Ultrafast exciton dynamics in bulk ZnO

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Outline

Motivation

ZnO excitons

Pump-probe reflectivity

Reflectivity data

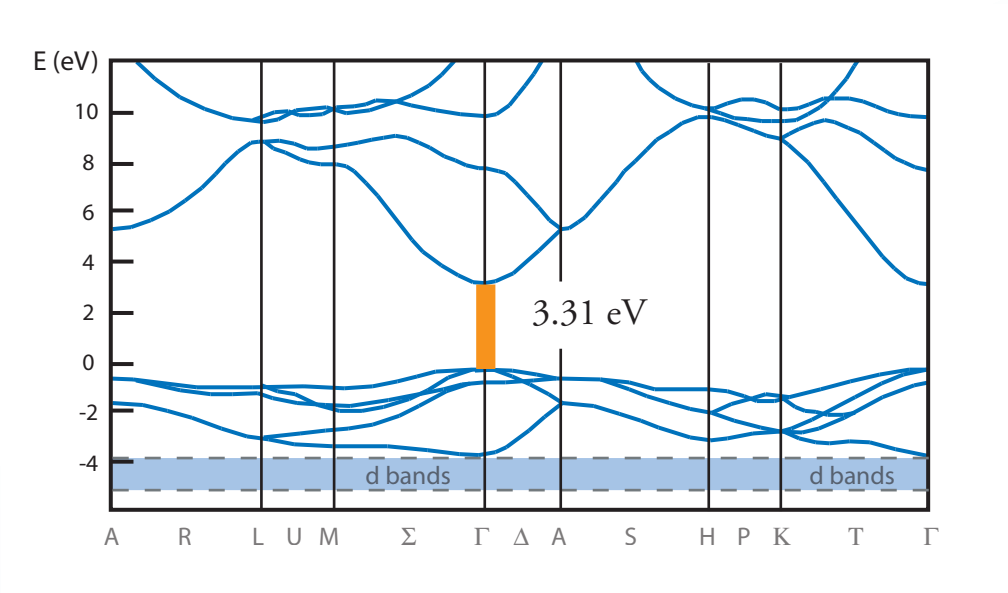
Time-resolved data

Conclusion

Motivation

Zinc Oxide (ZnO)

- Large direct bandgap (3.31 eV)
- Large exciton binding energy (60 meV)



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

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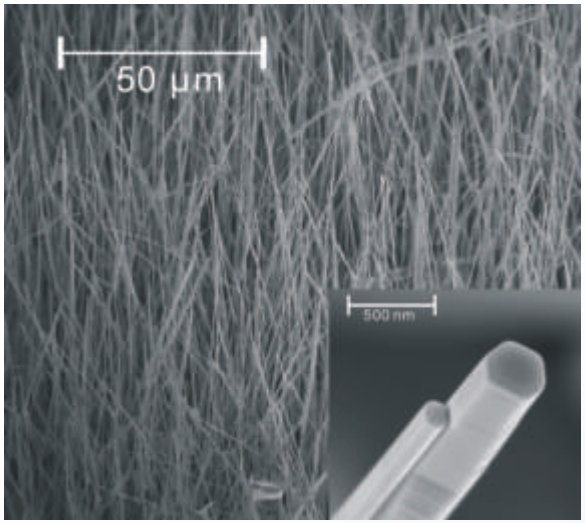

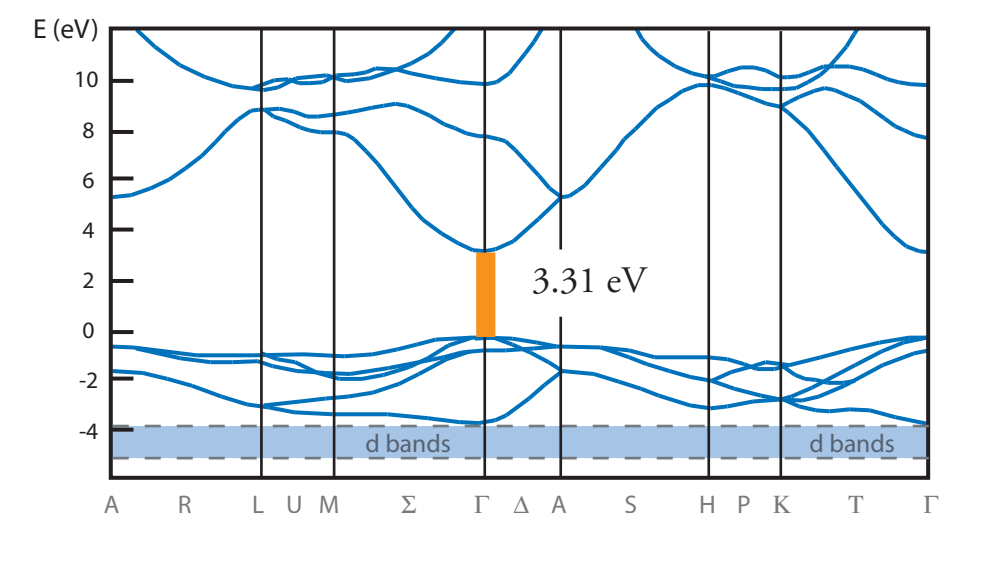

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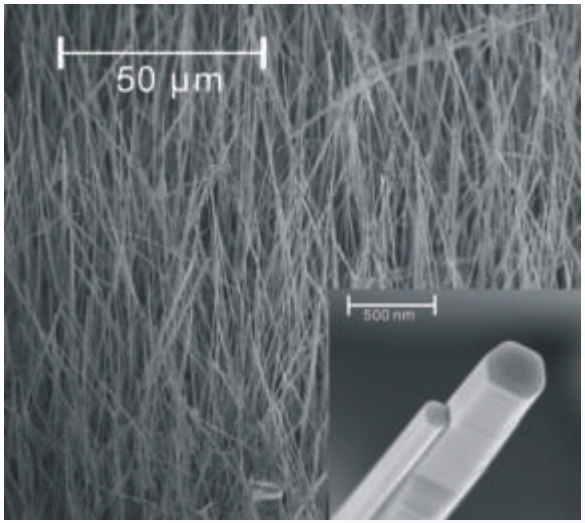

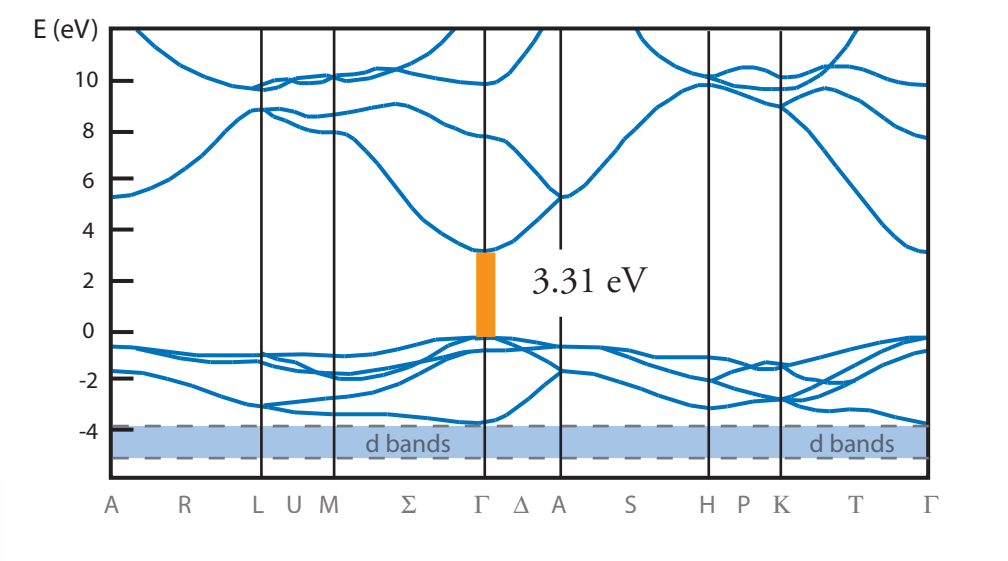

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ZnO-based Technologies

- Bulk and nanowire lasers
- Light Emitting Diodes

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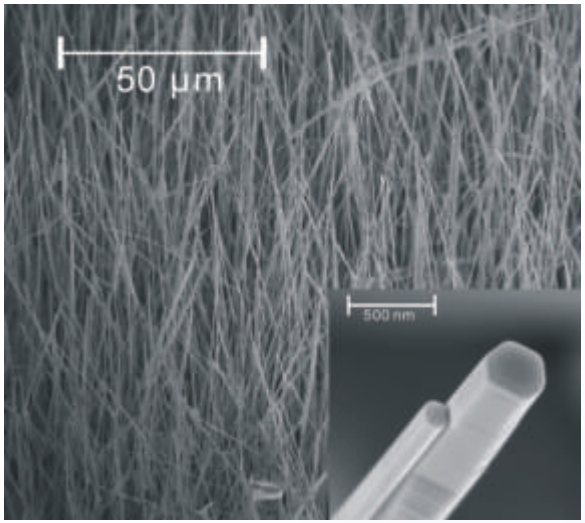

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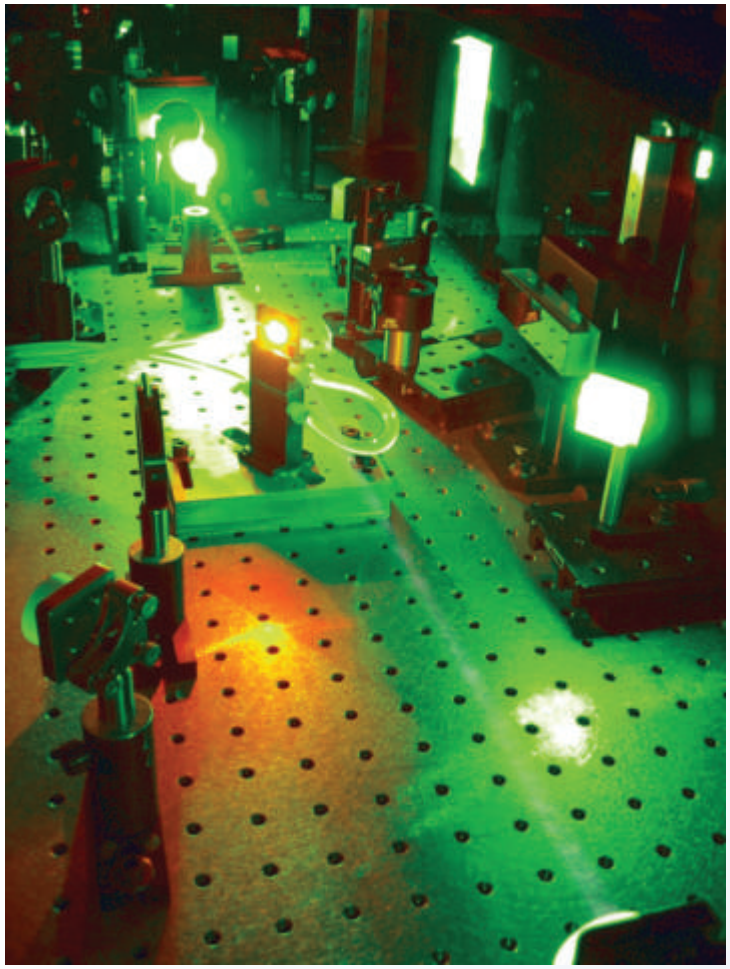
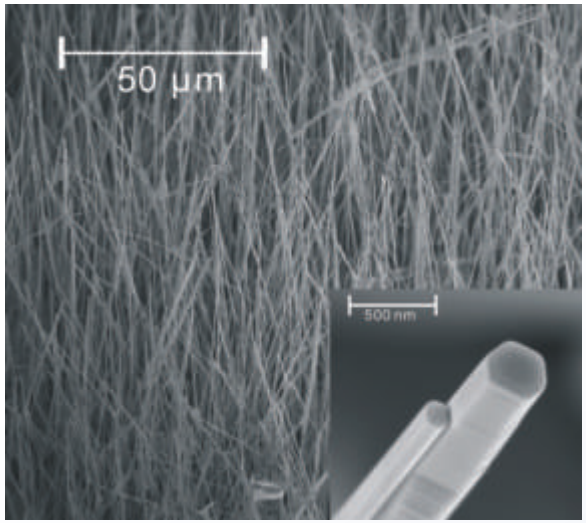
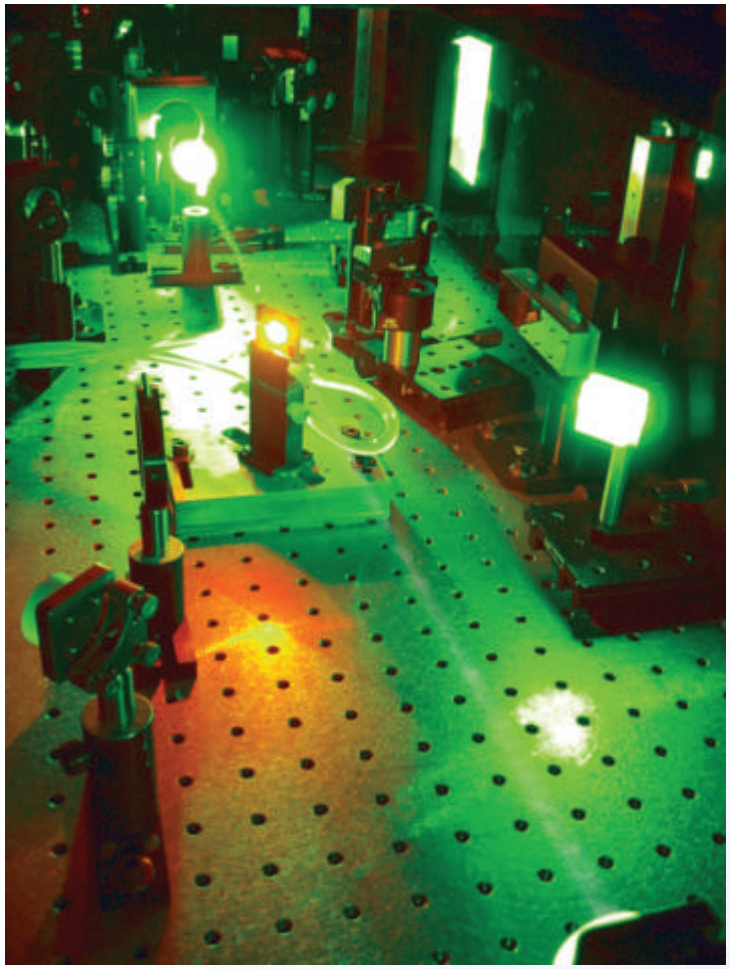
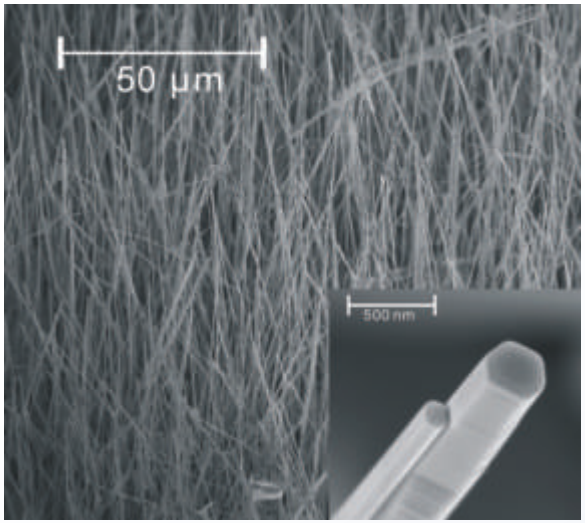




Motivation

Femtosecond lasers





increasing availability

less heating effects

low lasing threshold compared to
ns and ps pump pulse durations

Y.F. Hsu, *Opt. Mat.* **31**, 35-38 (2008)

What is the primary mechanism for lasing in ZnO?

Exciton-exciton scattering

Electron-hole plasma recombination

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Lasing mechanism?

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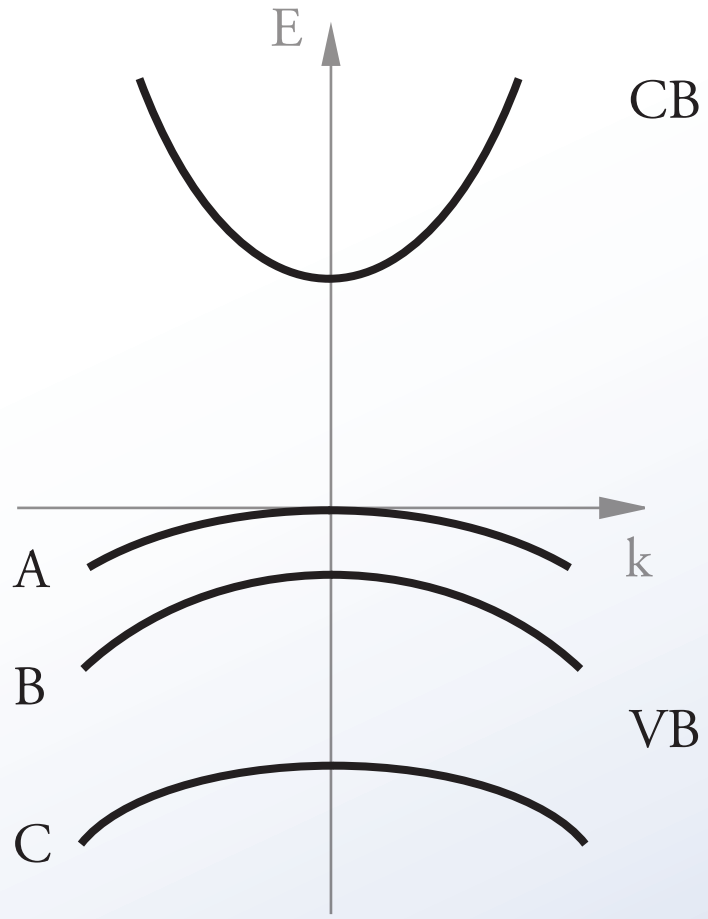
Excitons present at room temperature



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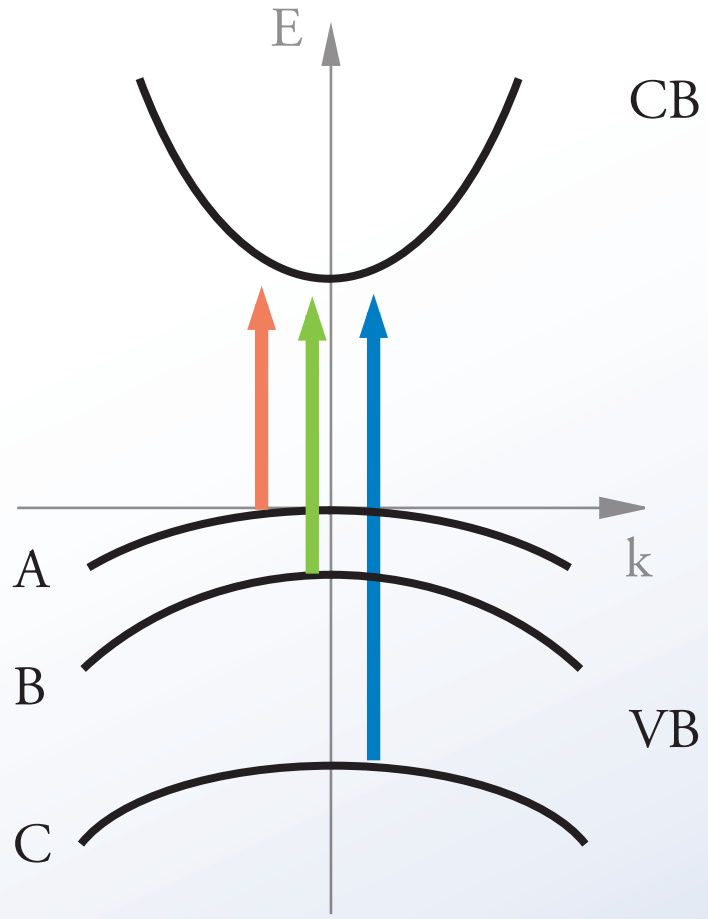

Lasing mechanism?

