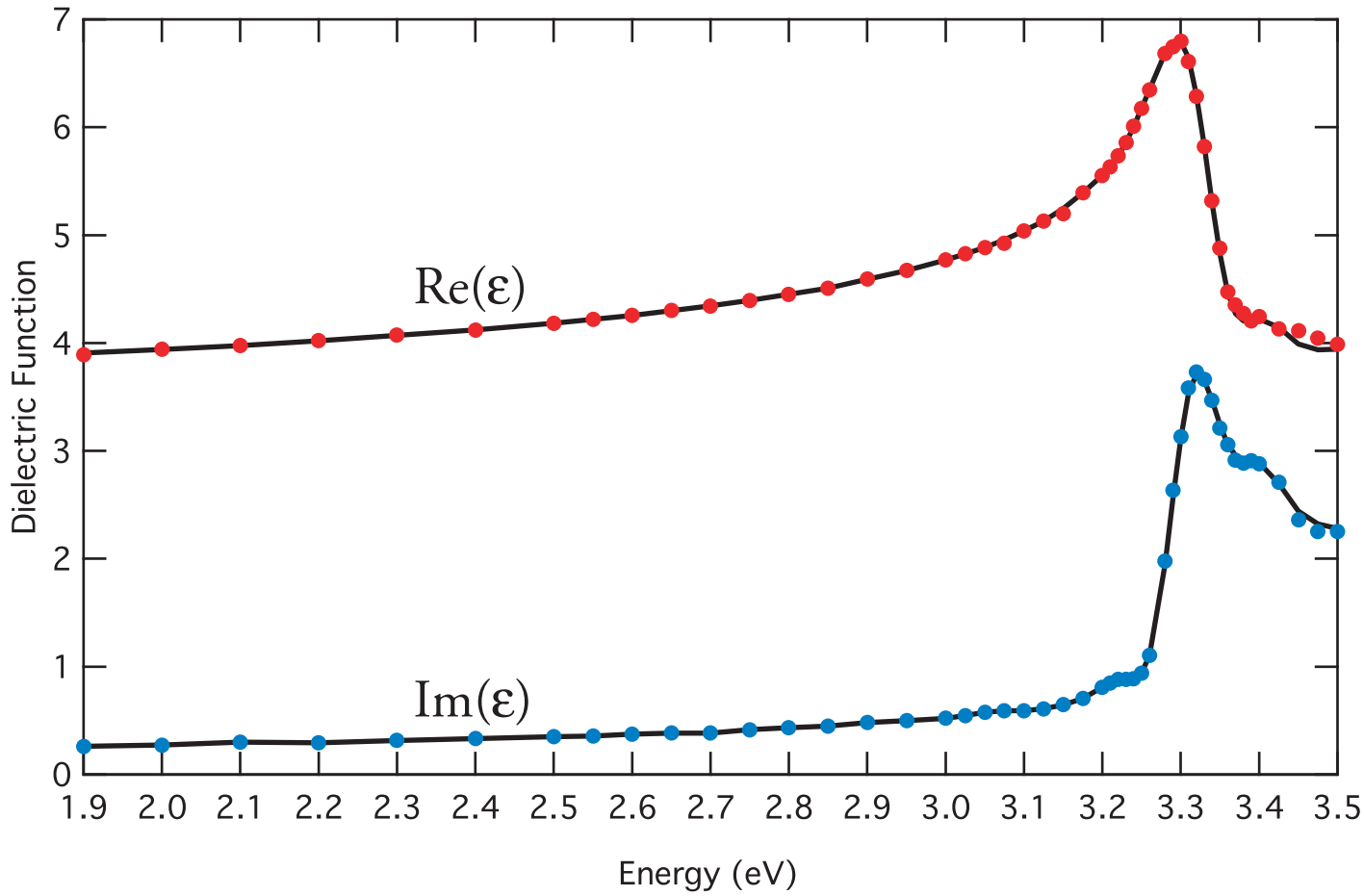


Ultrafast dielectric function dynamics



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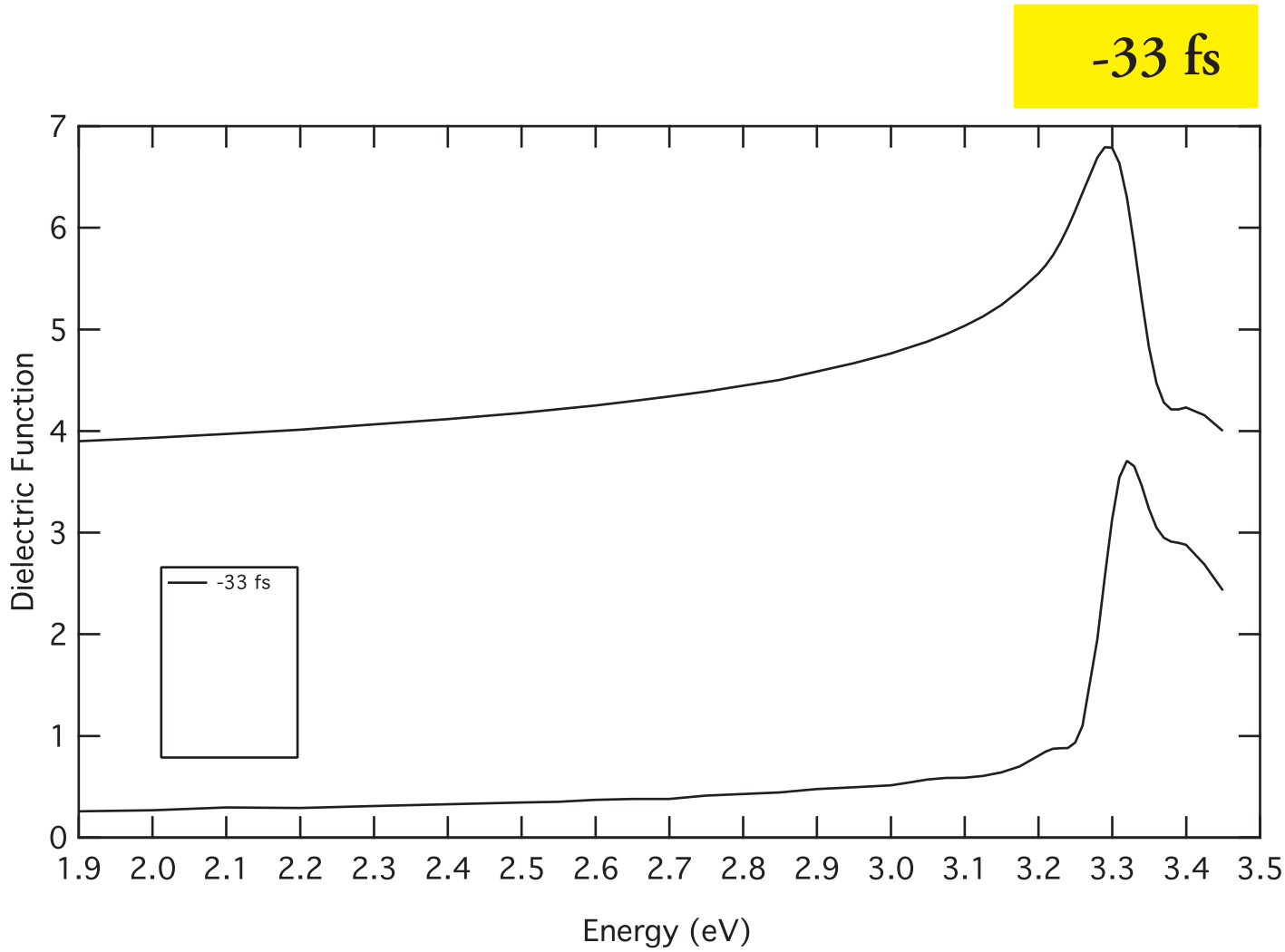
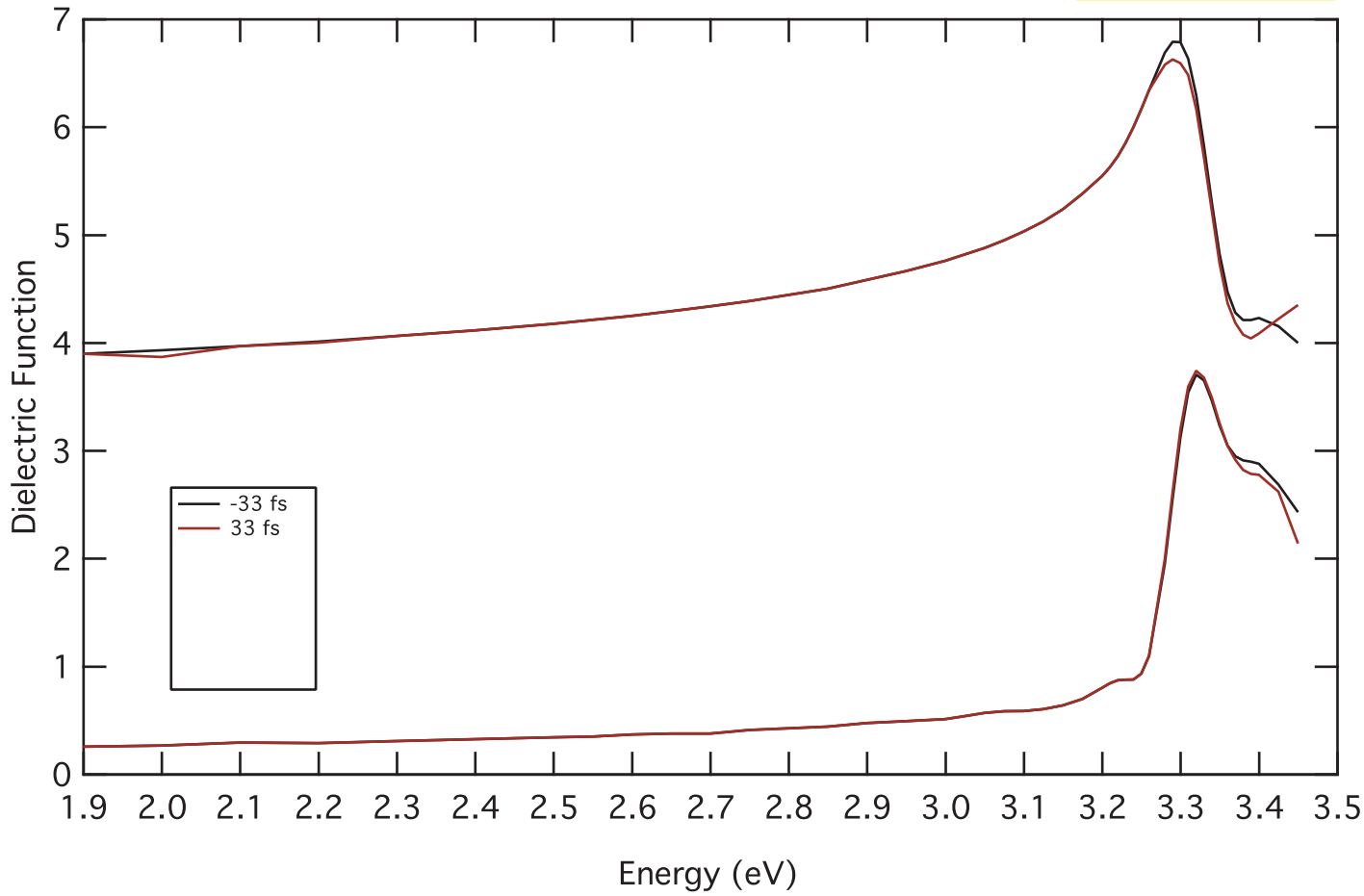
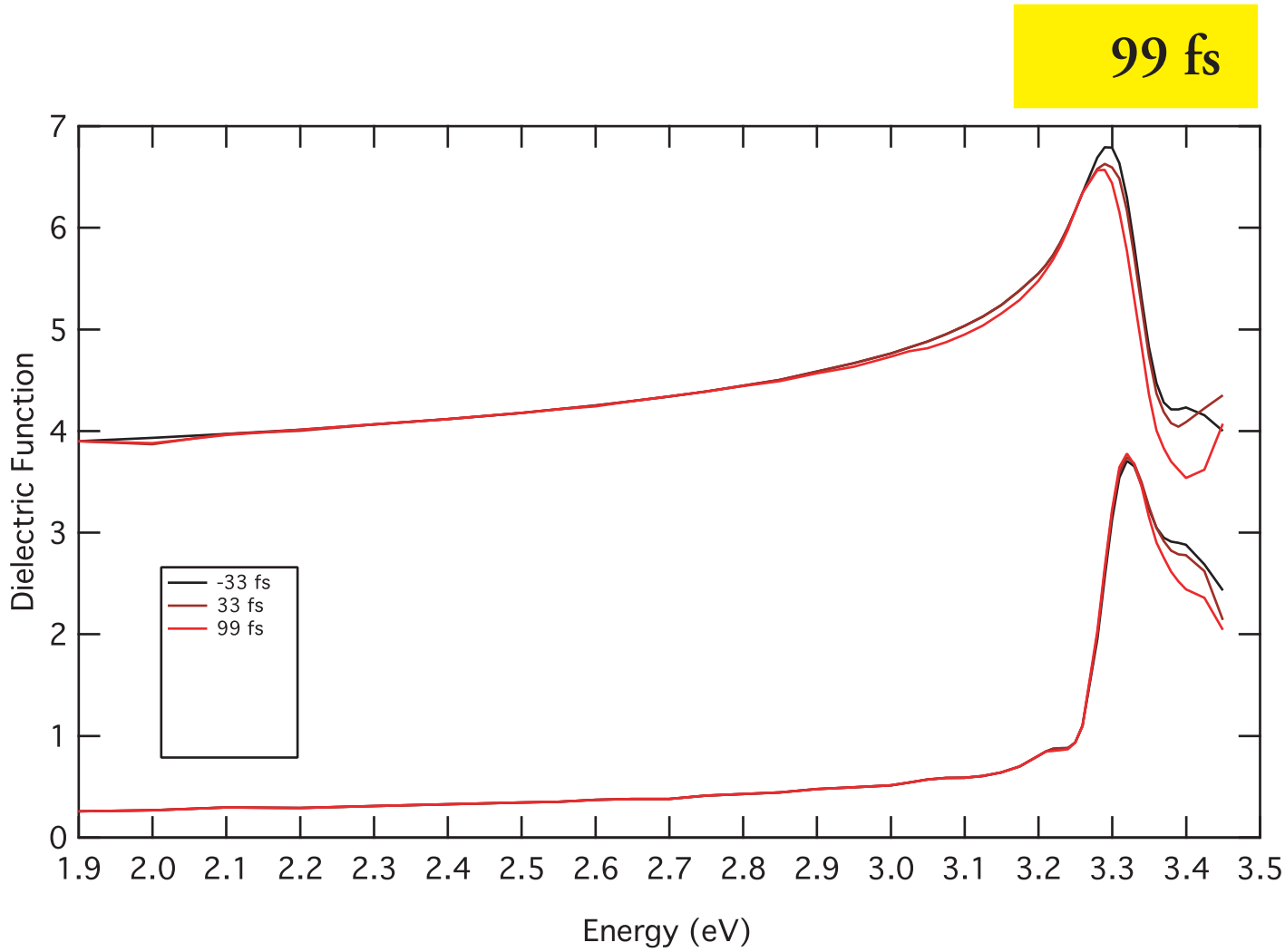