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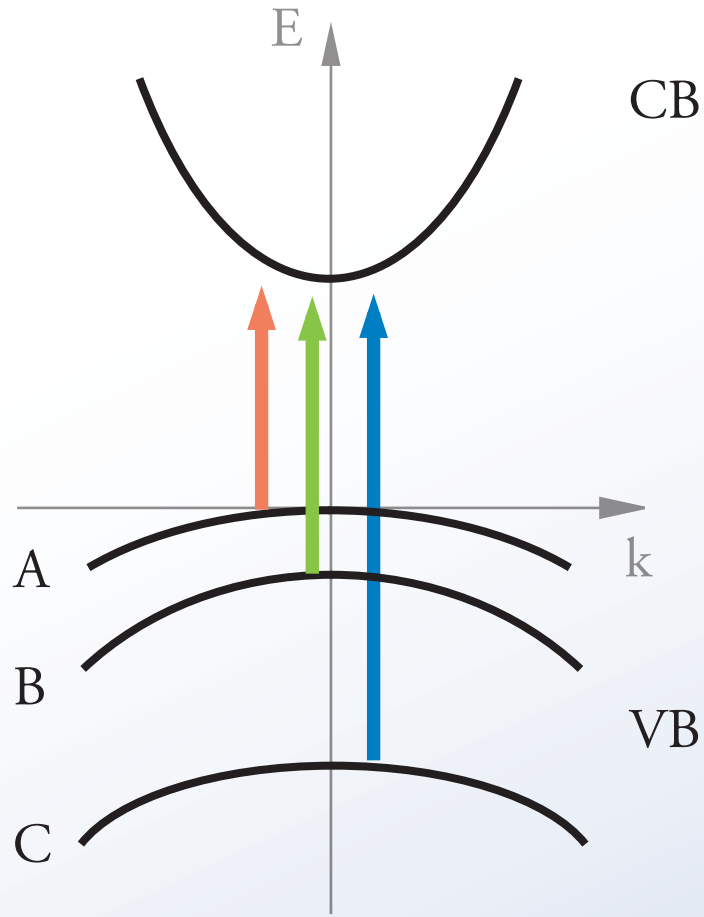
Two excitons scatter inelastically

- Up into higher energy state
- Down the photon-like part of the dispersion relation
- Emitting a photon



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Pump excitation creates free carriers

Exceed Mott density of $5*10^{17}$ /cm^3

from C. Klingshirn, *Chem. Phys. Chem.* **8**, 782 (2007)

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Significant Coulomb screening

Excitons no longer good quasiparticles

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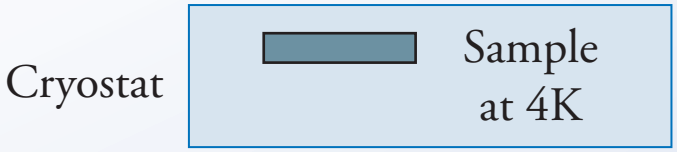
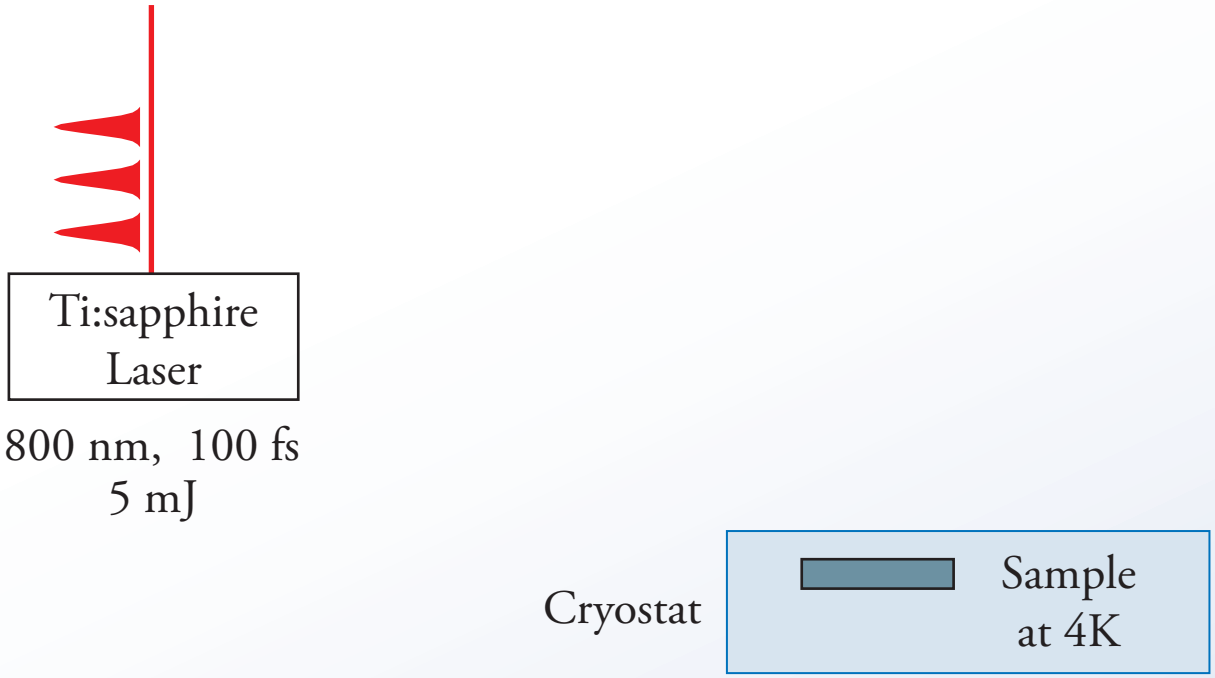
Exciton-exciton scattering vs. Electron-hole plasma recombination

Pump-probe spectroscopy



Micro-cryo broadband pump-probe reflectivity setup

Pump-probe spectroscopy



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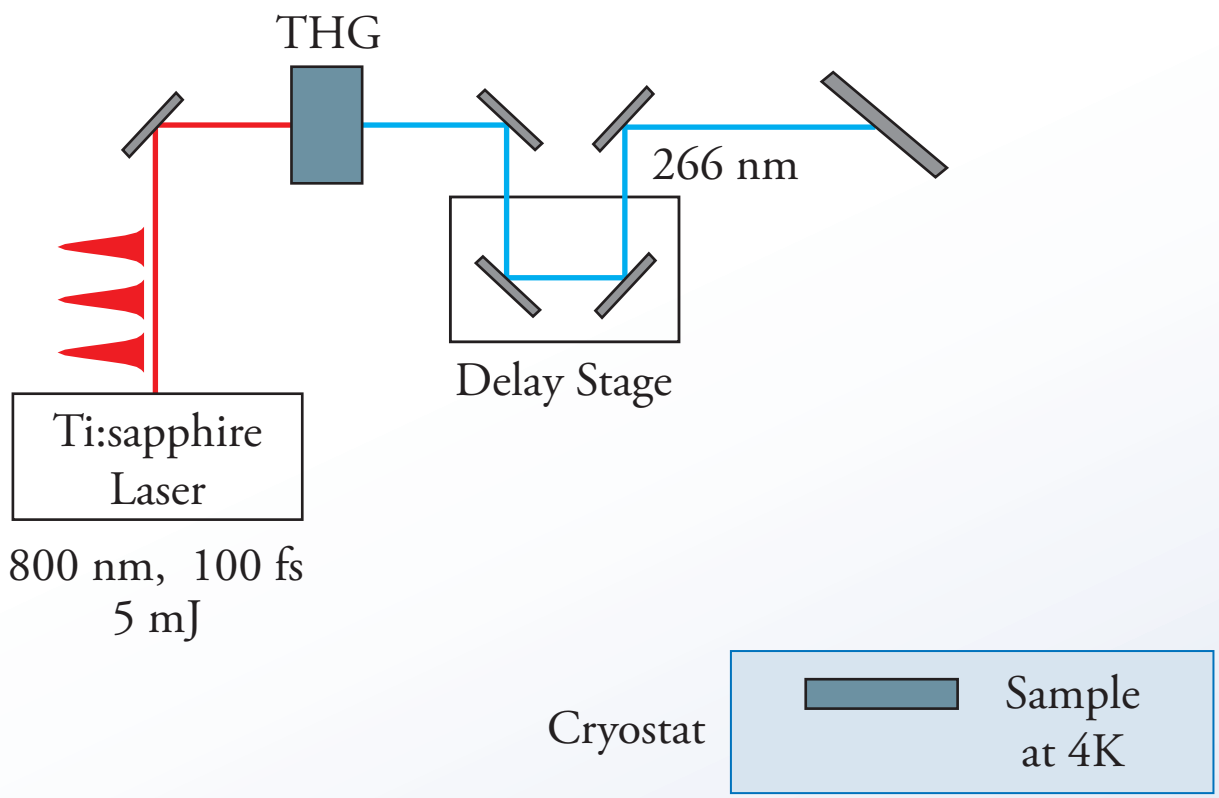

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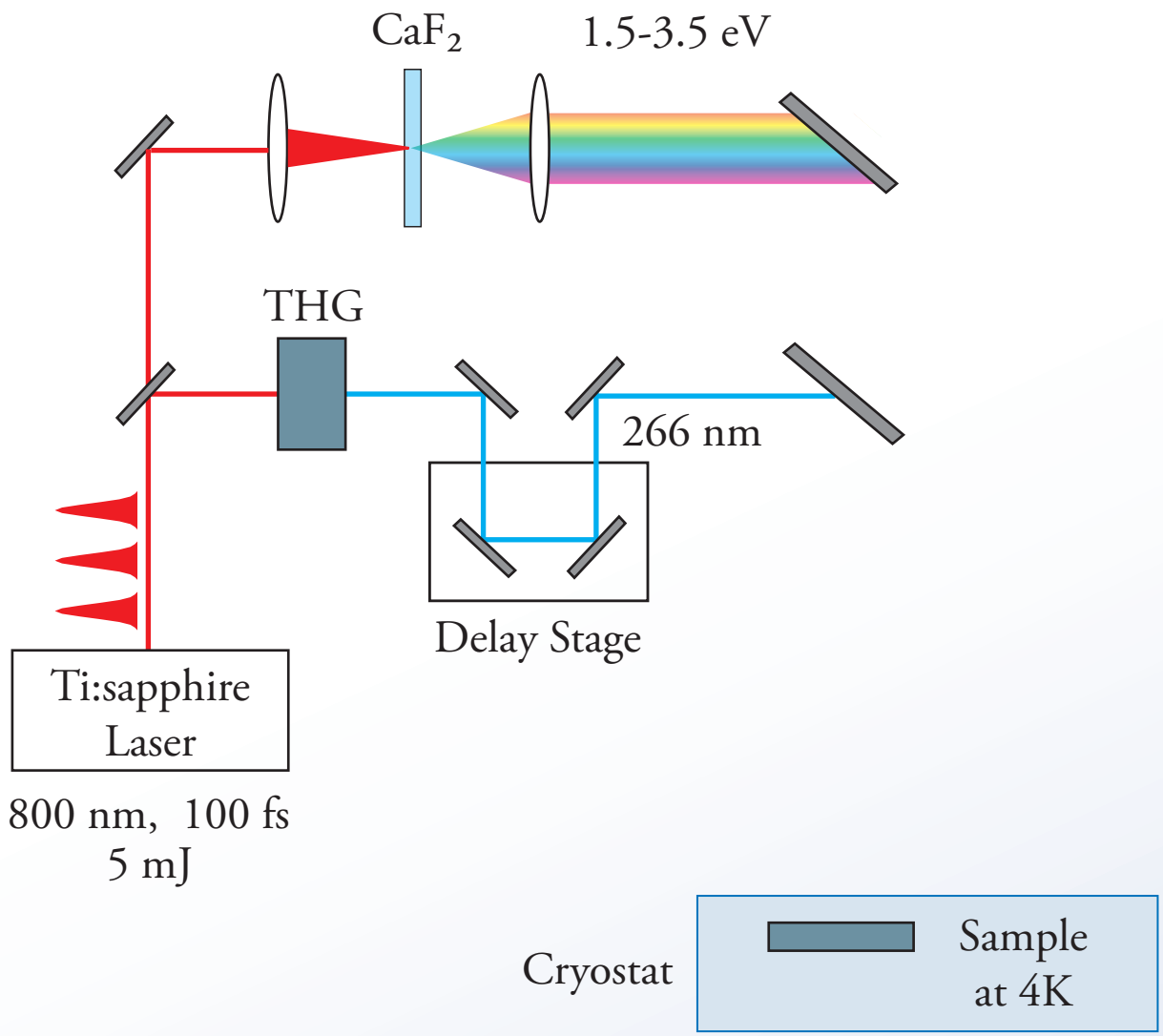

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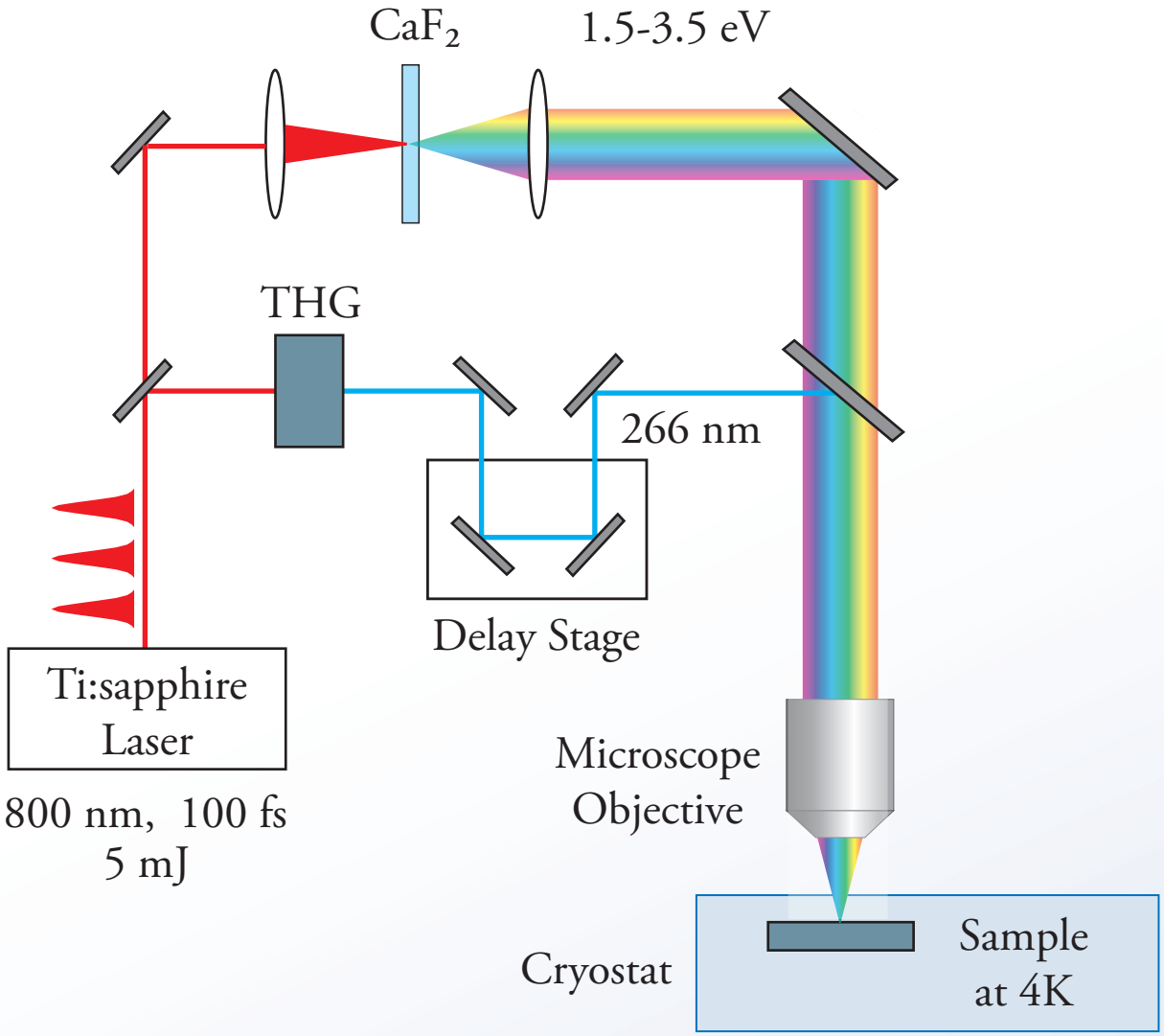