Ultrafast dielectric function dynamics

33 fs

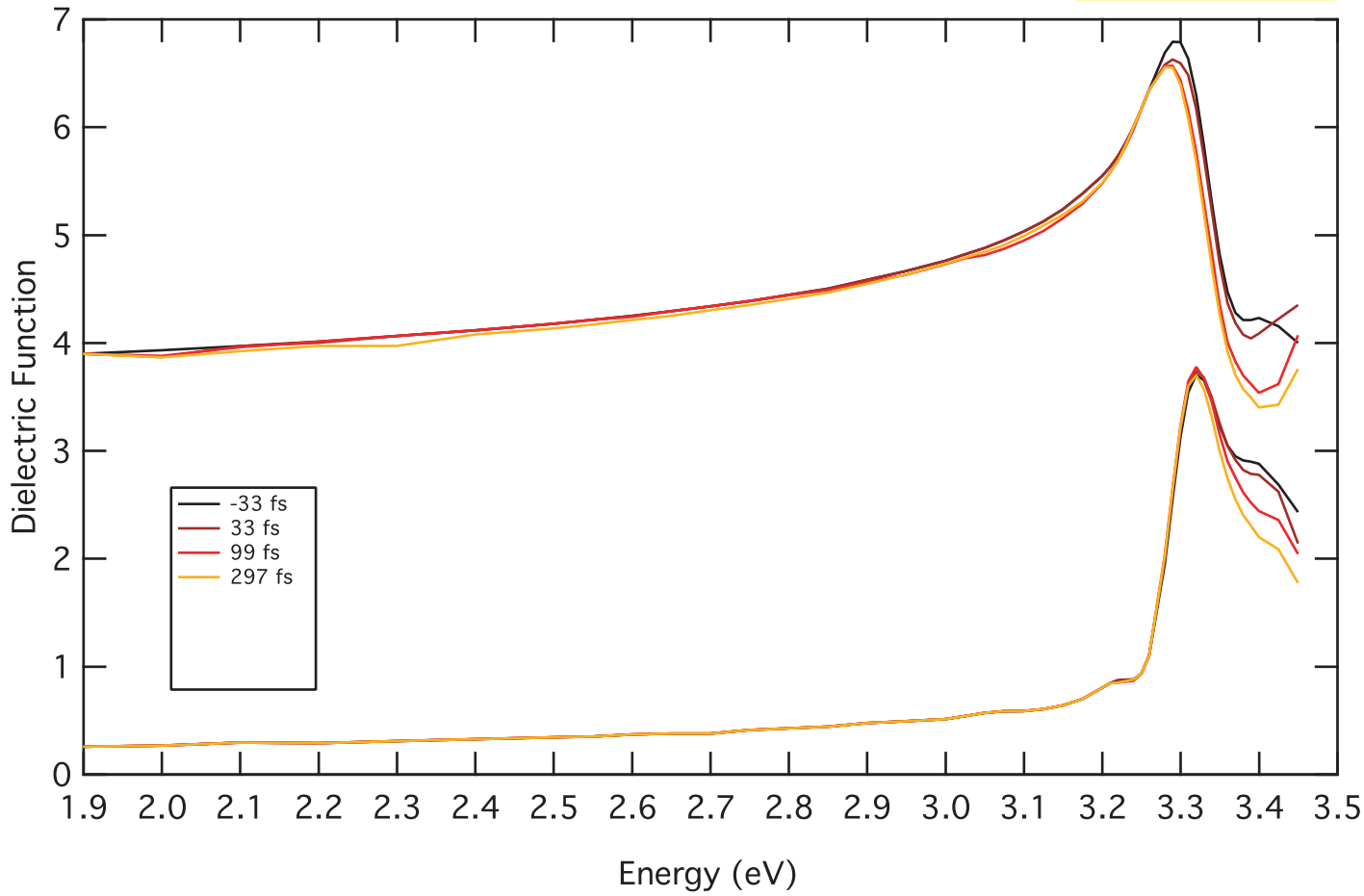


Ultrafast dielectric function dynamics

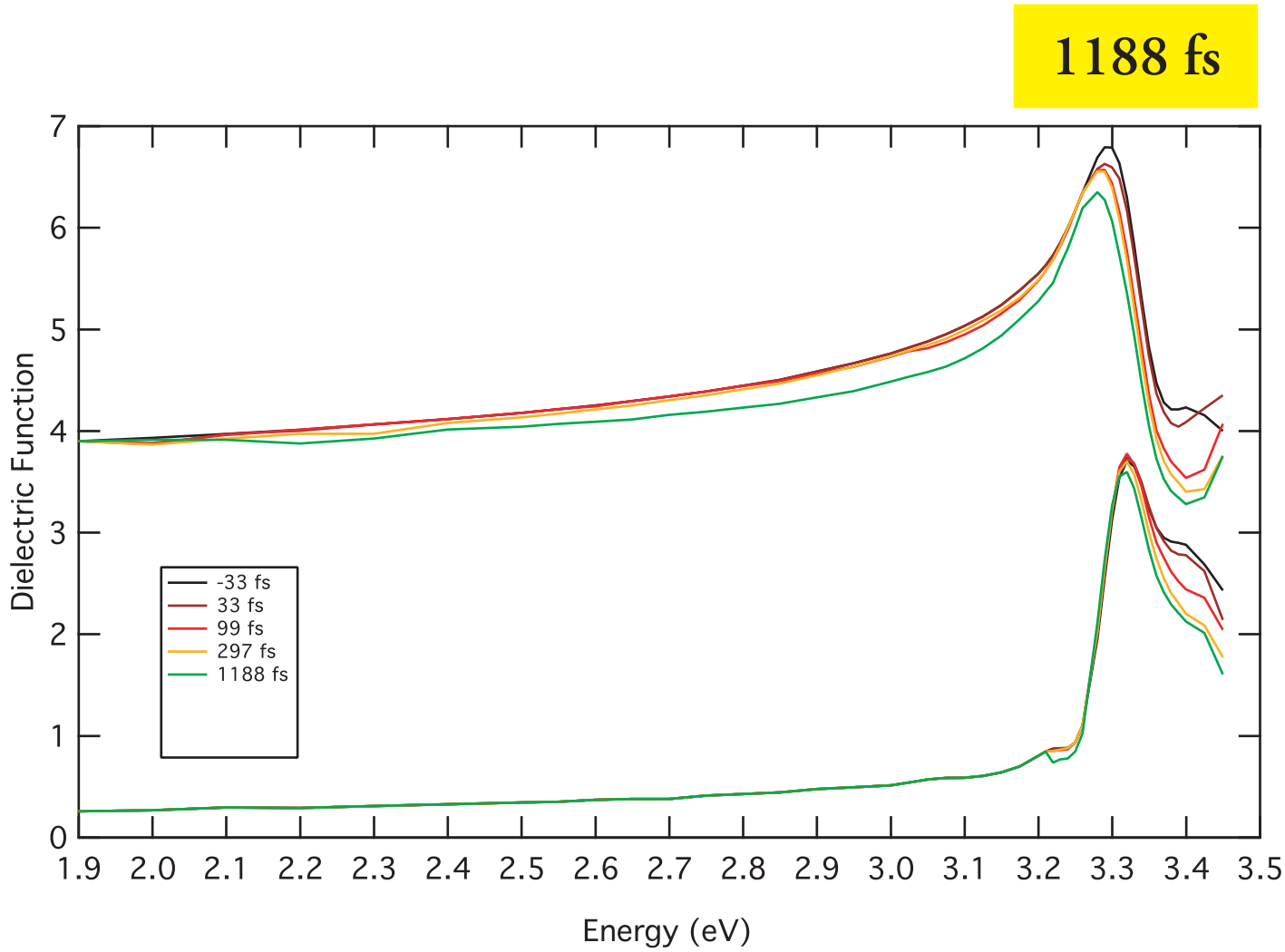


Ultrafast dielectric function dynamics

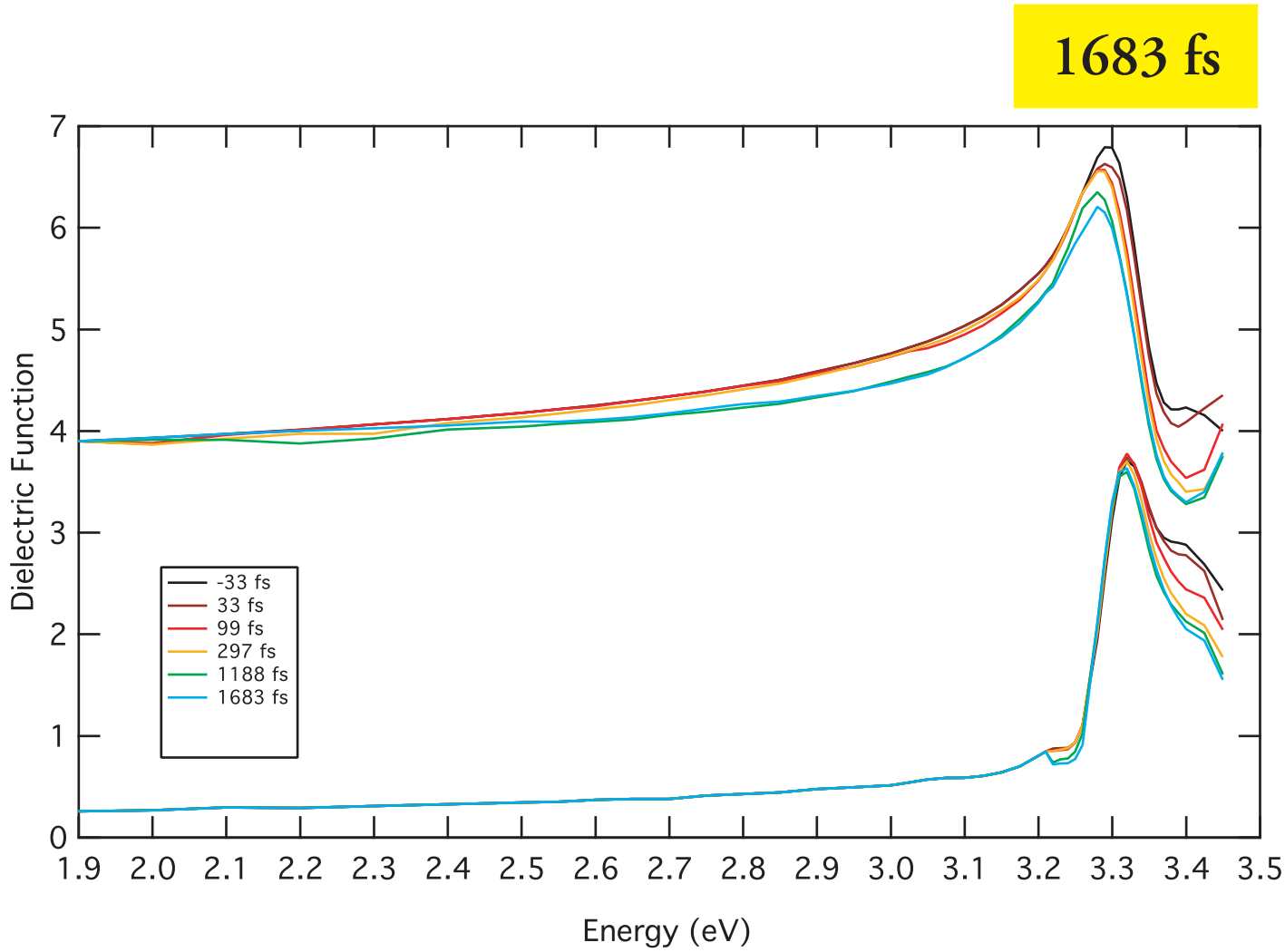
297 fs



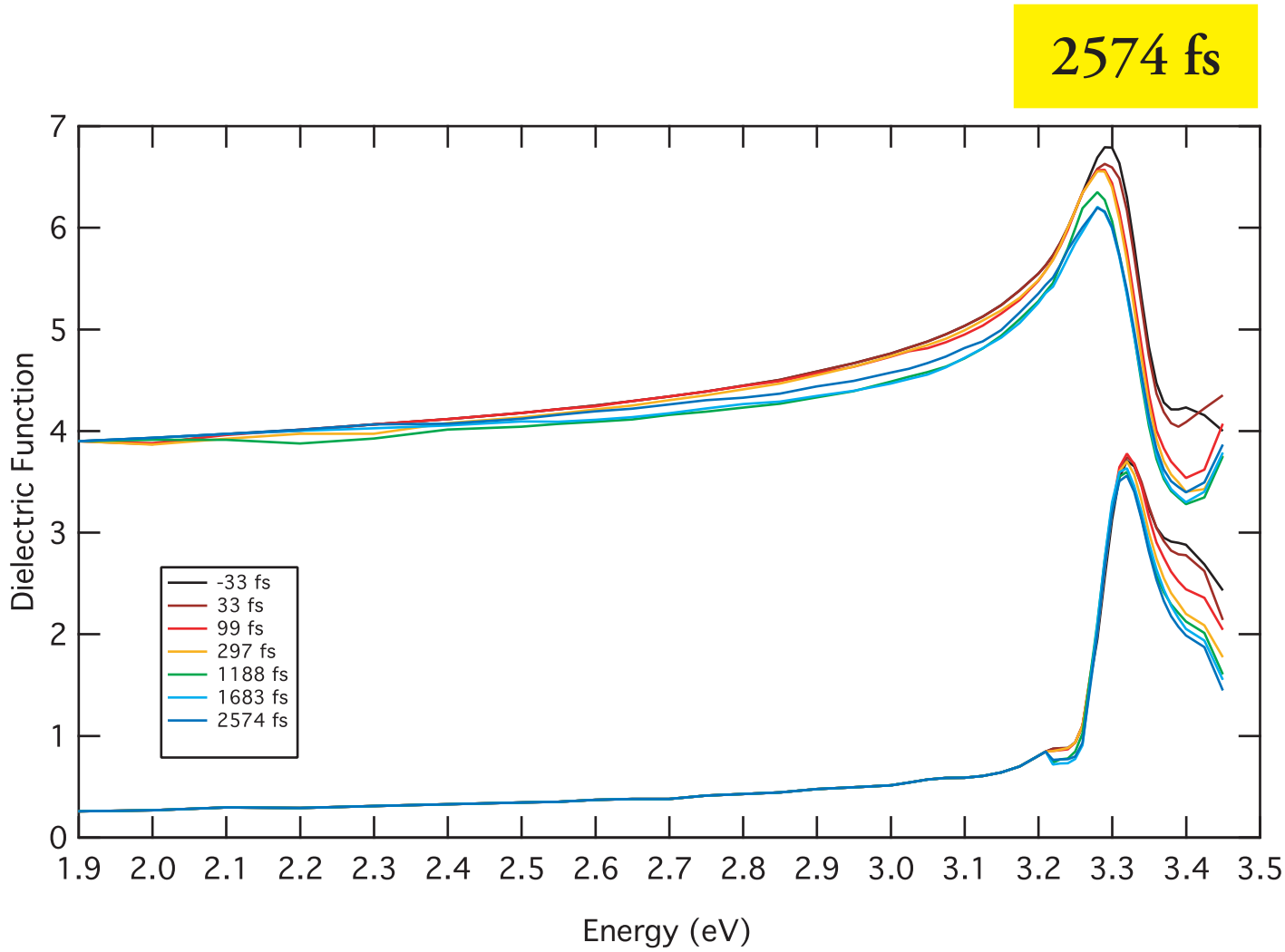
Ultrafast dielectric function dynamics



Ultrafast dielectric function dynamics



Ultrafast dielectric function dynamics



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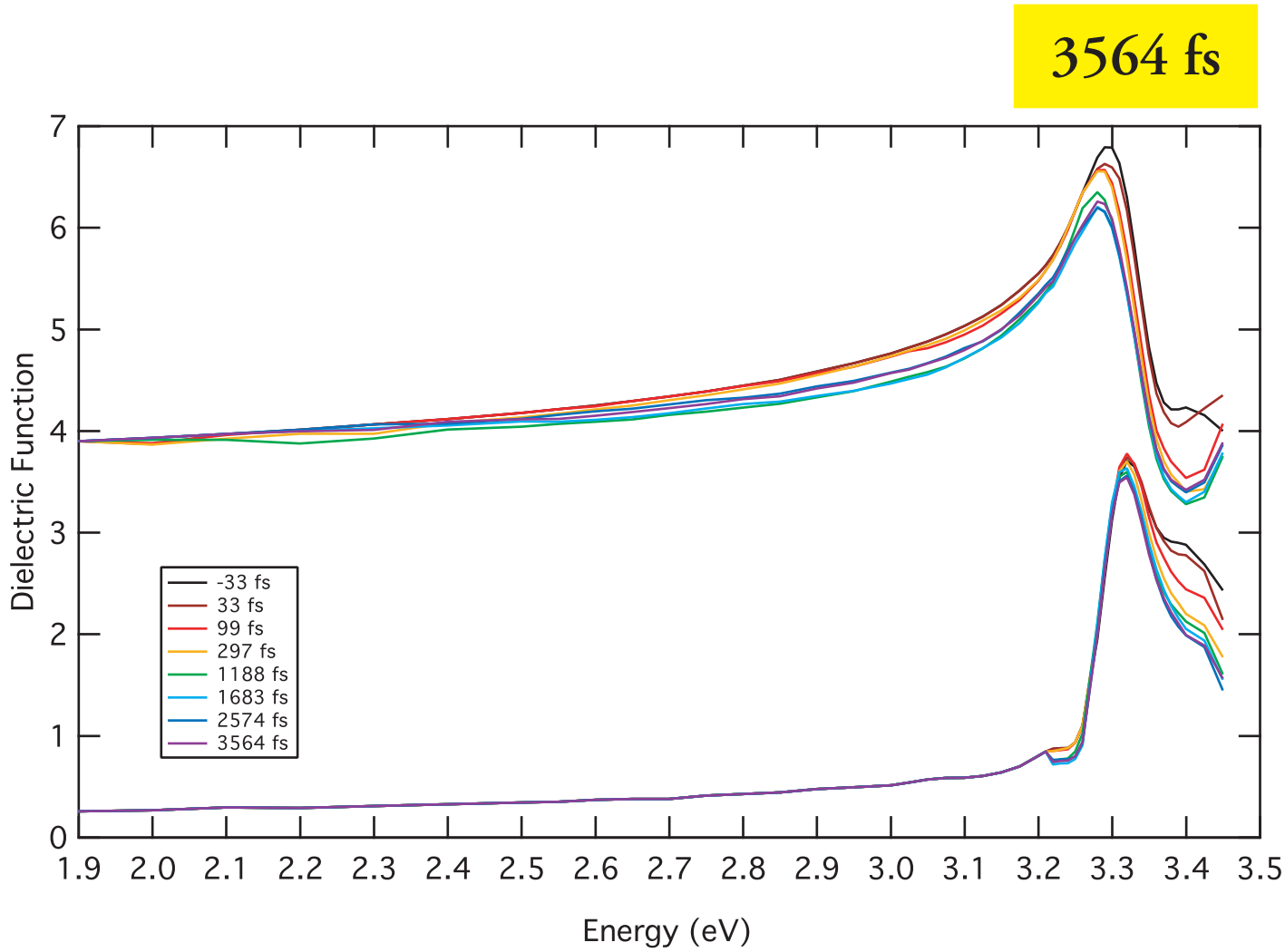

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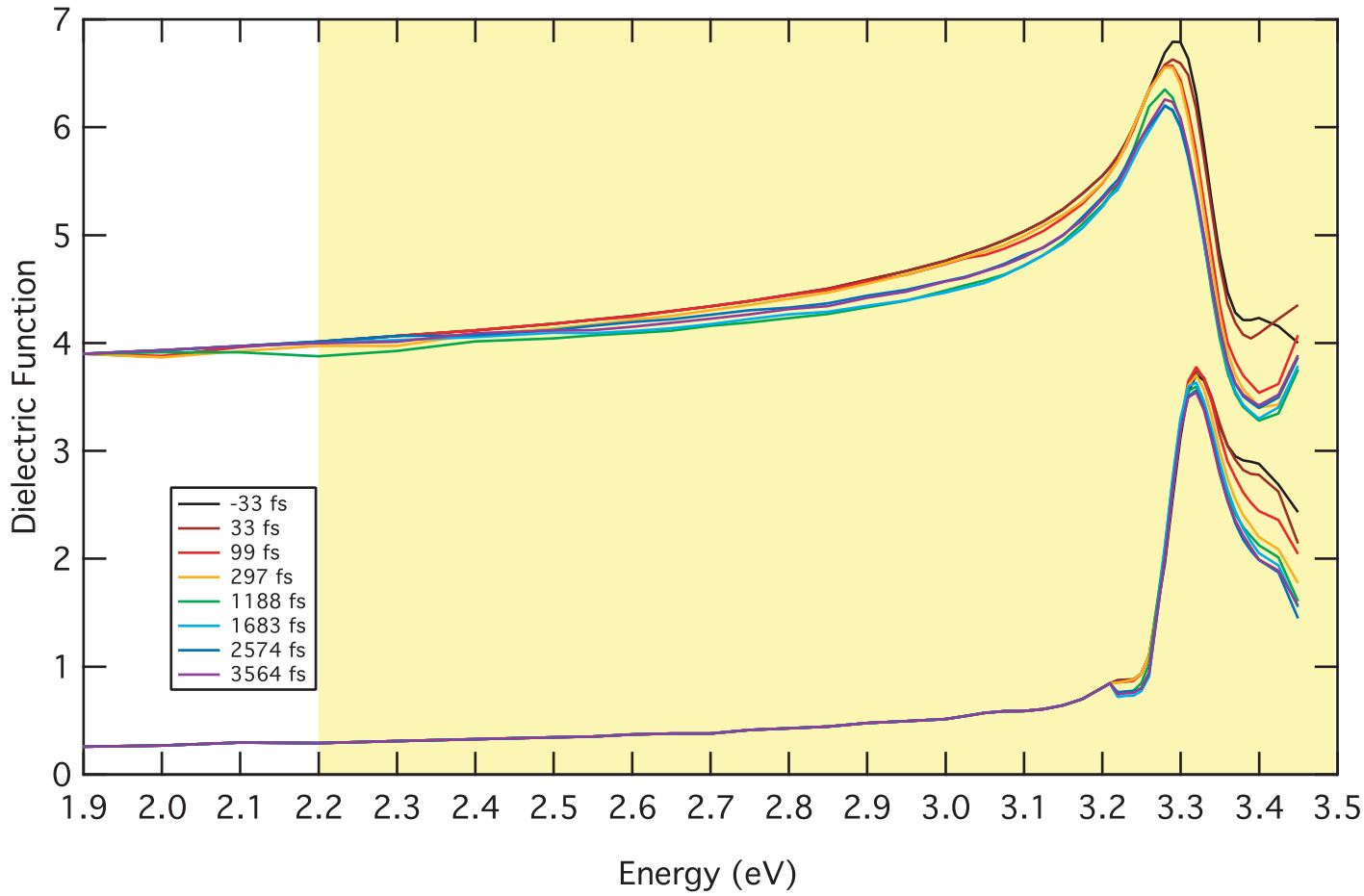