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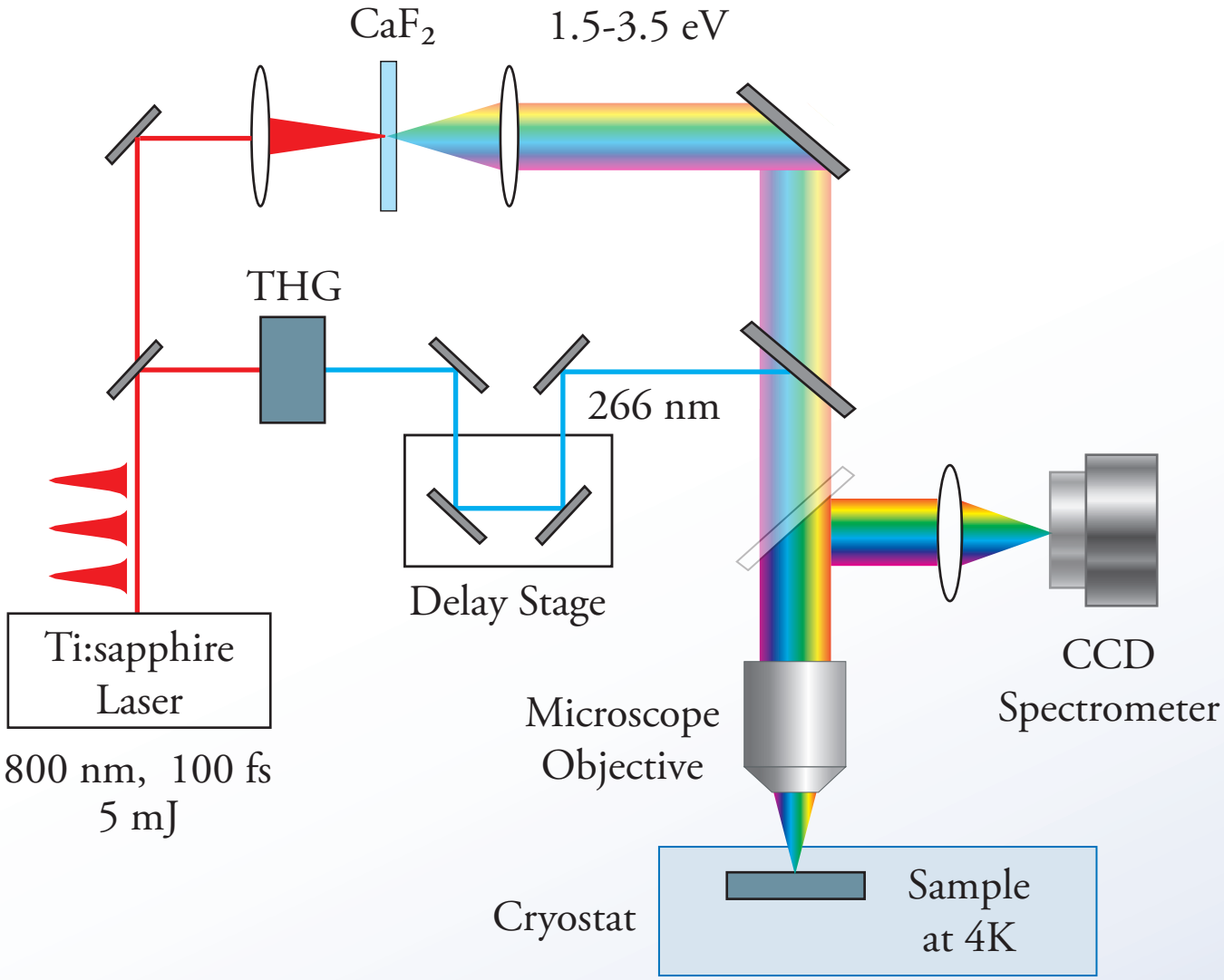
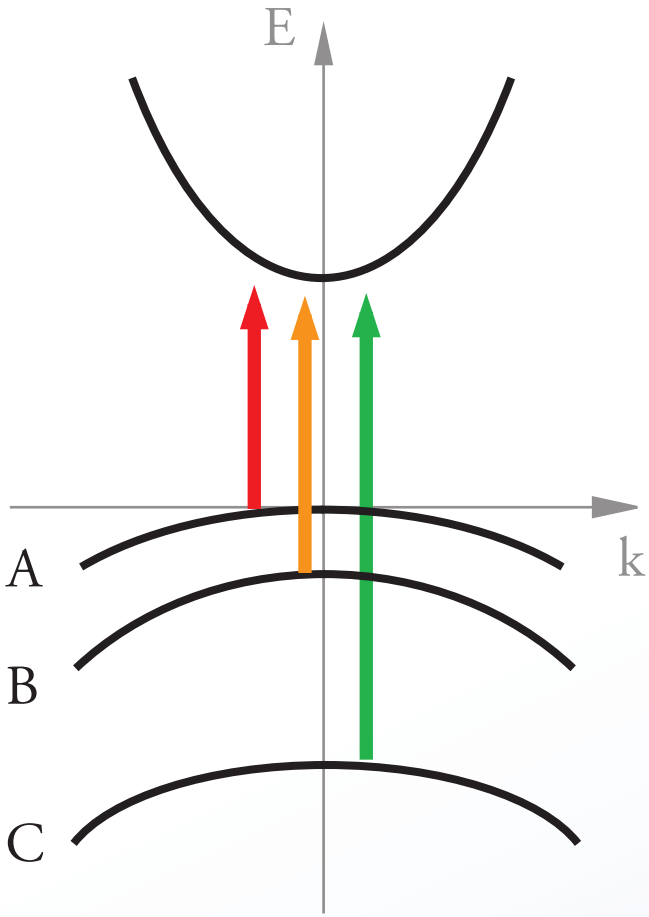


Micro-cryo broadband pump-probe reflectivity setup

Can we “see” the excitons?

Reflectivity data



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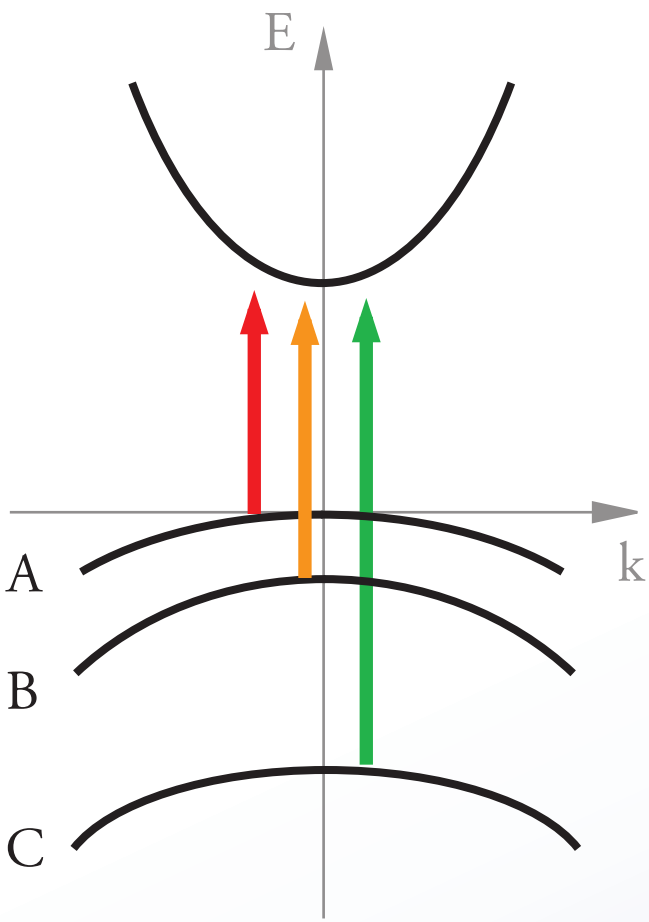

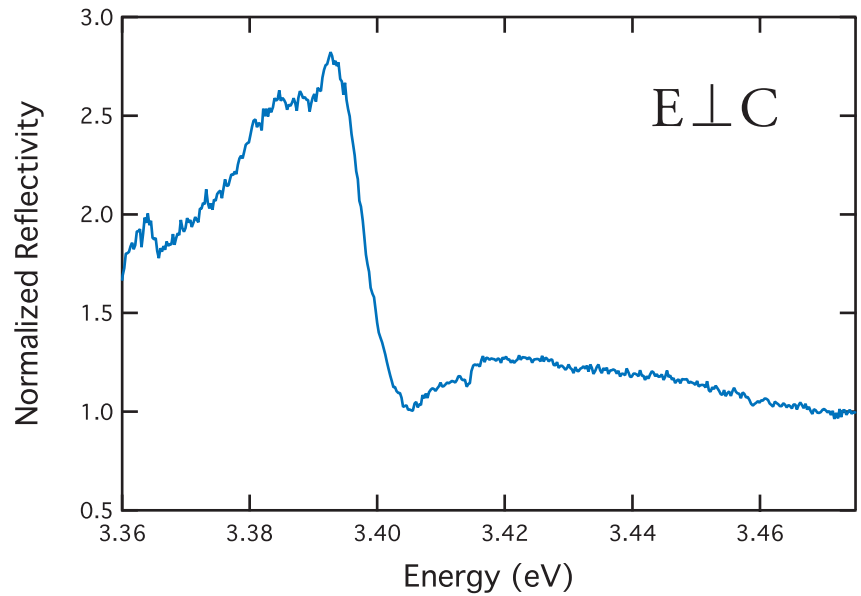

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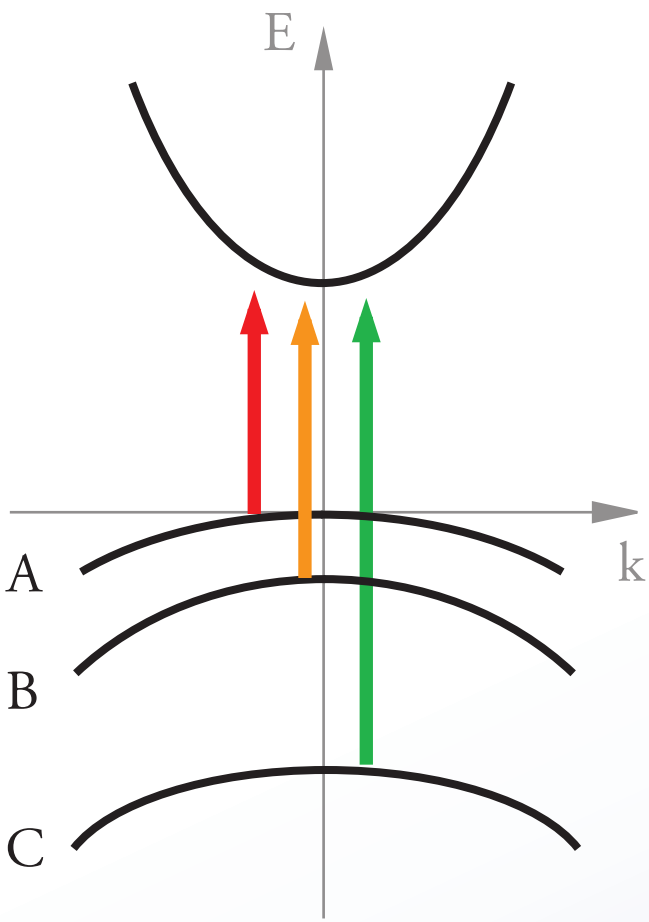

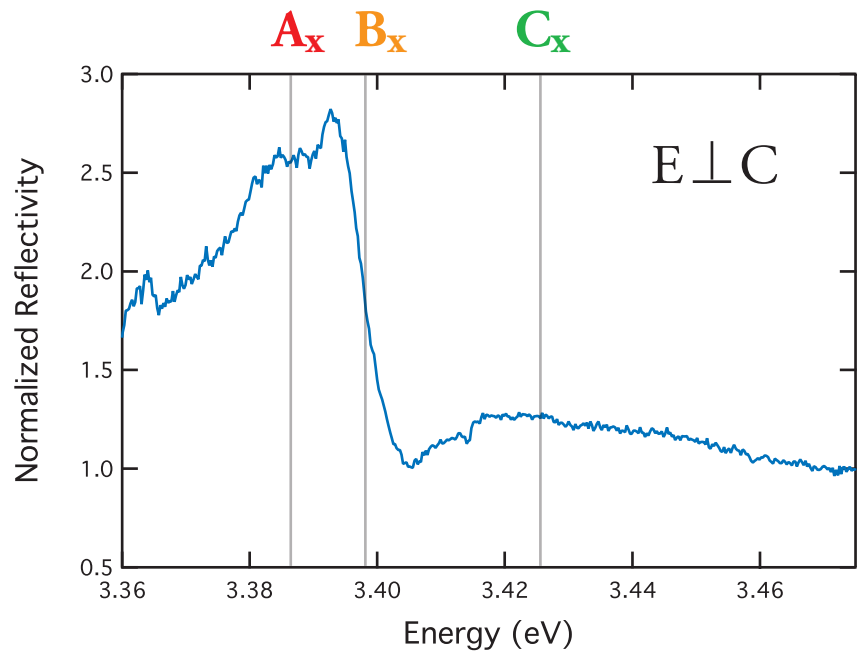

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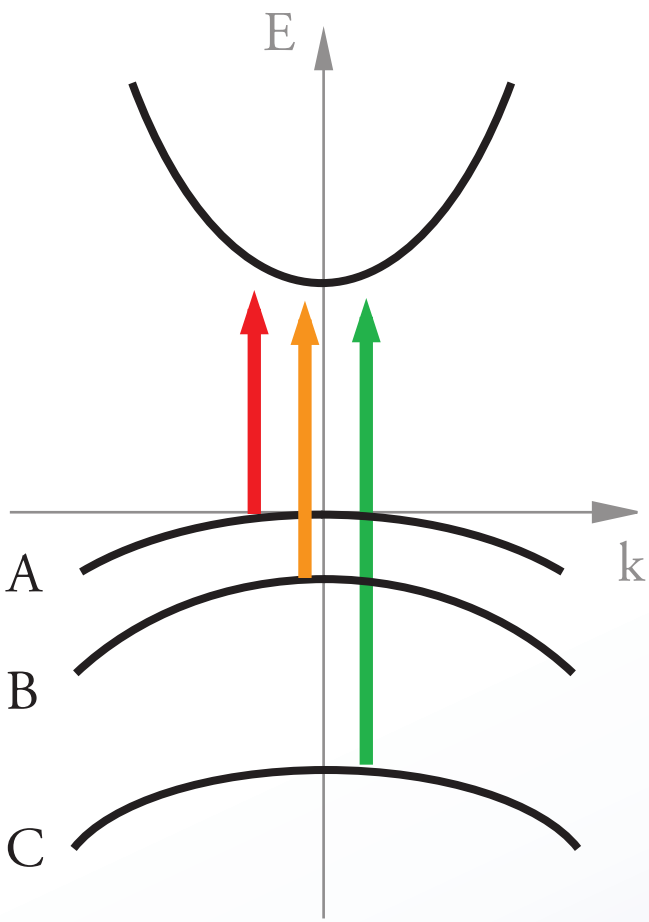

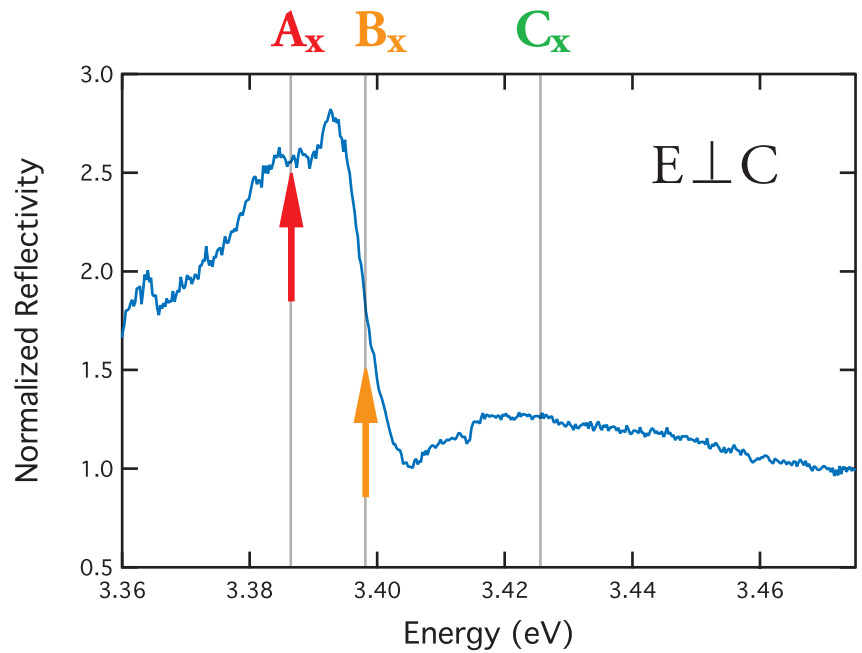

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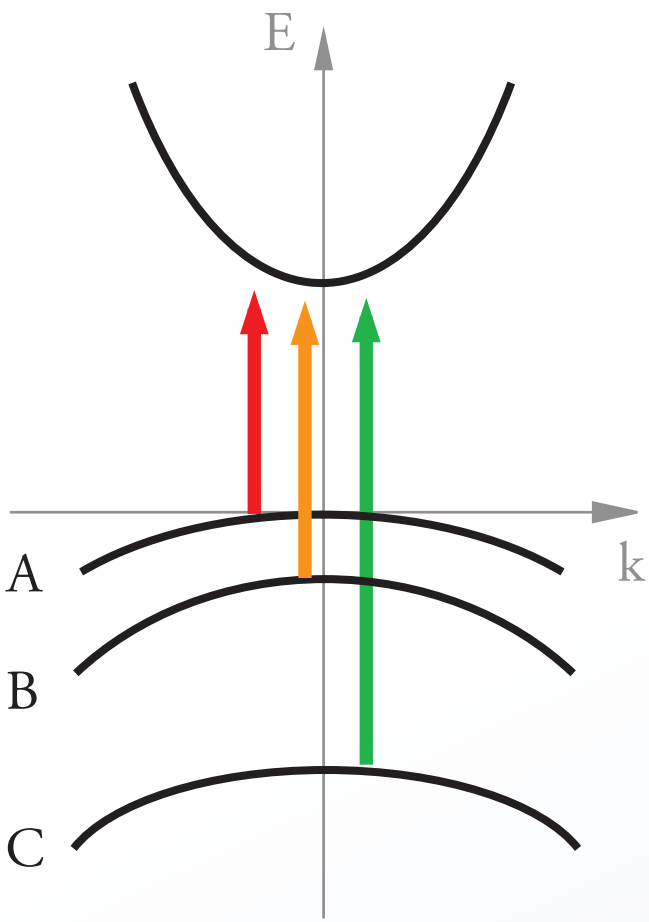