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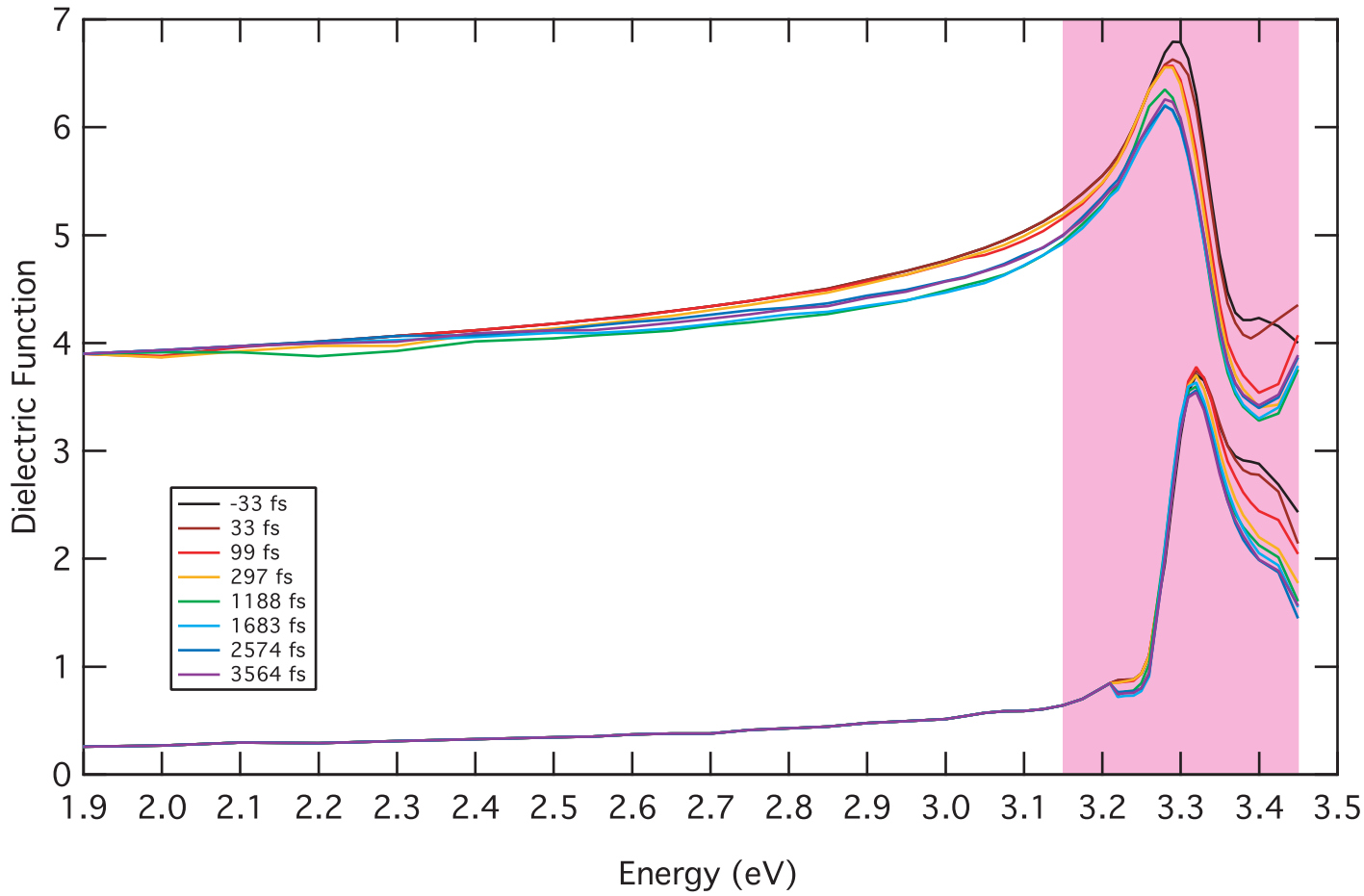
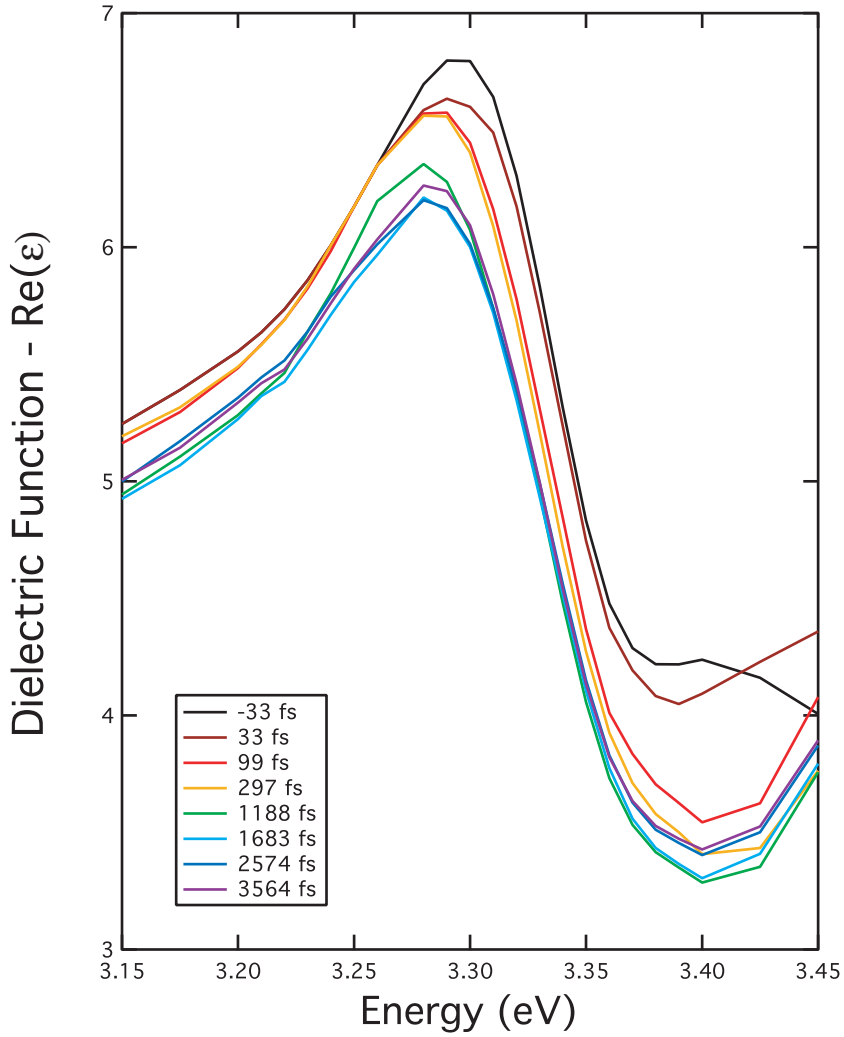


Ultrafast dielectric function dynamics



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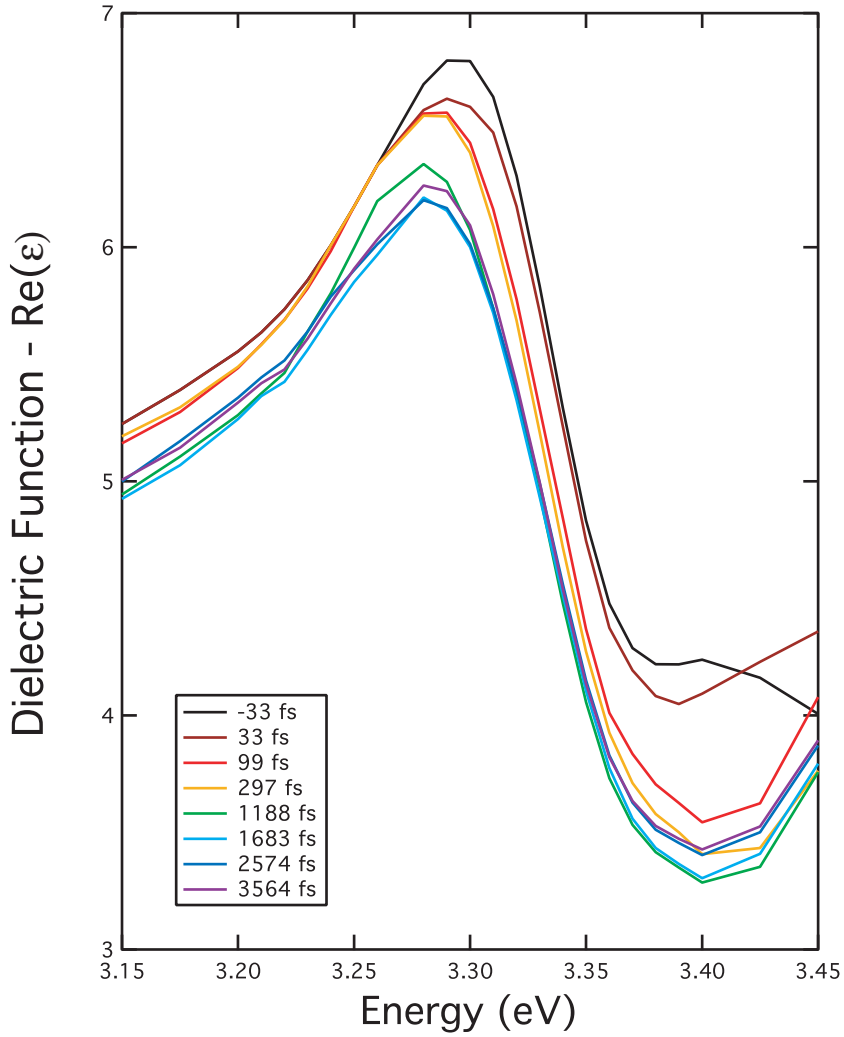
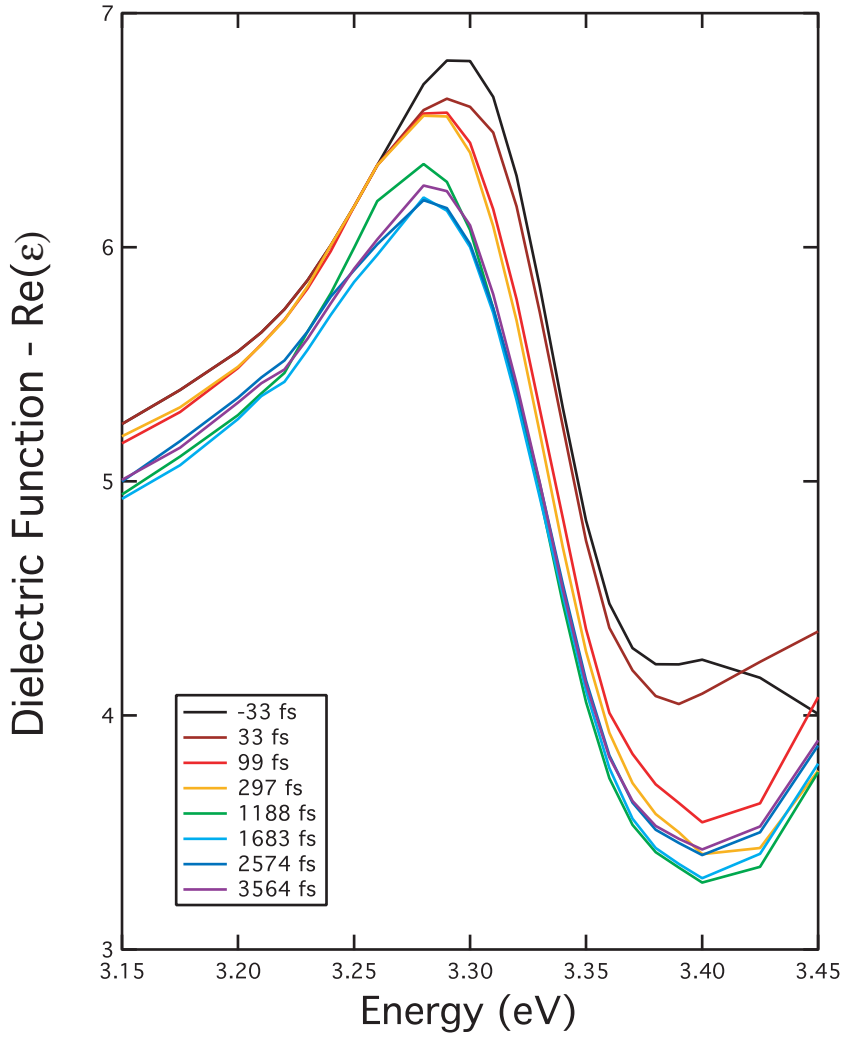