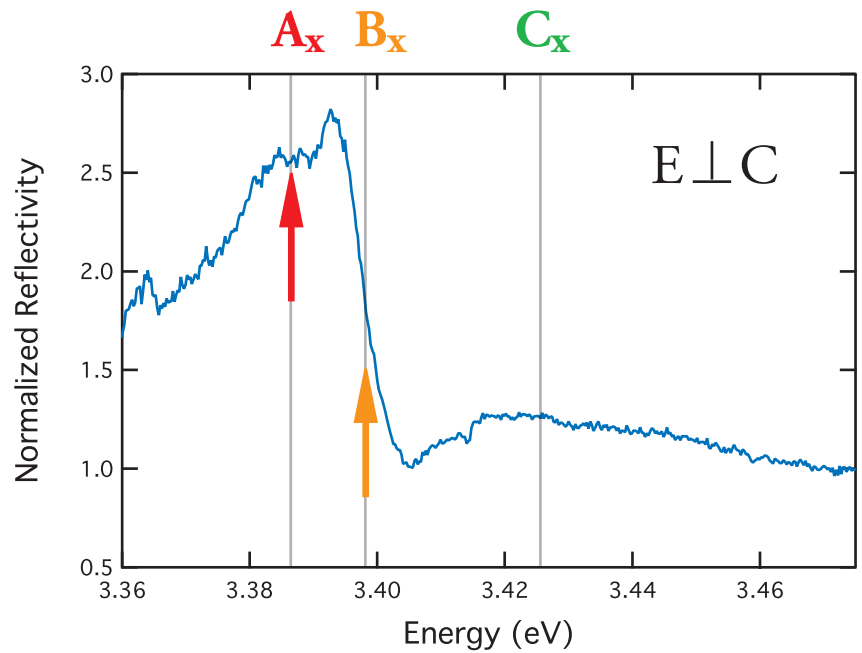

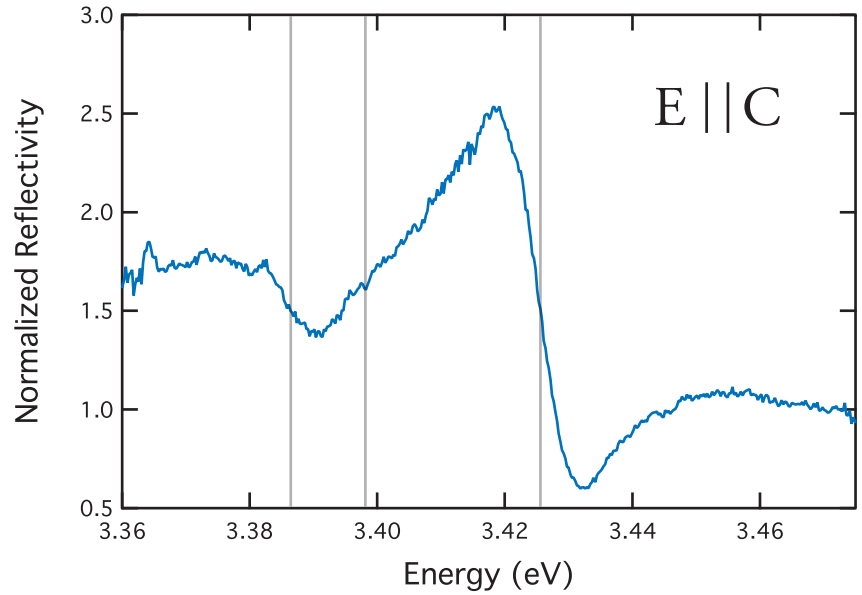

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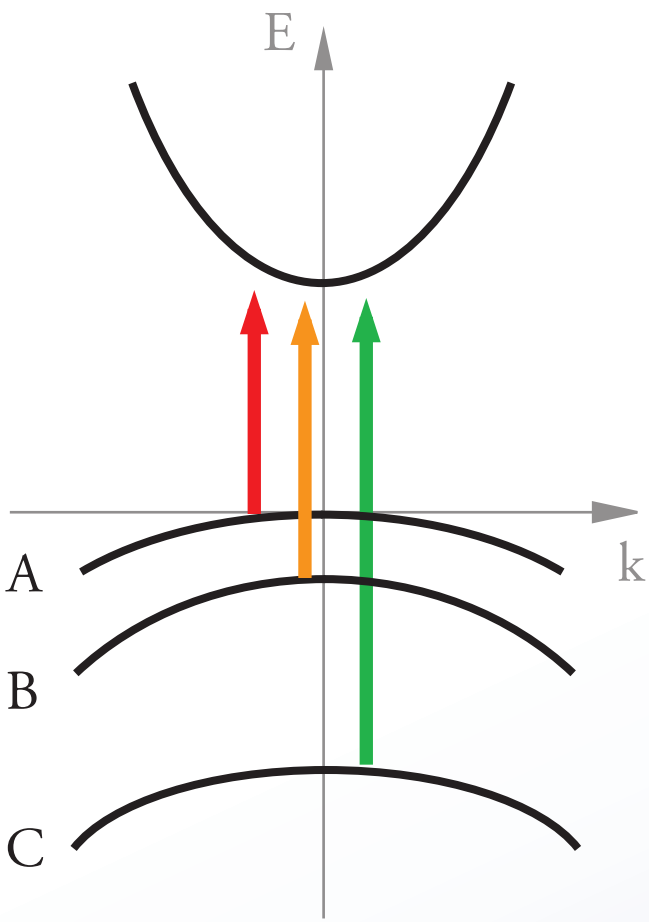
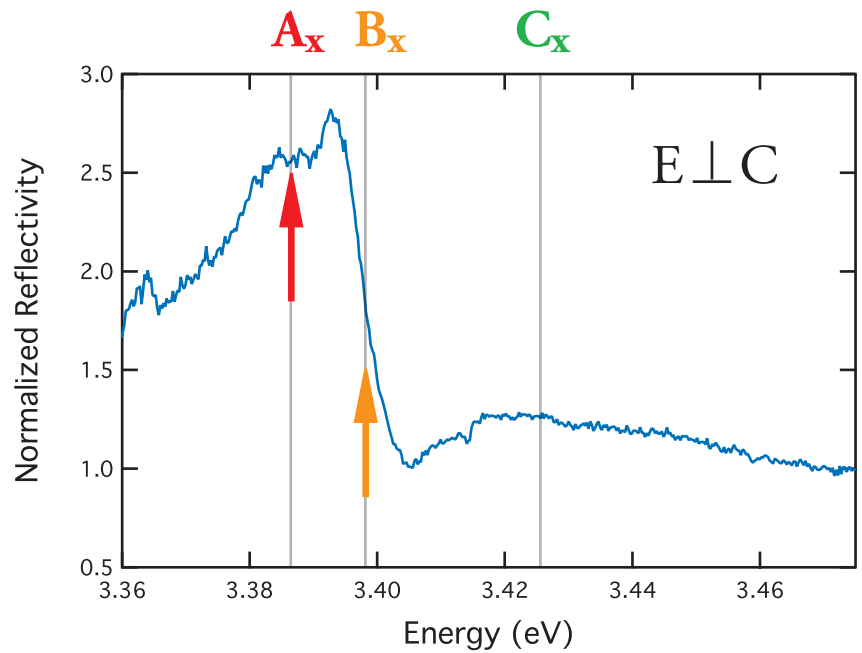
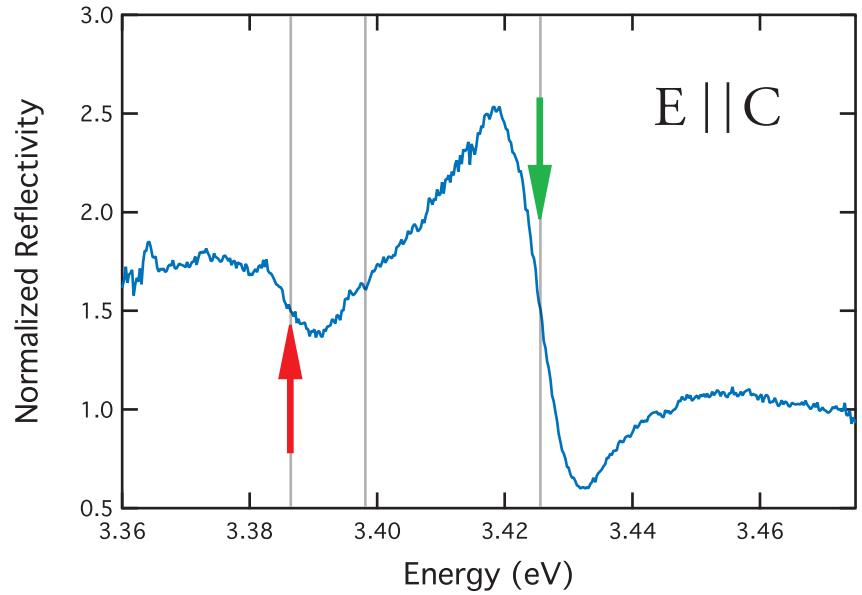
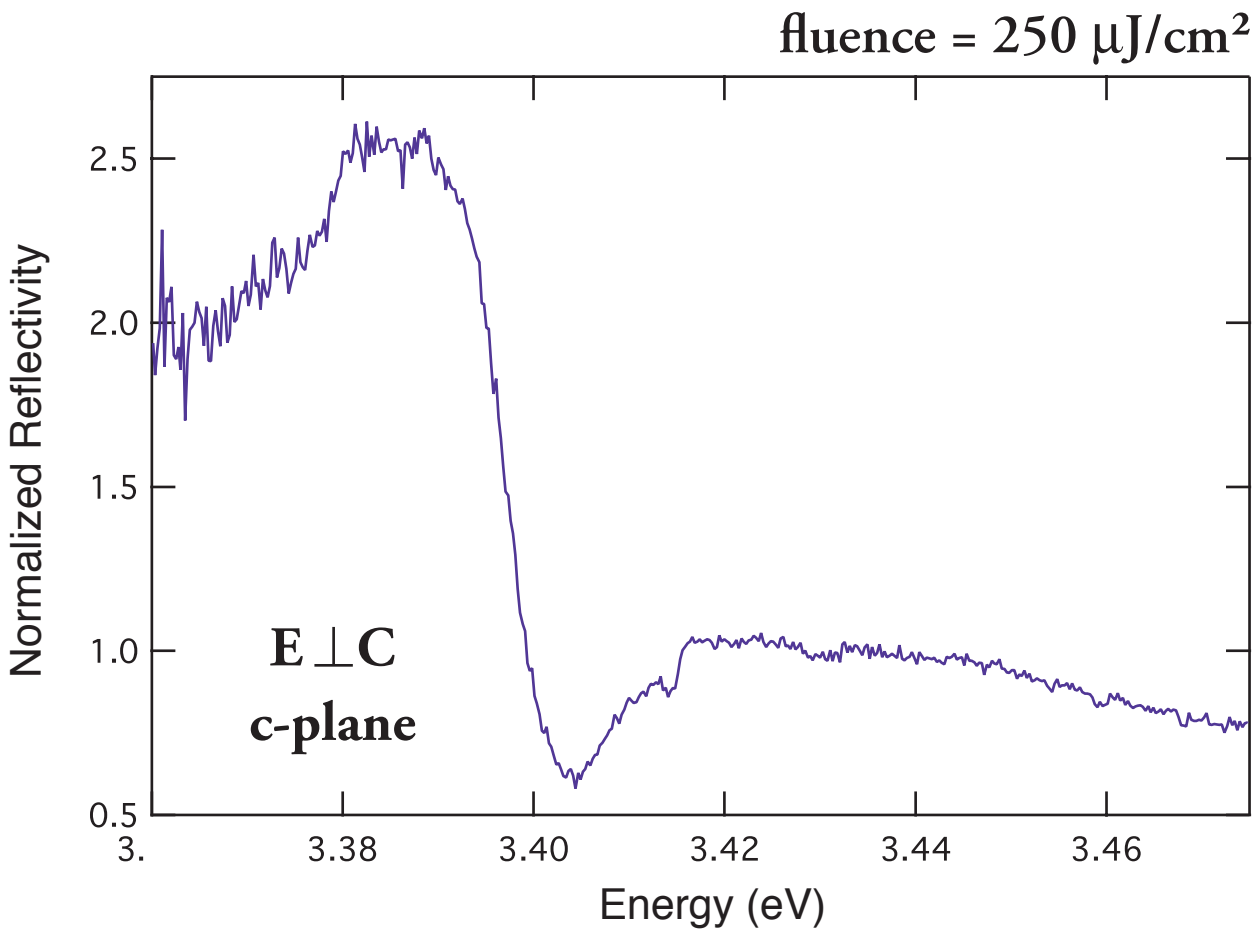






Are there exciton dynamics as a result of fs-excitation?

Time-resolved data



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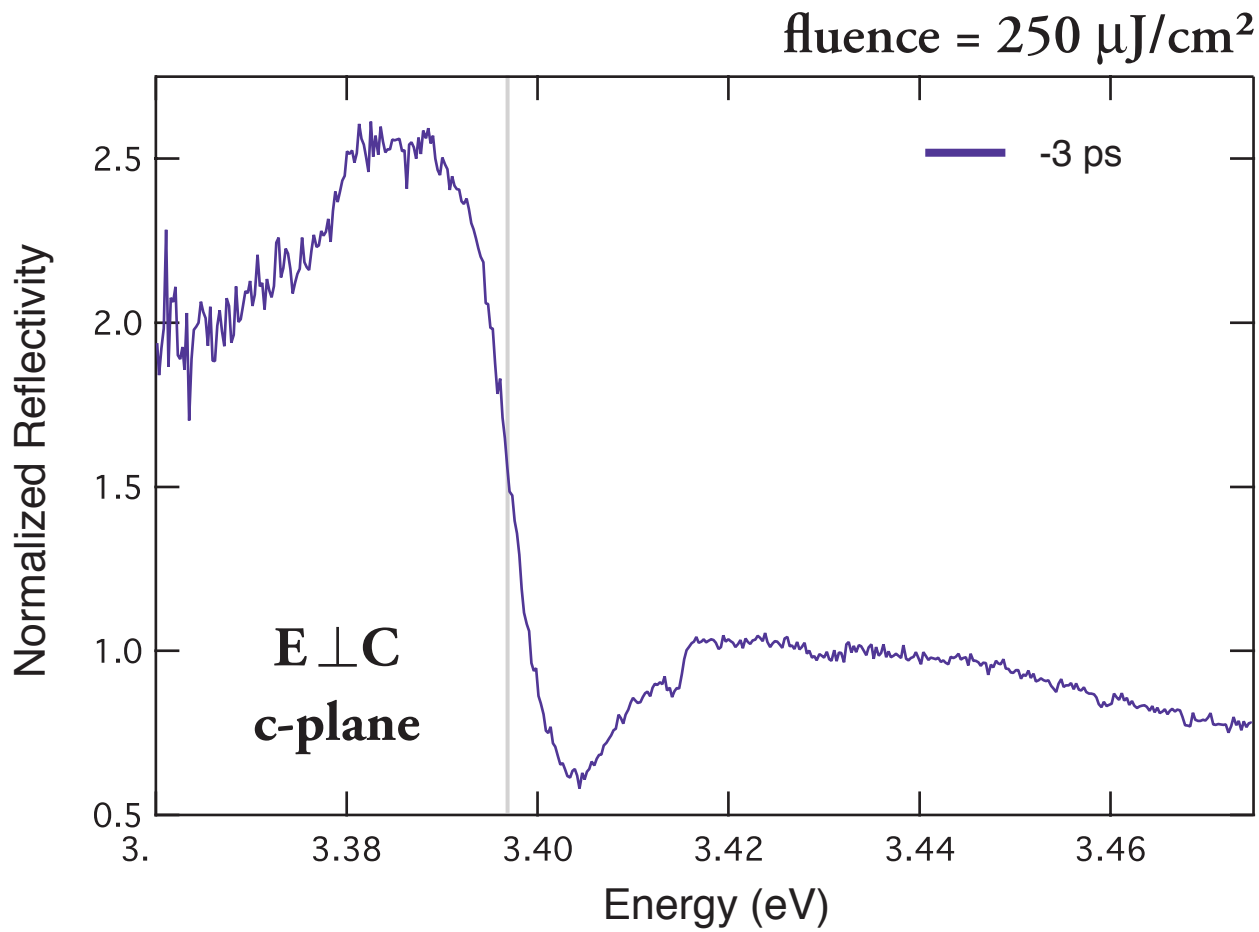
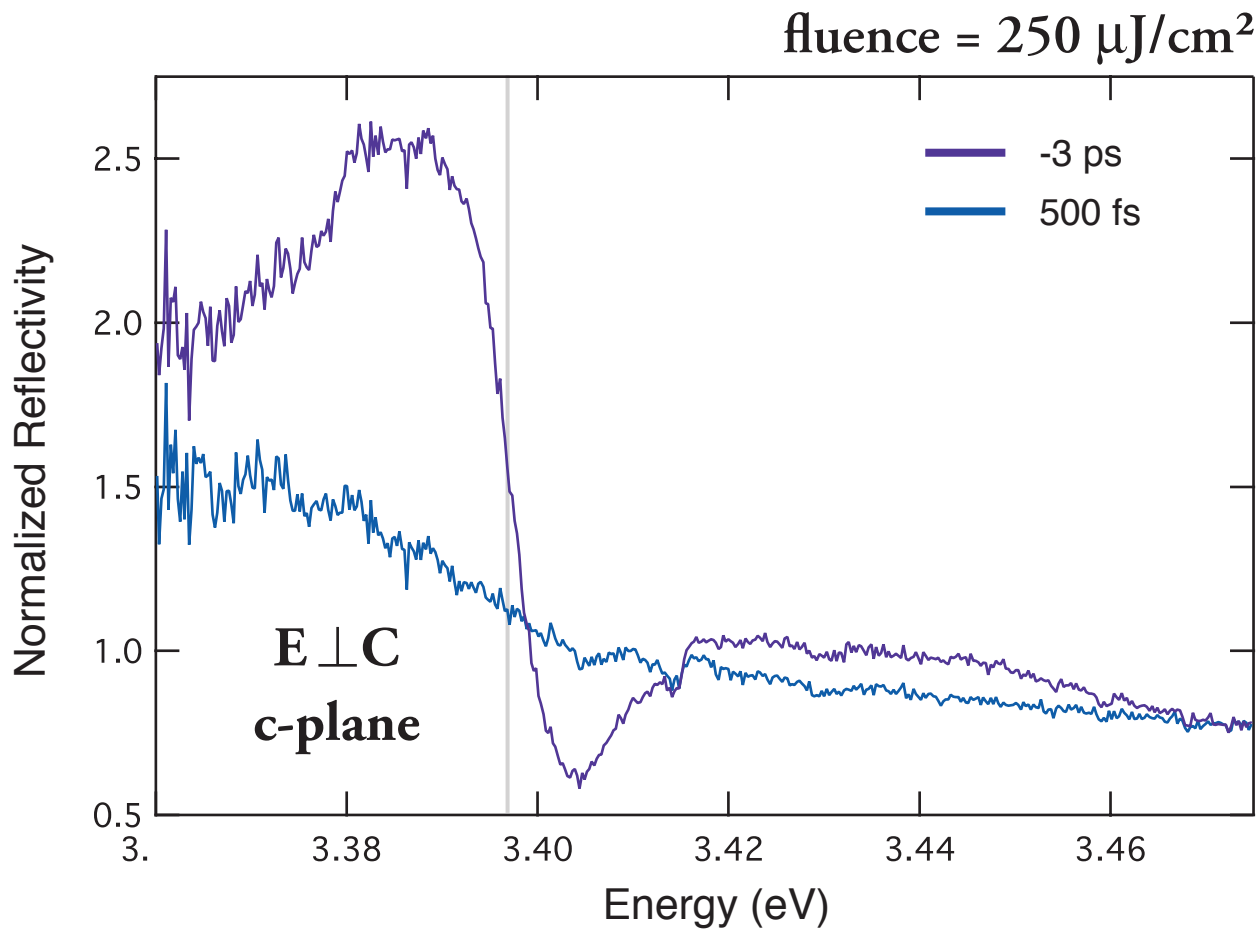


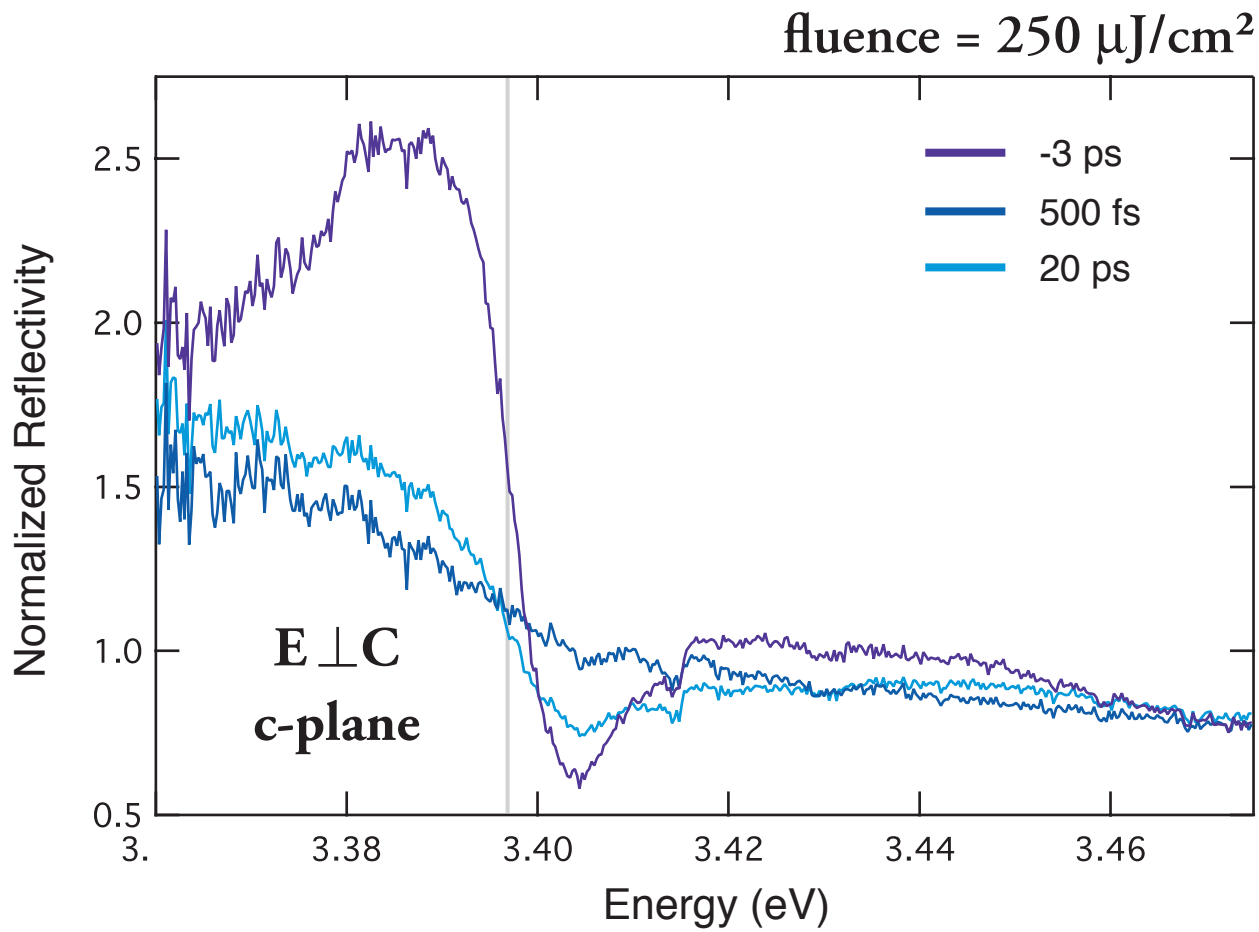
Exciton resonance features present before time zero

Time-resolved data



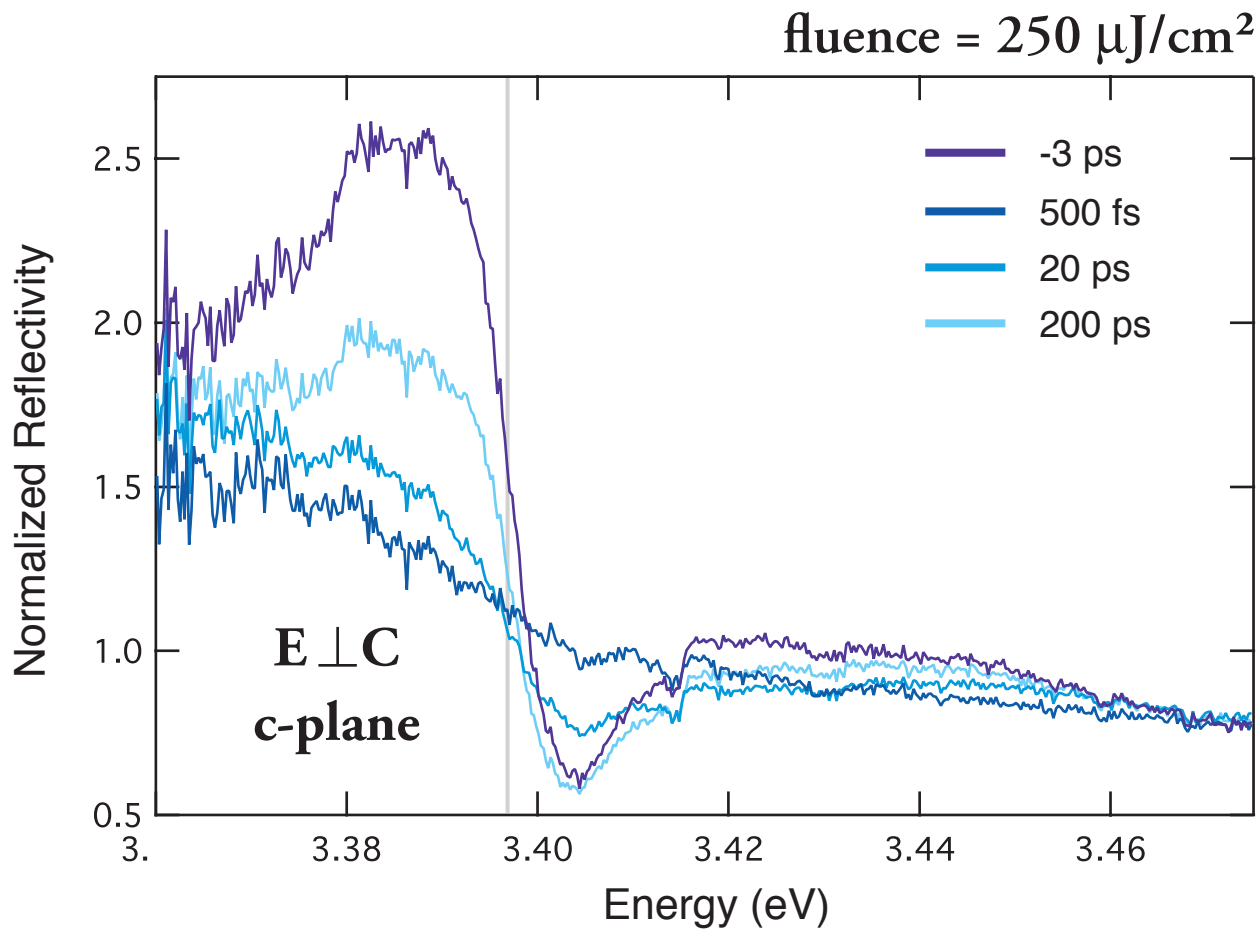
Resonance completely damps out after hundreds of femtoseconds

Time-resolved data



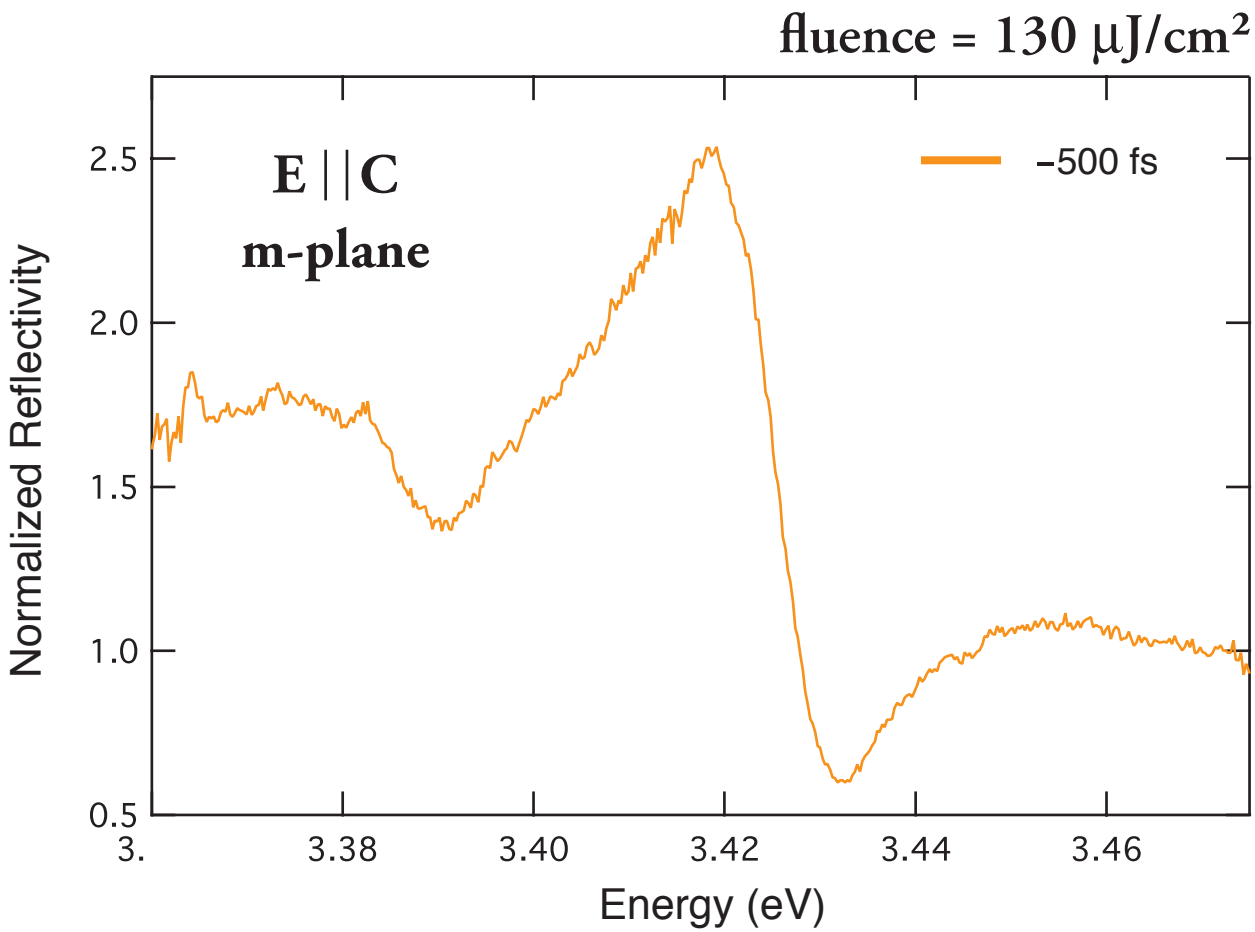
As carriers recombine, Coulomb screening reduces

Time-resolved data



Exciton resonance recovers slowly over hundreds of picoseconds

Time-resolved data



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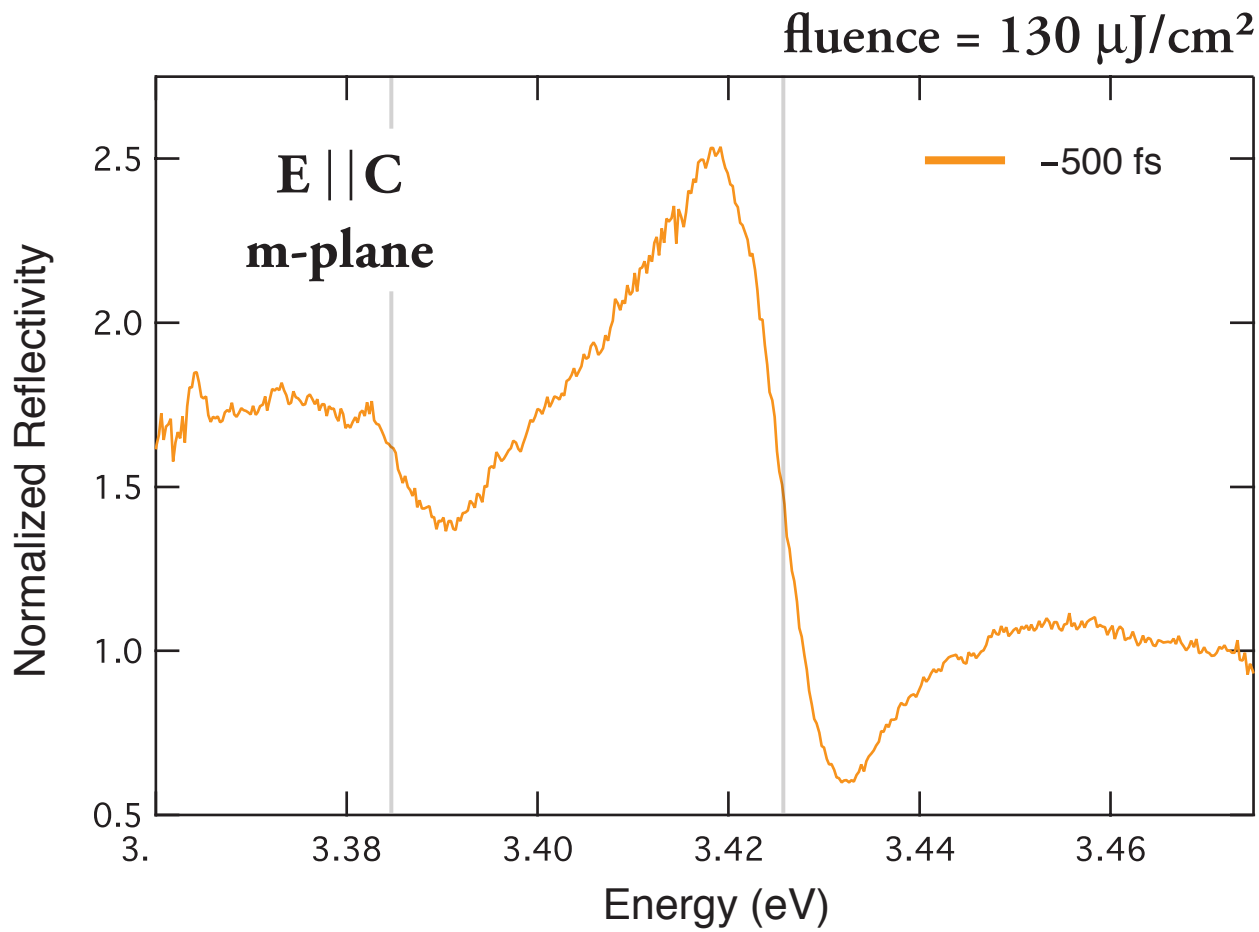
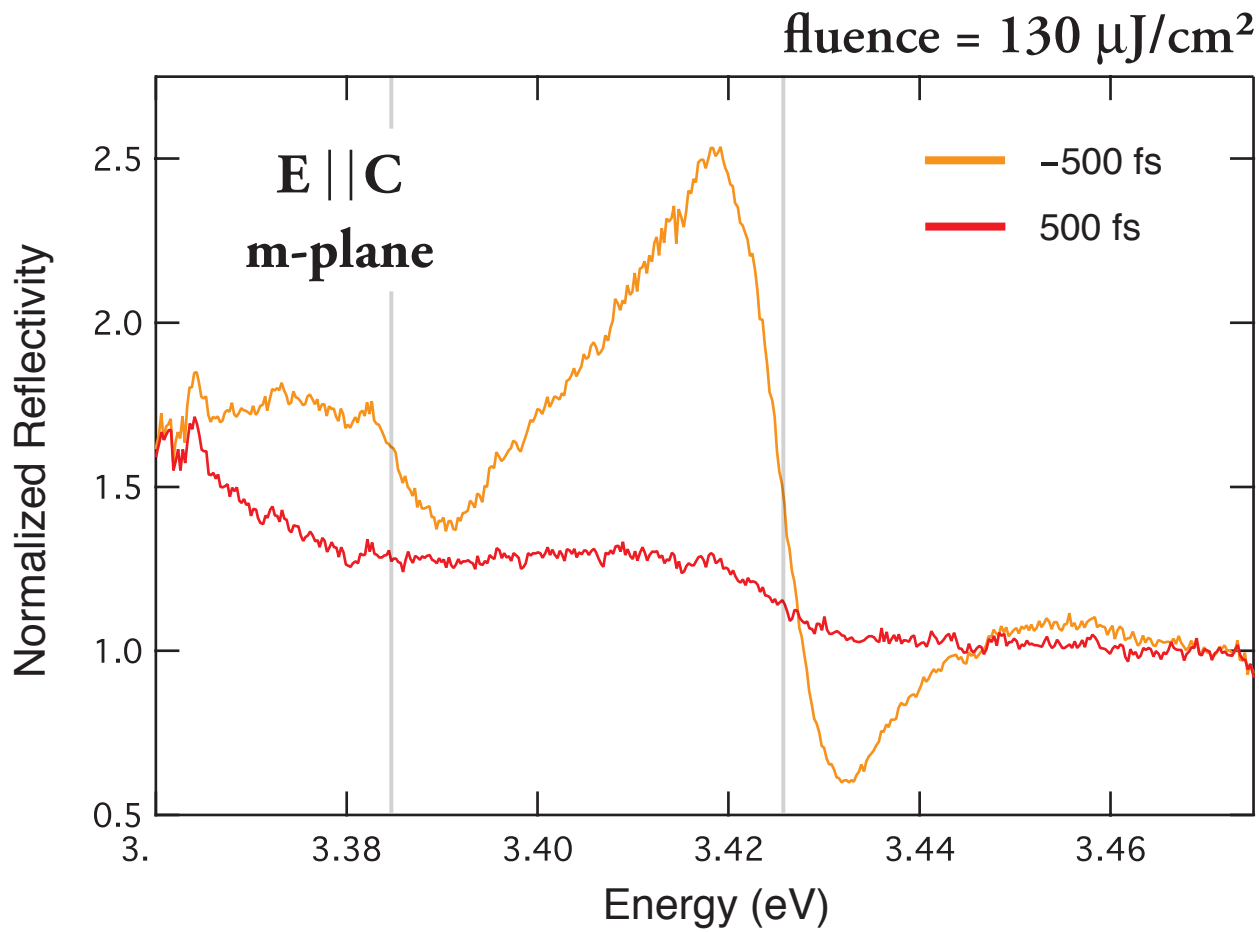


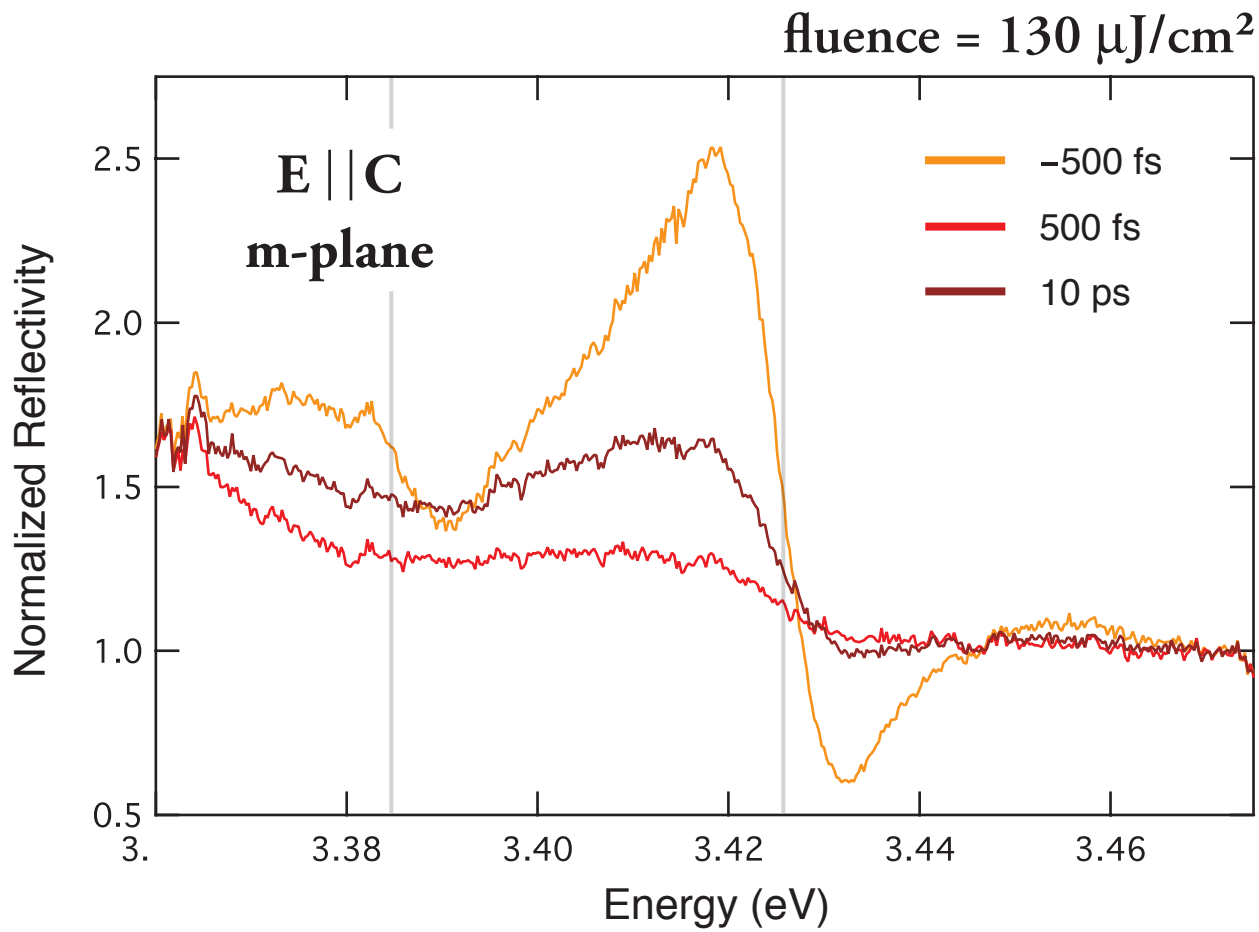
Clear exciton resonance features before time zero