Red shift of peak near 3.3 eV

Bandgap renormalization

Ultrafast dielectric function dynamics



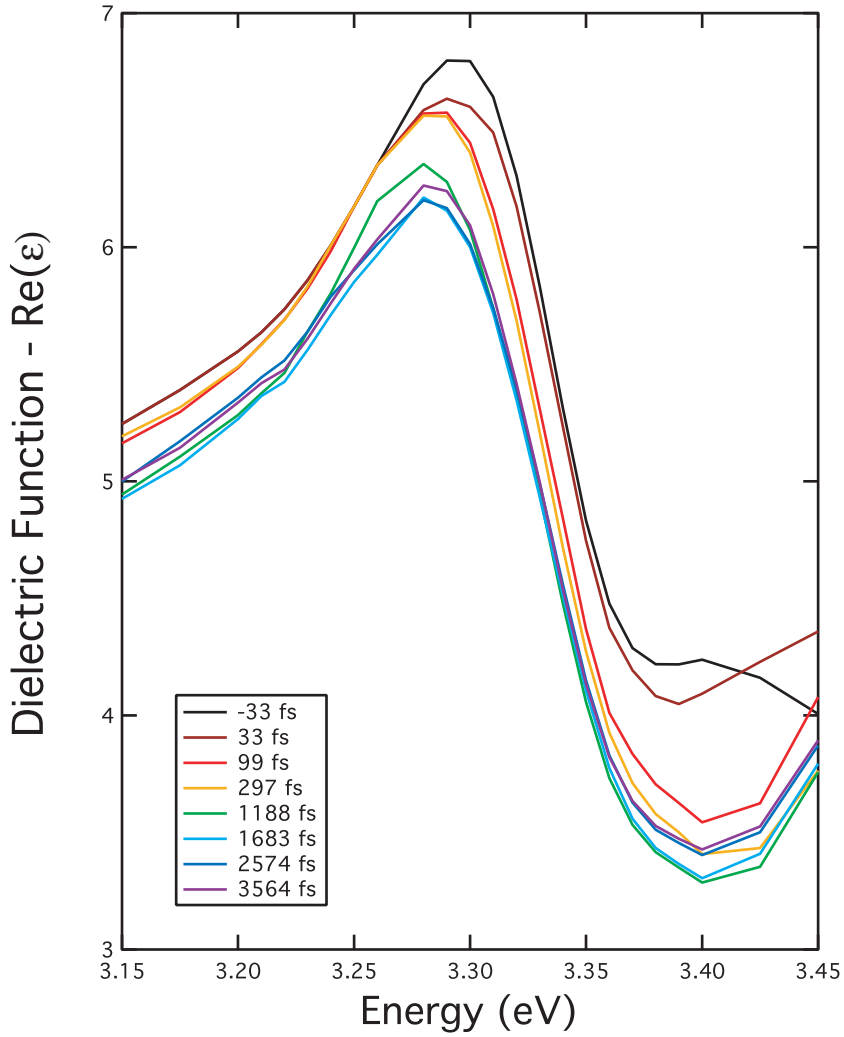
Red shift of peak near 3.3 eV

Bandgap renormalization

Blue shift of valley near 3.4 eV

Broadening of excitonic resonance

Ultrafast dielectric function dynamics



Red shift of peak near 3.3 eV

Bandgap renormalization

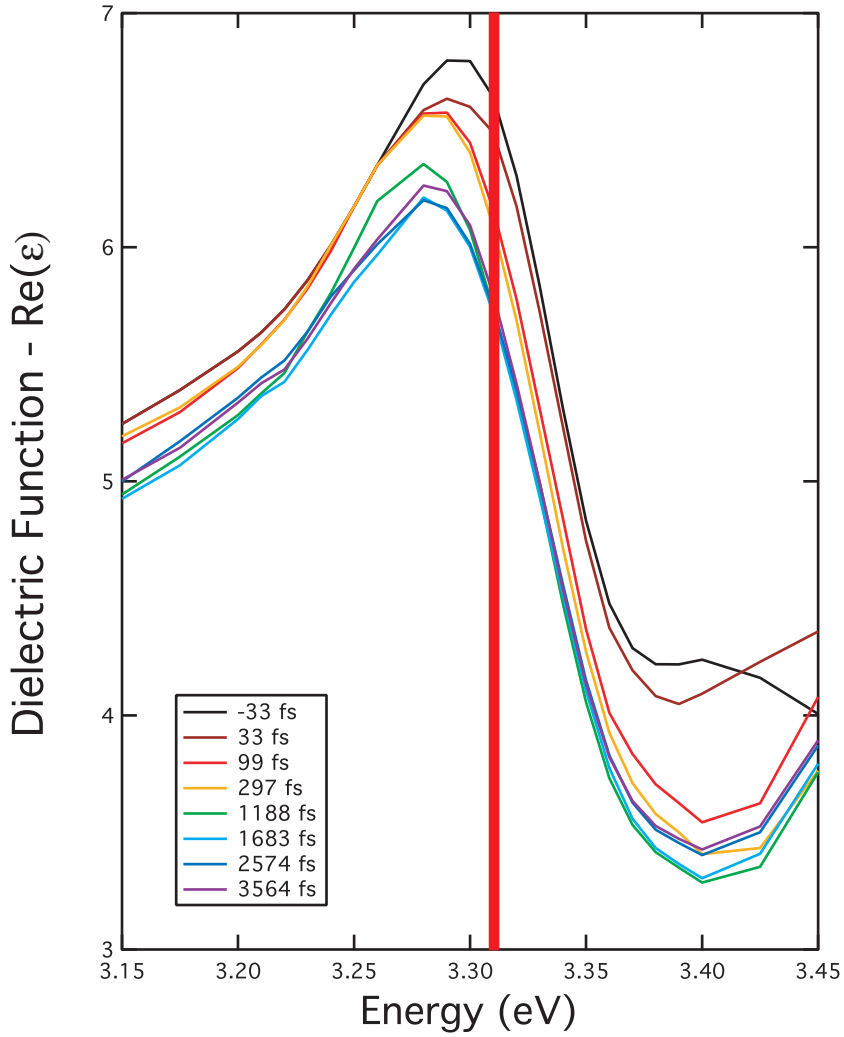
Blue shift of valley near 3.4 eV