Time-resolved data



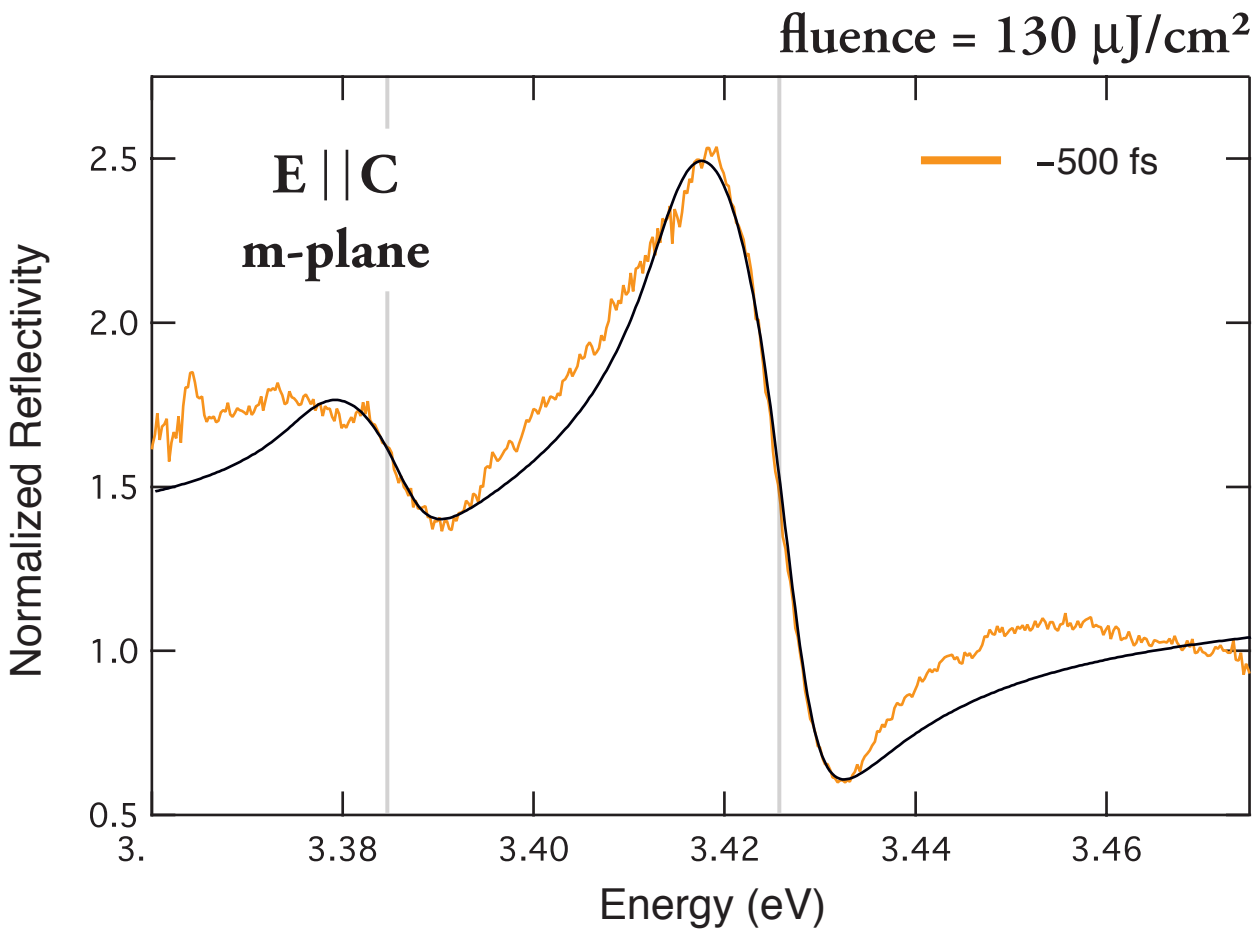
Complete dampening of exciton resonance

Time-resolved data



Recovery starts after tens of picoseconds

Time-resolved data



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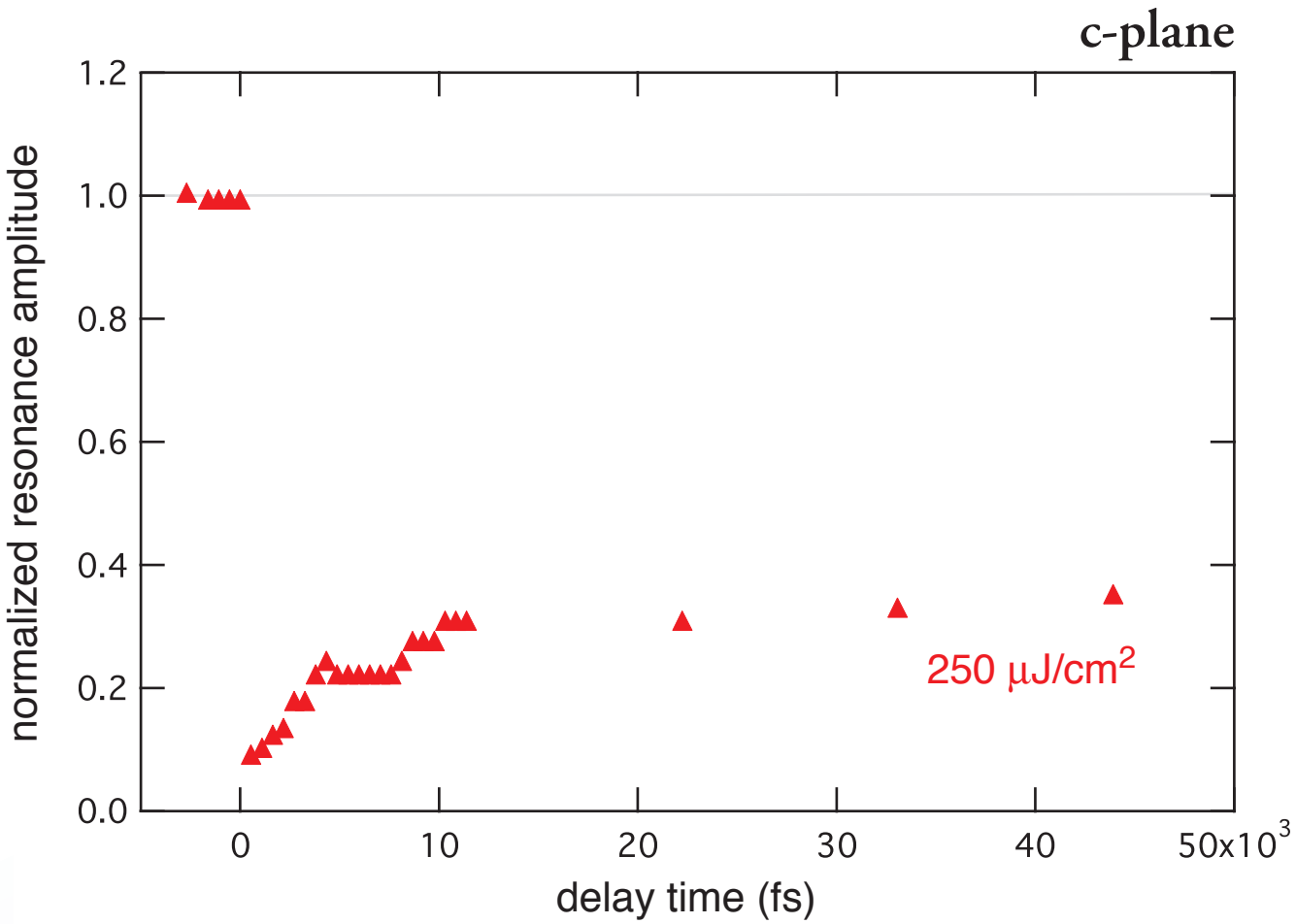

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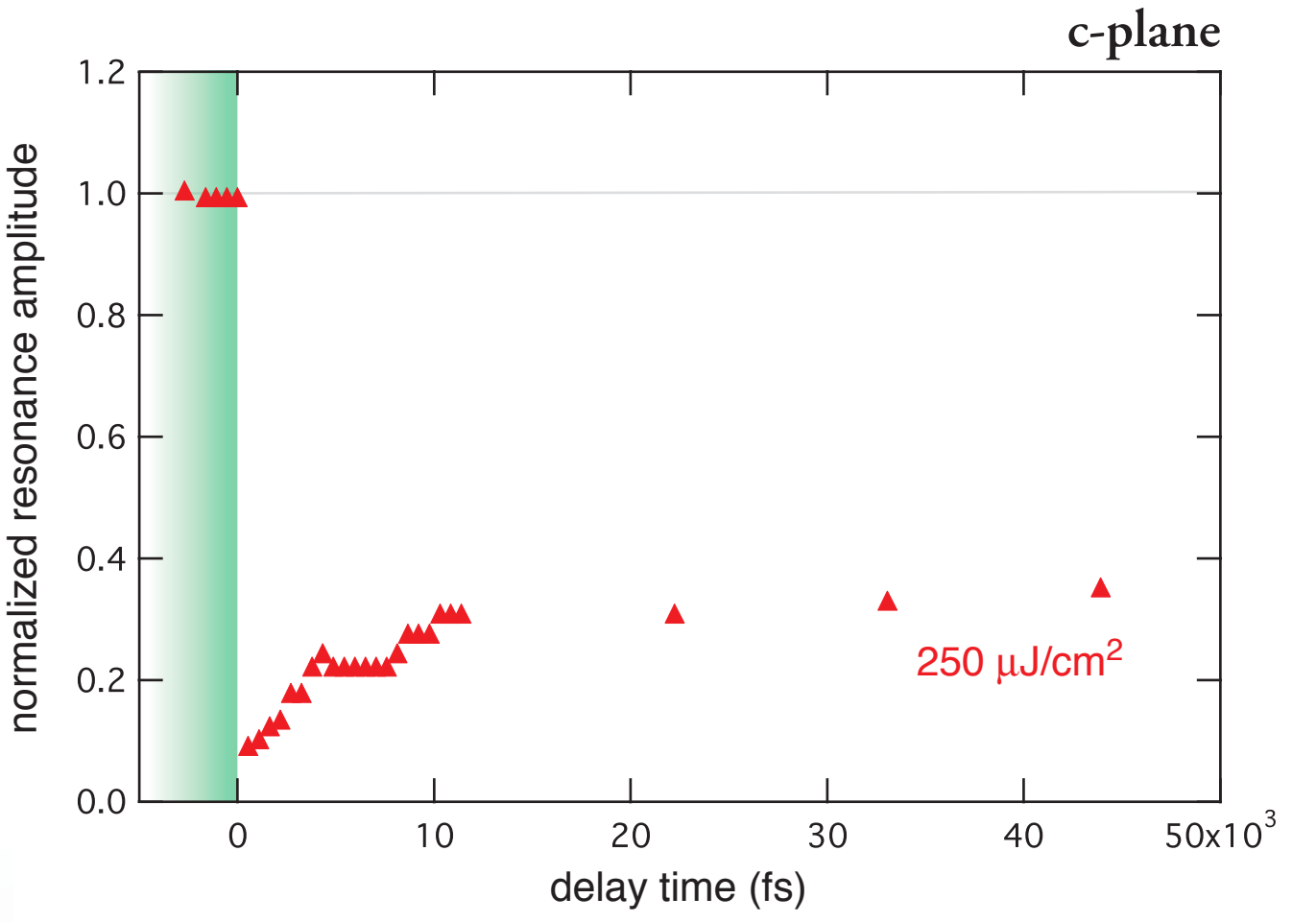
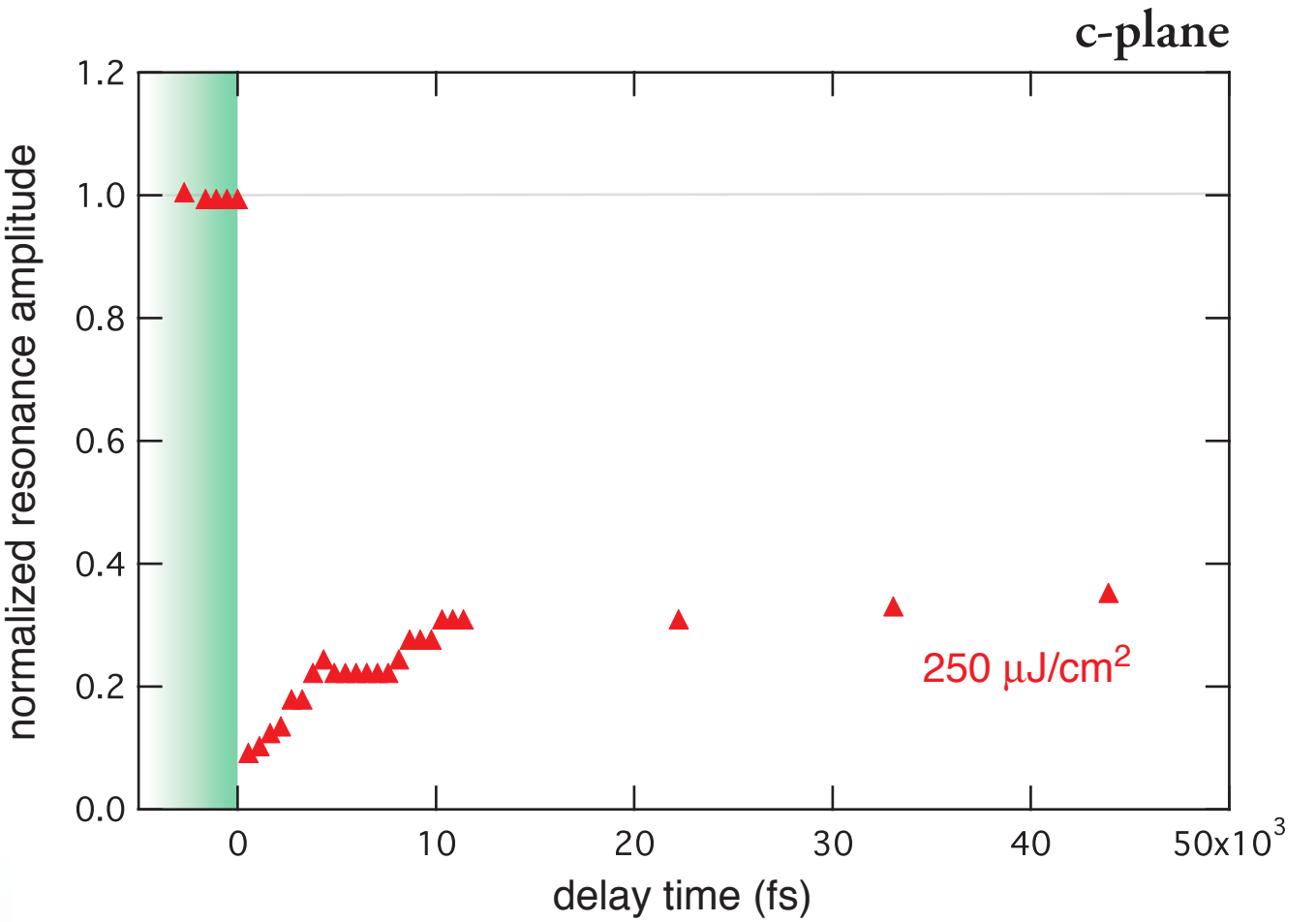


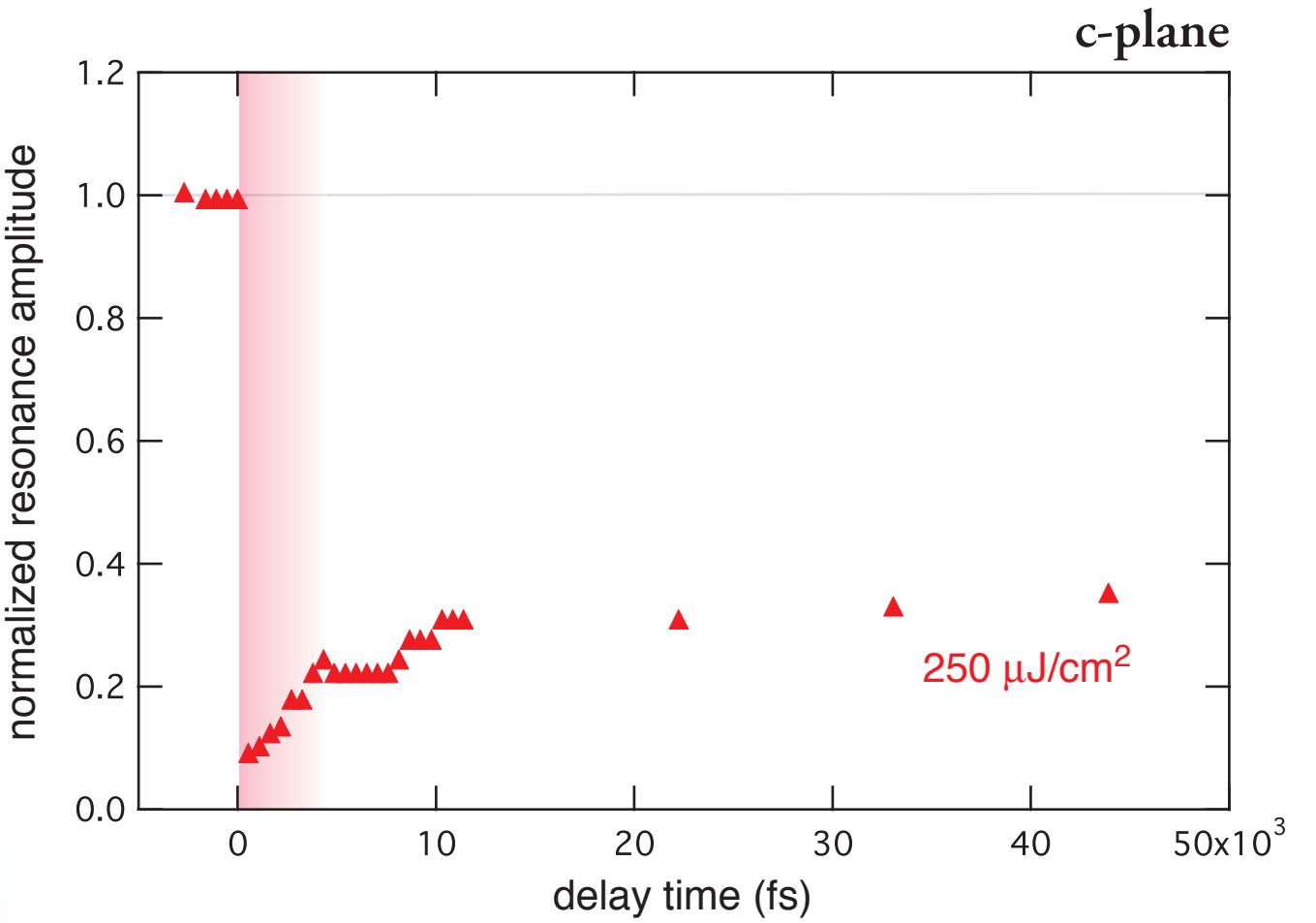
Excitation creates a very high density of carriers around time zero