Broadening of excitonic resonance

Maximum decrease of ~0.6

Damping of excitonic response

Ultrafast dielectric function dynamics



Red shift of peak near 3.3 eV

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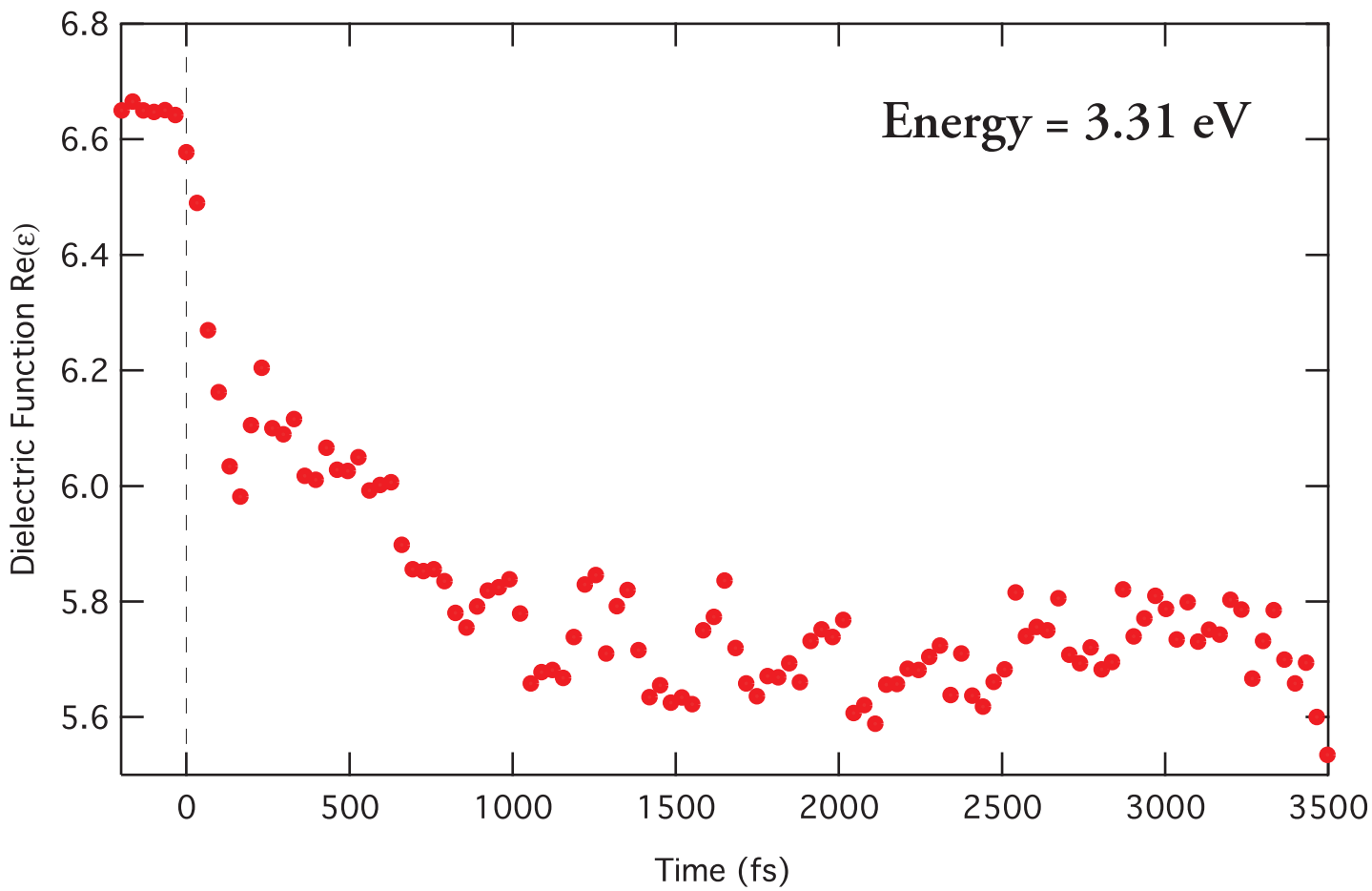
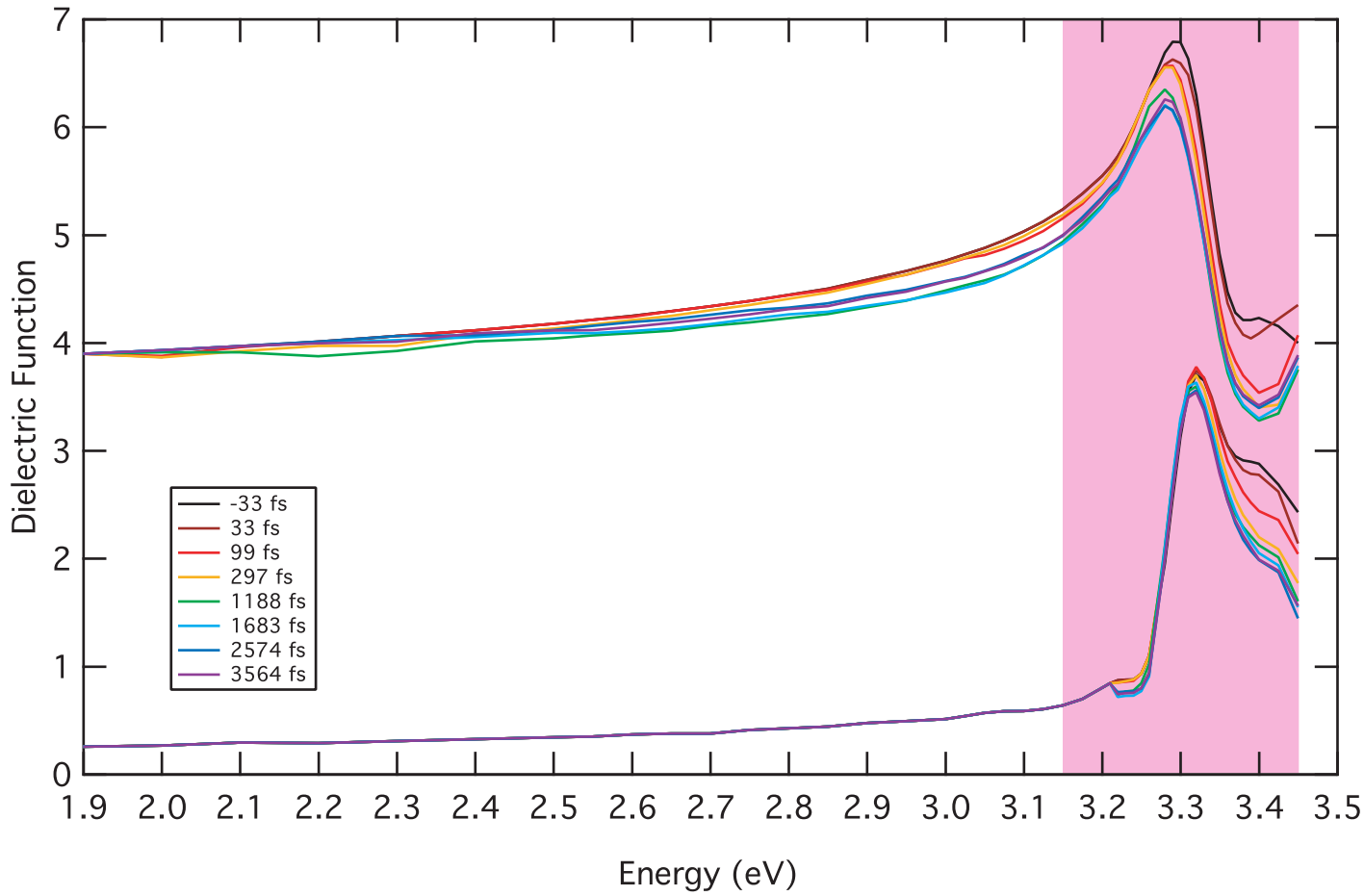


Ultrafast dielectric function dynamics



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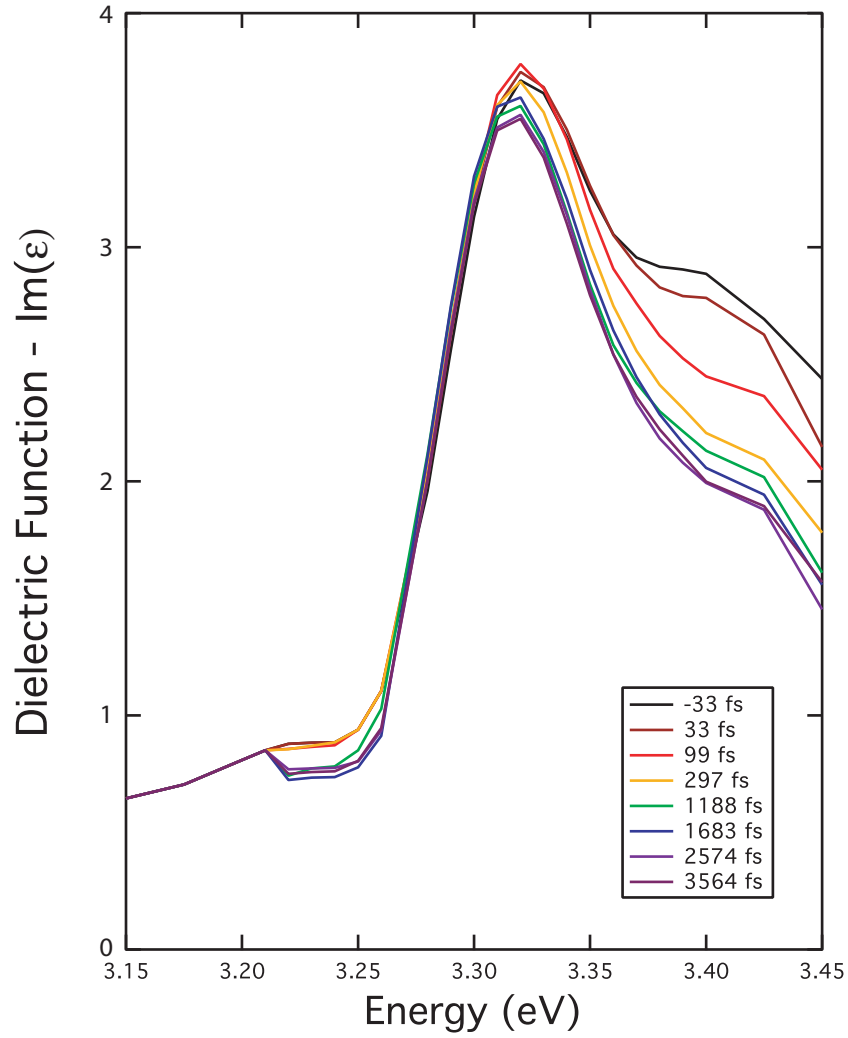