Time-resolved data



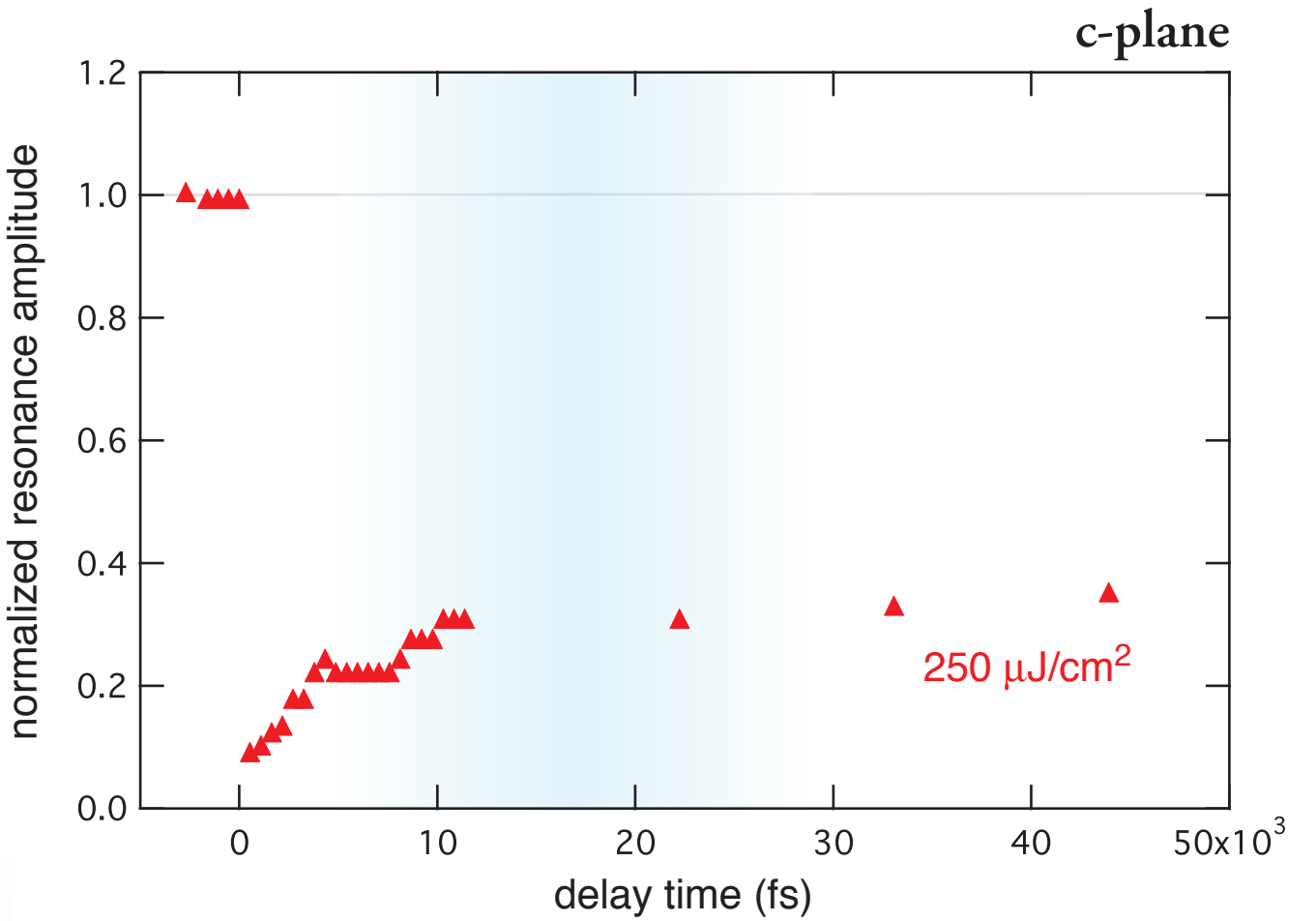
Calculated carrier concentration: $1.6*10^{18}/cm^3$ > Mott density: $5*10^{17}/cm^3$

Time-resolved data



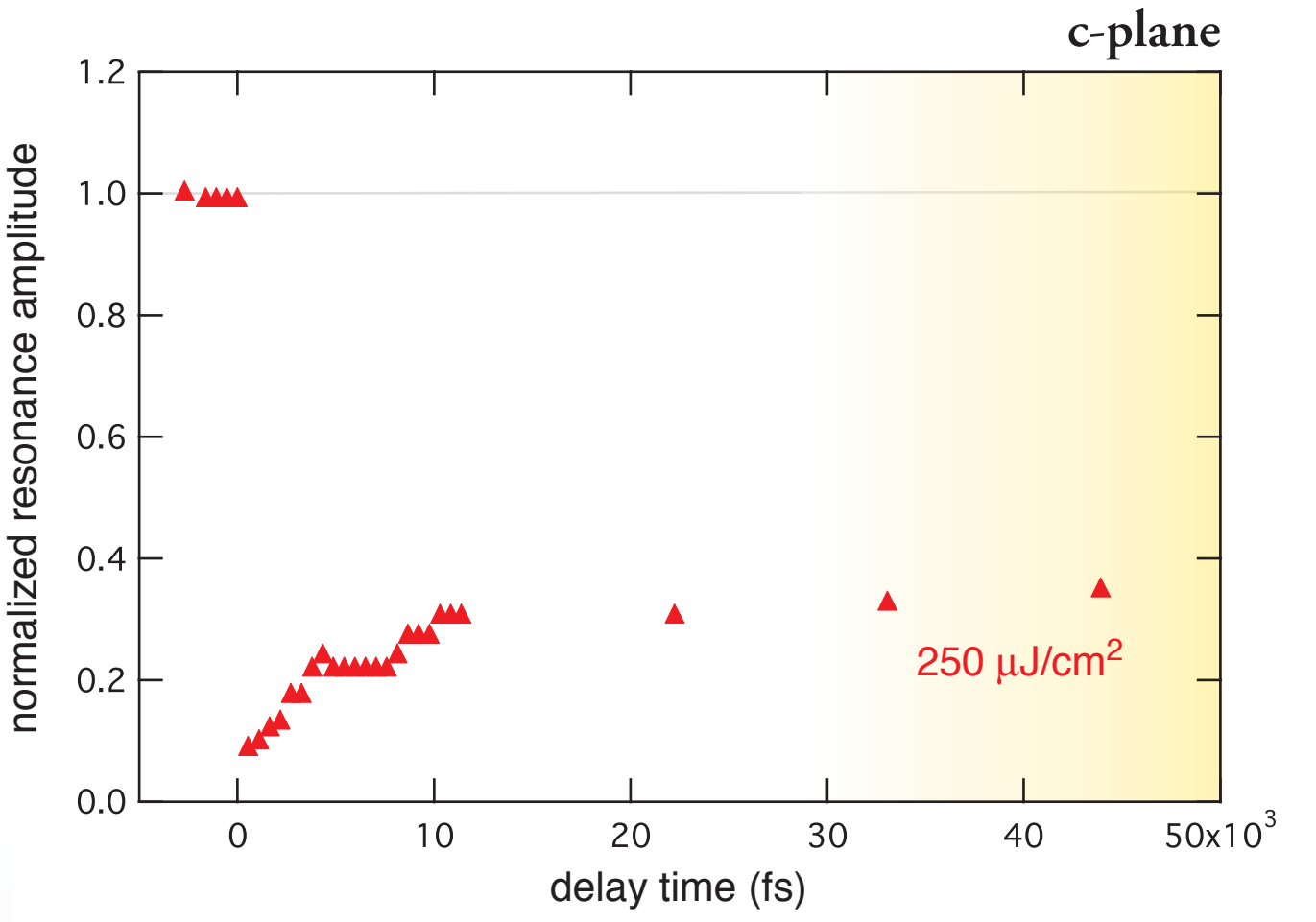
Carrier scattering and recombination will start shortly after excitation

Time-resolved data



As carrier density reduces, Coulomb screening becomes less

Time-resolved data



After tens of picoseconds, exciton resonance slowly recovers

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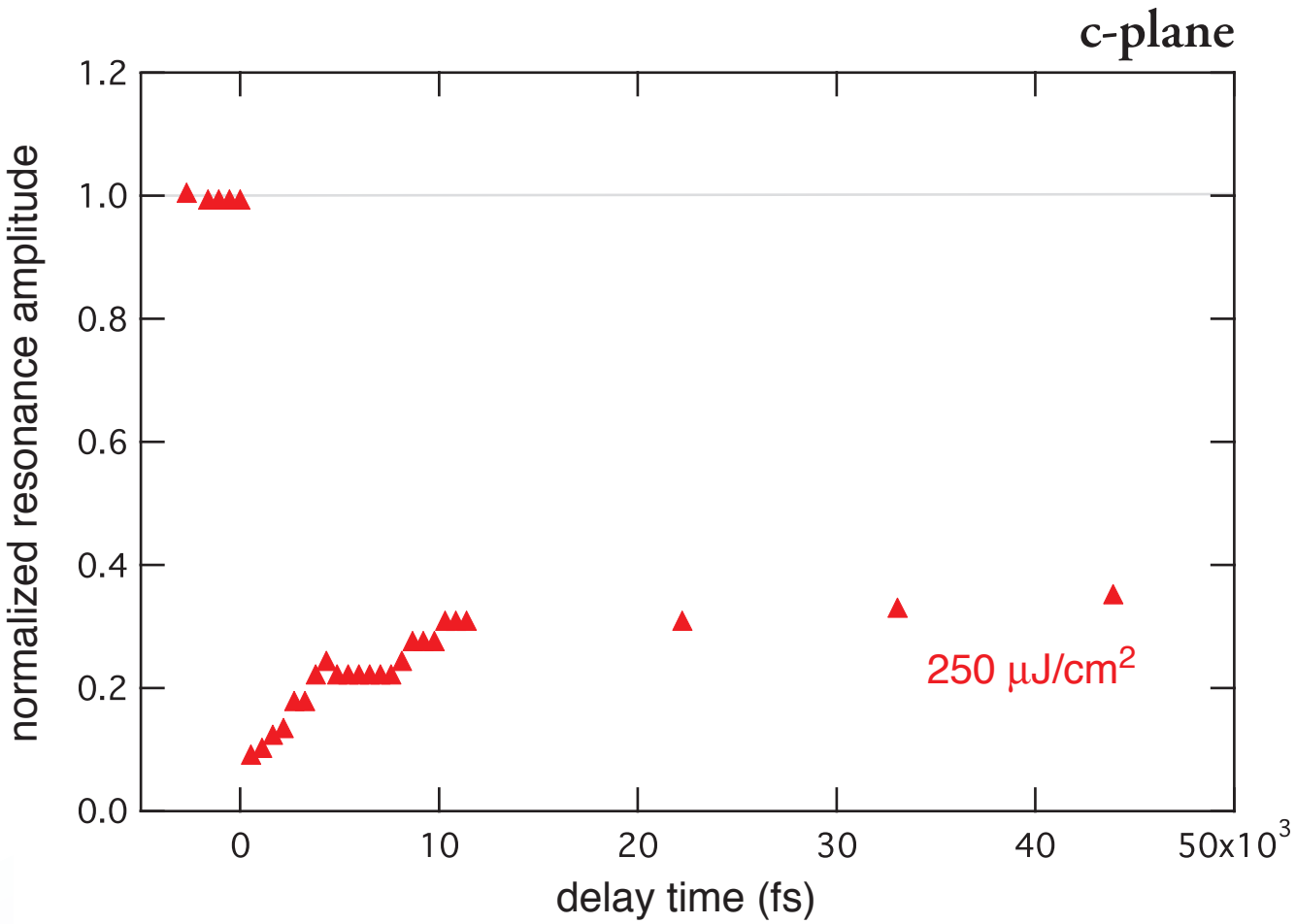

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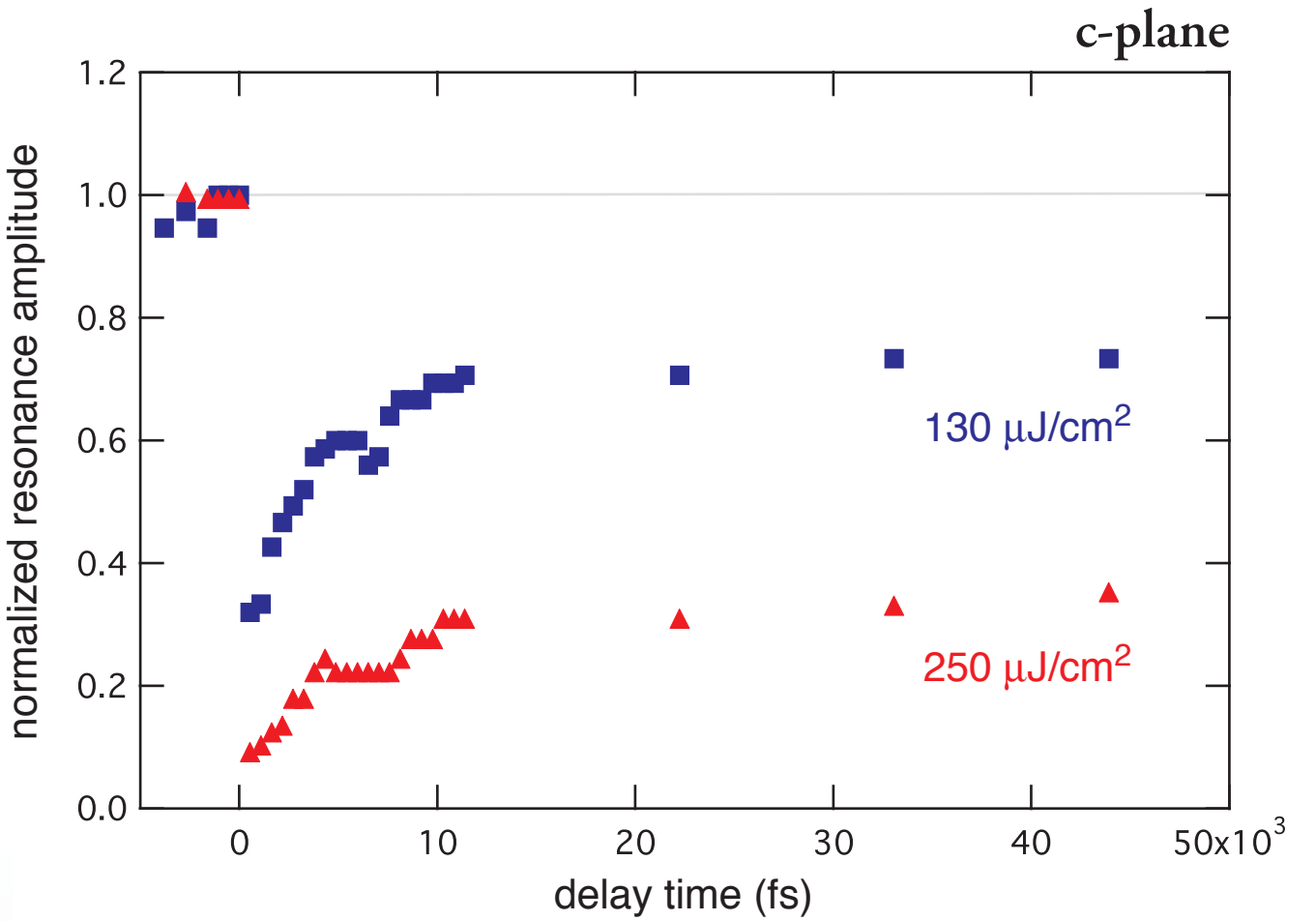
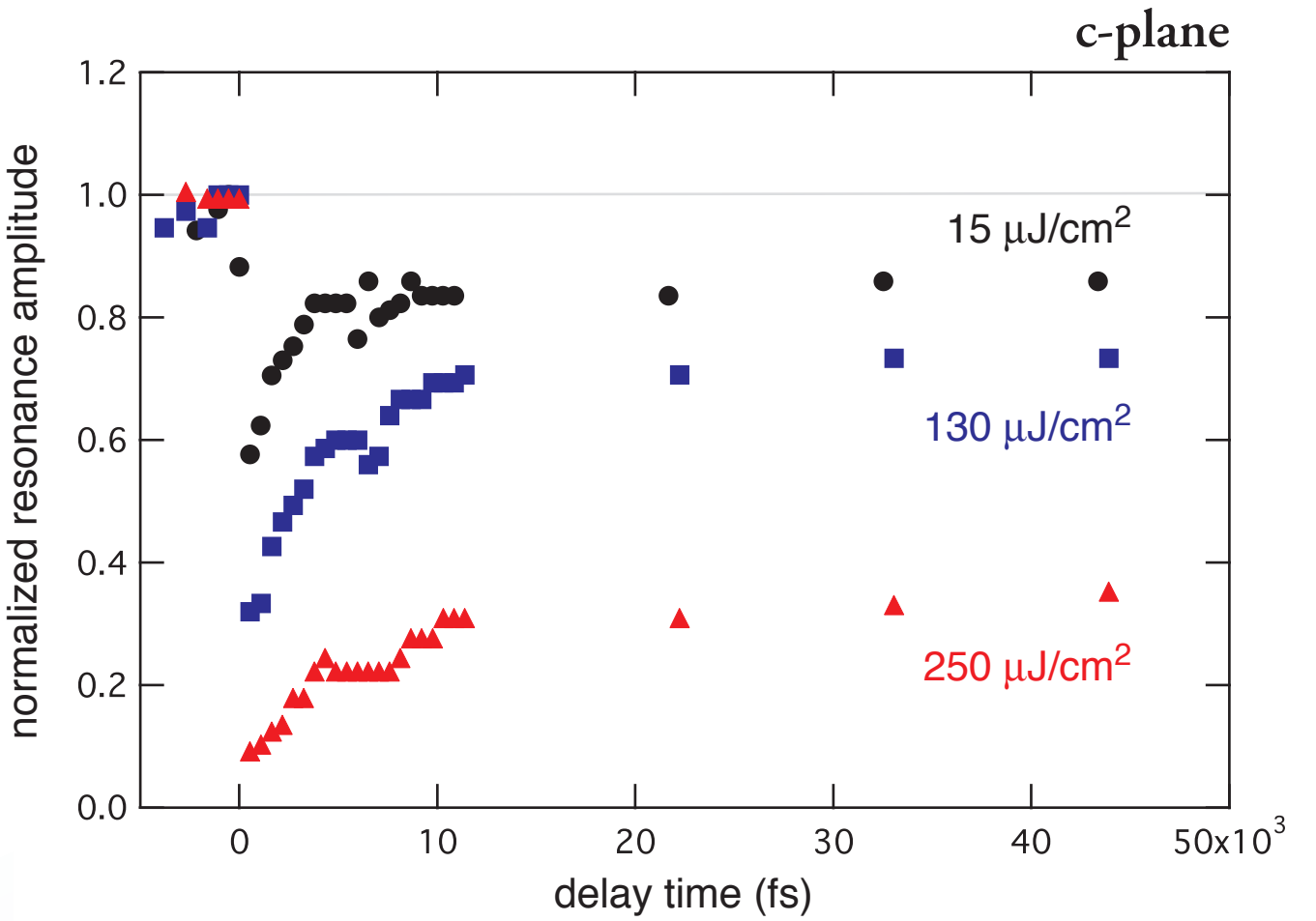


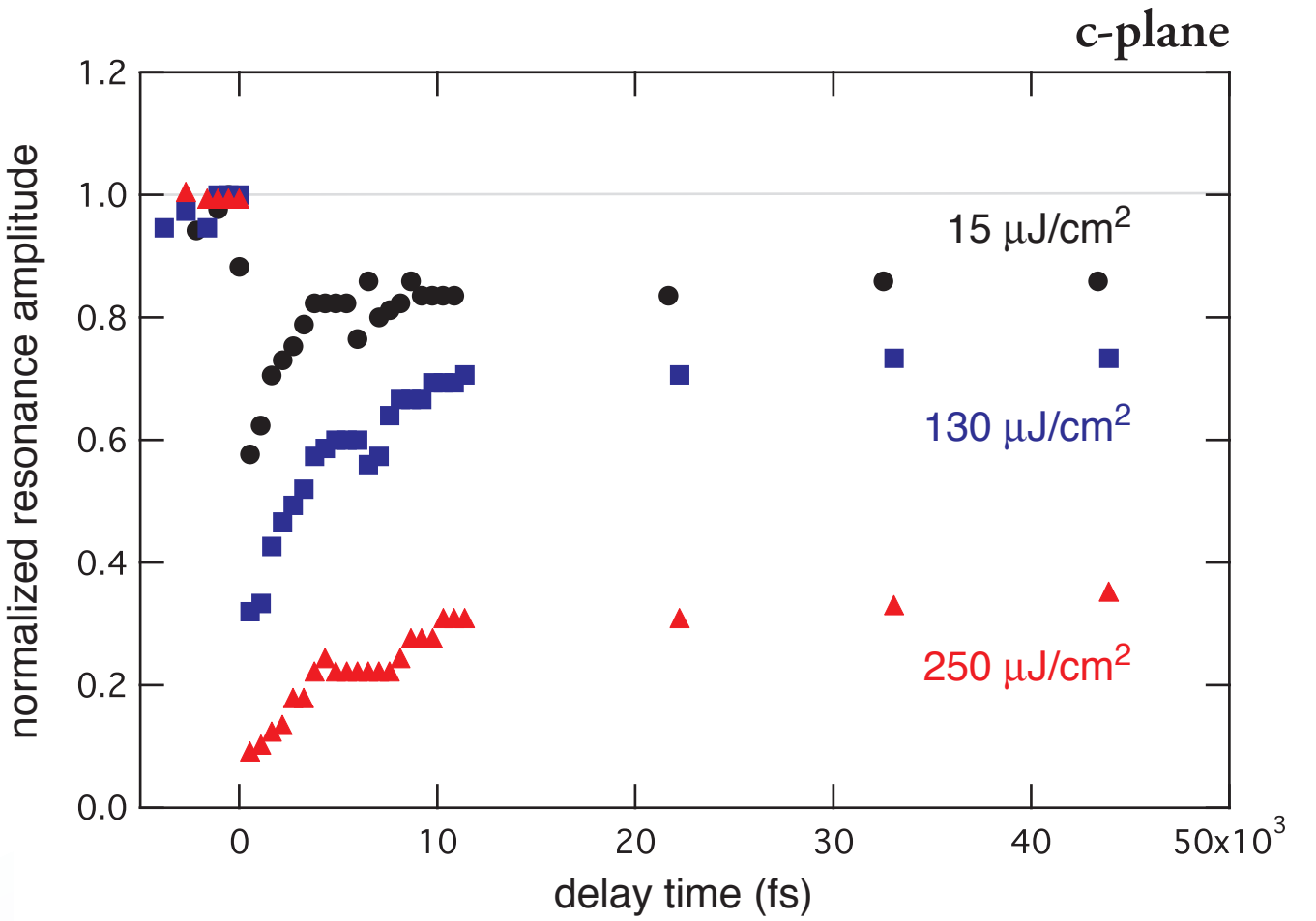
Calculated carrier concentration: $8.4*10^{17}/cm^3$ > Mott density: $5*10^{17}/cm^3$

Time-resolved data



Calculated carrier concentration: $9.6*10^{16}/cm^3$ > Mott density: $5*10^{17}/cm^3$

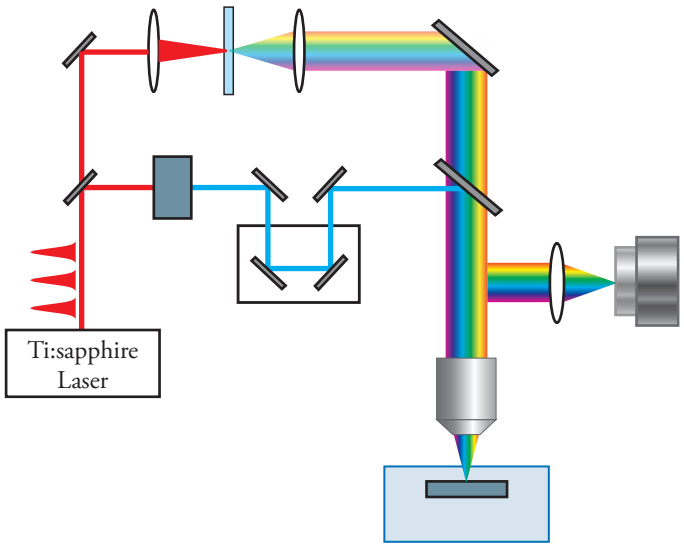
Time-resolved data



Excitons contribute to lasing, but it is initially dominated by e-h plasma emission

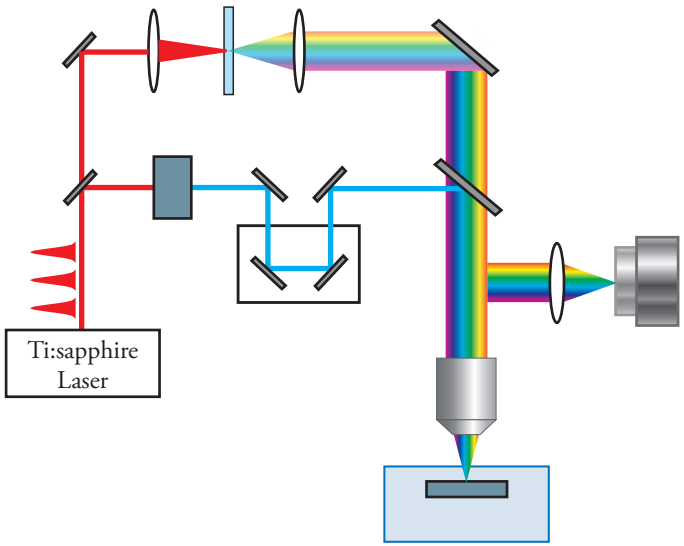
Conclusion

Conclusion

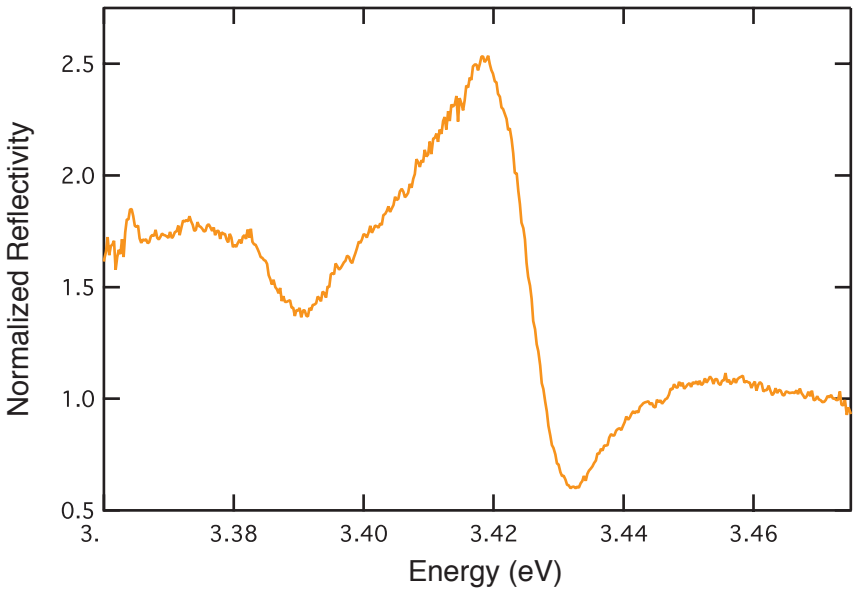


Pump-pump probe reflectivity

Conclusion

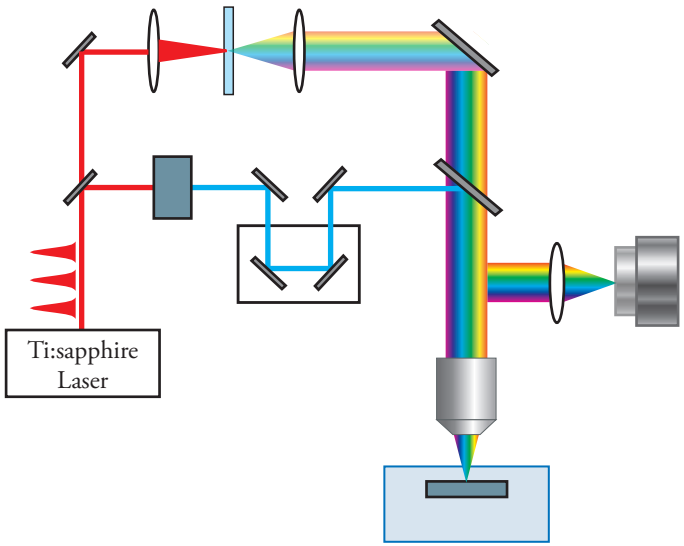


Pump-pump probe reflectivity

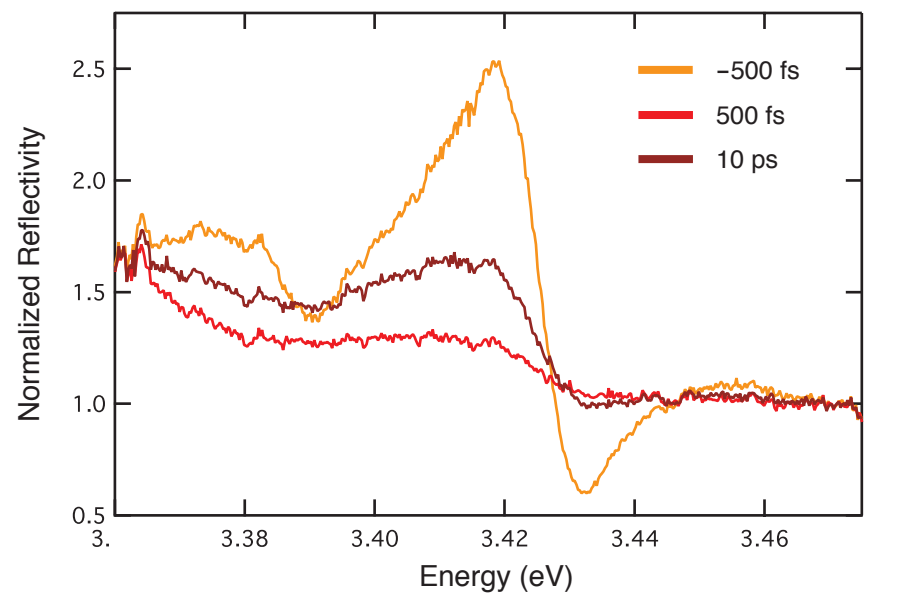


Observe exciton resonances

Conclusion



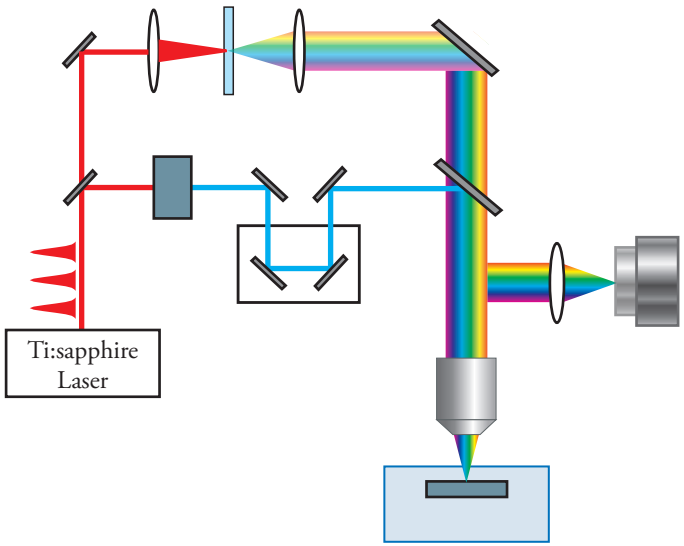
Pump-pump probe reflectivity



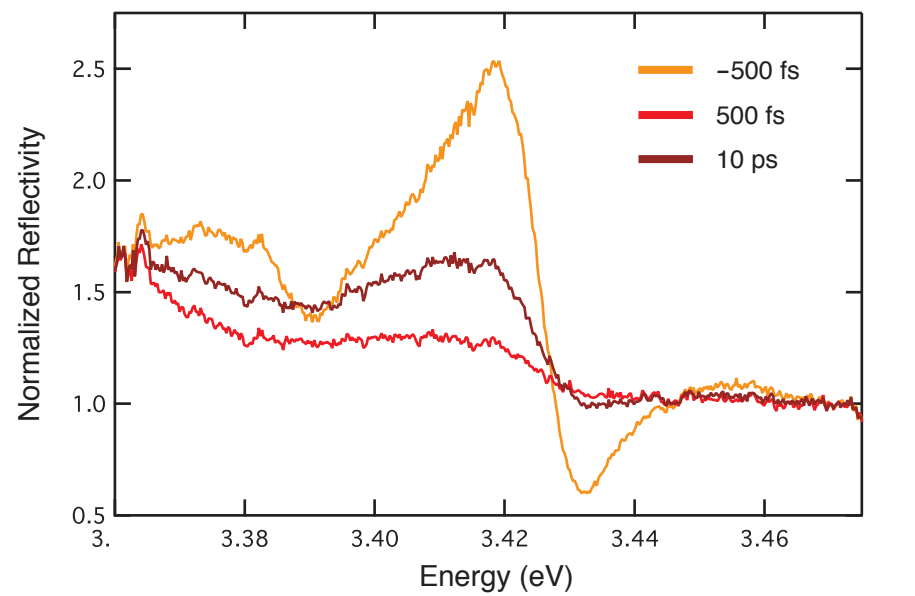
Observe exciton resonances

Monitor excitons' time-resolved response

Conclusion



Pump-pump probe reflectivity



Observe exciton resonances

Monitor excitons' time-resolved response

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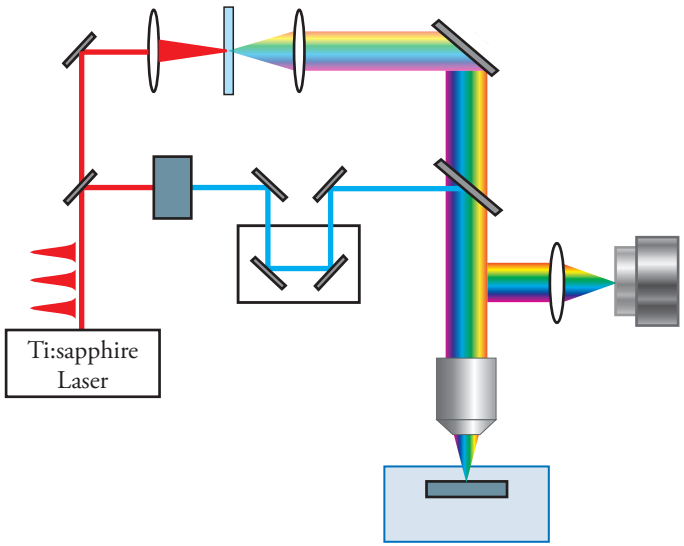
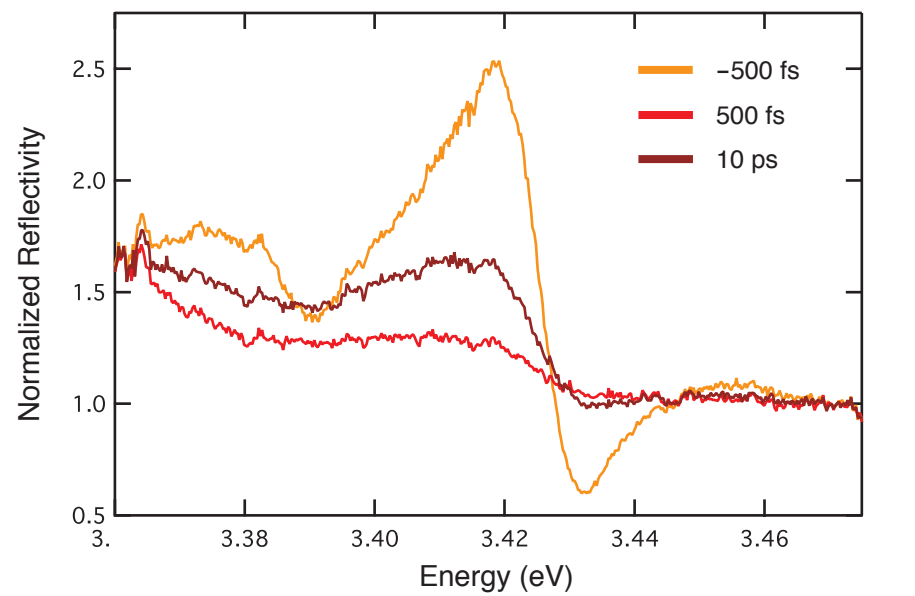
ZnO lasing dominated by electron-hole plasma

Conclusion



Pump-pump probe reflectivity



Observe exciton resonances

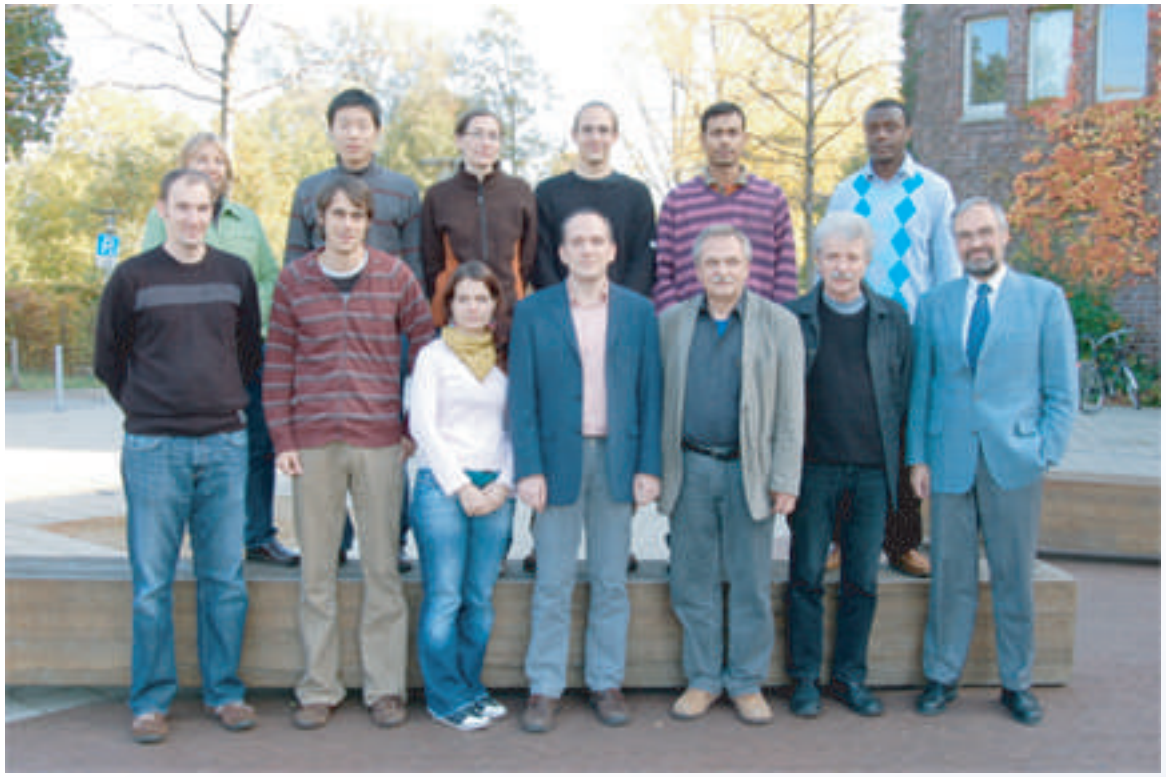
Monitor excitons' time-resolved response

ZnO lasing dominated by electron-hole plasma

Future Work

- Nanostructures
- Low dimensional effects

Acknowledgements



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