Ultrafast dielectric function dynamics

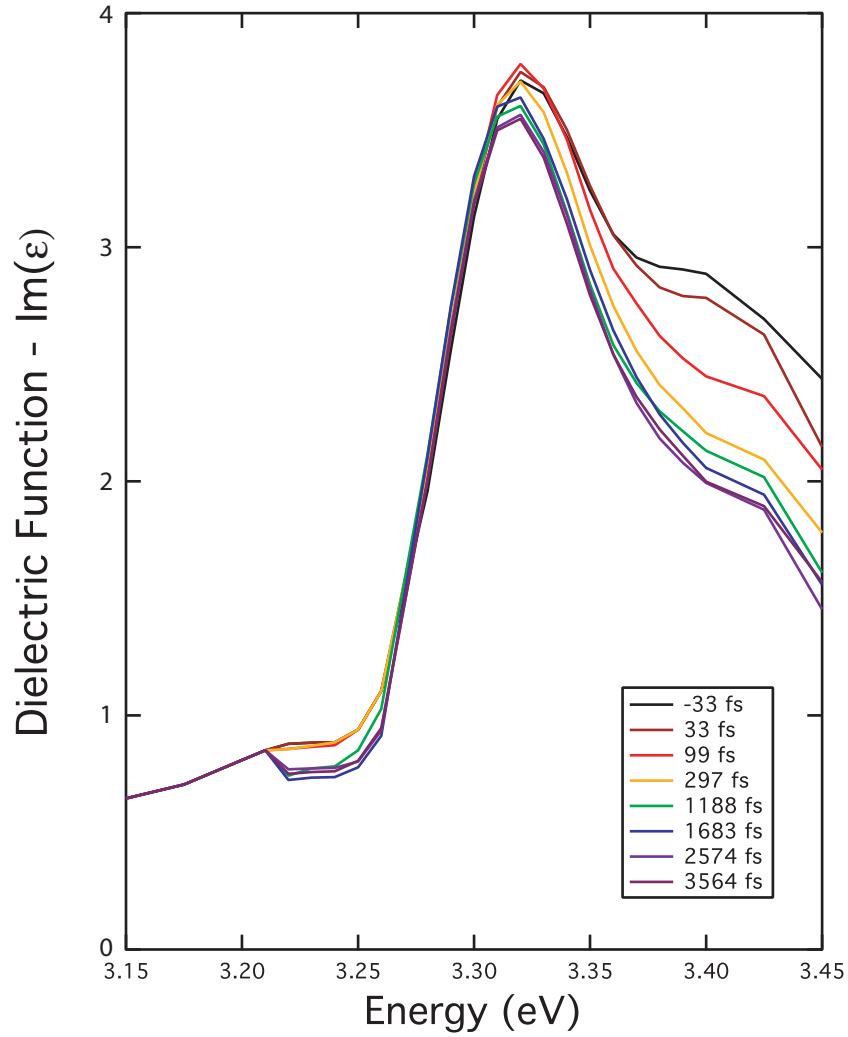
Above-bandgap excitation

Scattering of carriers

Filling of states

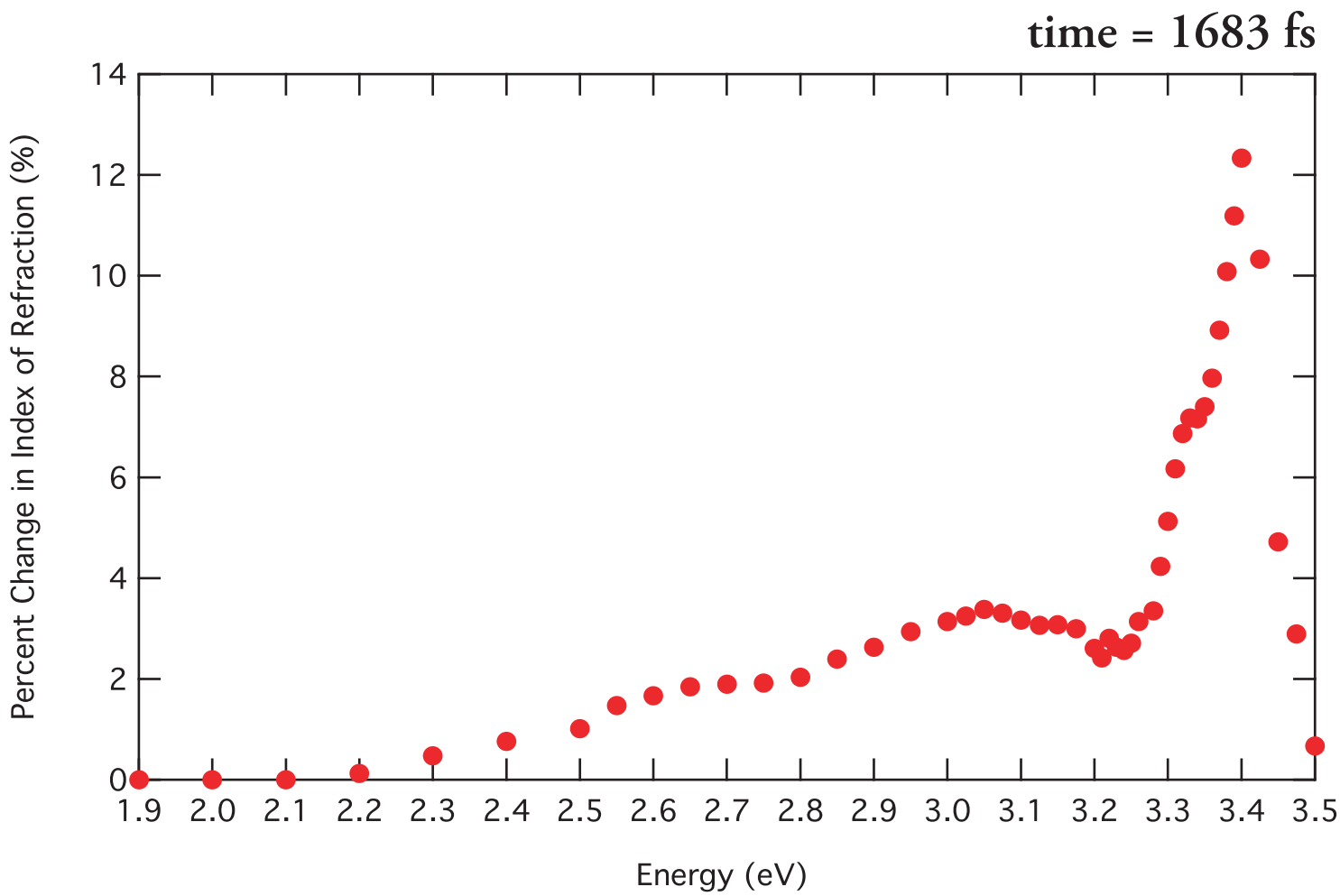
Bleaching of resonance

Decrease in absorption of ~0.7

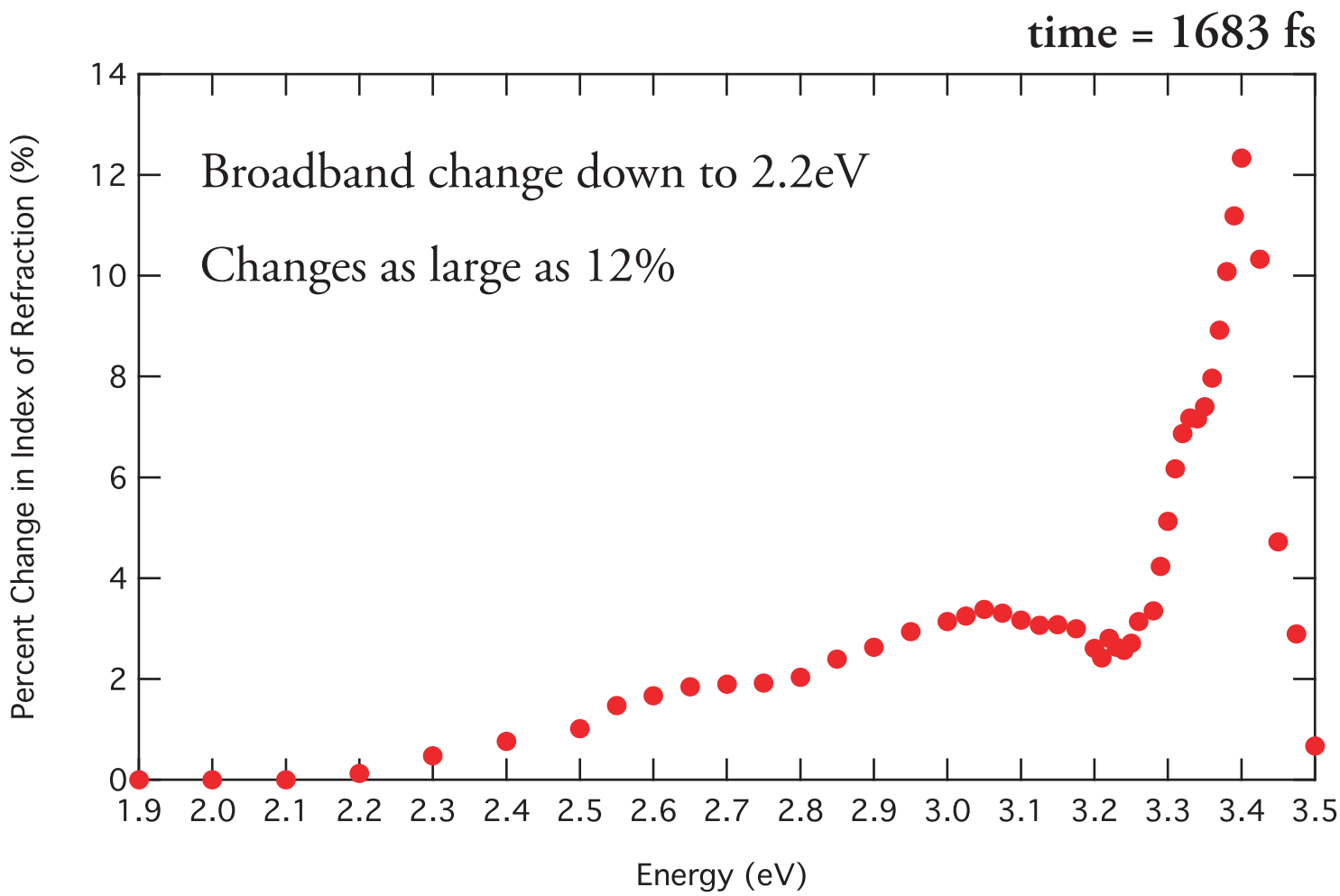


Link to real applications

Ultrafast index of refraction dynamics



Ultrafast index of refraction dynamics



Ultrafast index of refraction dynamics

Link to real applications

High excitation pumping

Excitation density of ~10^{16} carriers/cm^3

Index changes persist over ~80 ps

Ultrafast index of refraction dynamics

Link to real applications

High excitation pumping

Excitation density of ~10^{16} carriers/cm^3

Index changes persist over ~80 ps

Determines optical length of resonators

Determines lasing modes

Ultrafast index of refraction dynamics

Link to real applications

High excitation pumping

Excitation density of ~10^{16} carriers/cm^3

Index changes persist over ~80 ps

Determines optical length of resonators

Determines lasing modes

Optimization of ZnO laser

Outline

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Pump-probe reflectometry

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Time-Resolved ZnO Dielectric Function

Dynamics in 1-3 ps

~10% change in Re(ε) and Im(ε)

Broadening of resonance

Bandgap renormalization

Filling of states

Summary

Time-Resolved ZnO Dielectric Function

Dynamics in 1-3 ps

~10% change in Re(ε) and Im(ε)

Broadening of resonance

Bandgap renormalization

Filling of states

Applications

Directly measure index of refraction

Maximum of 12% change

Large carrier excitation density

Optimize design of ZnO lasers

Thanks to the members of the Mazur group

This research is supported by the National Science Foundation