



PHASE TRANSFORMATIONS INDUCED BY FEMTOSECOND LASER PULSES

Eric Mazur

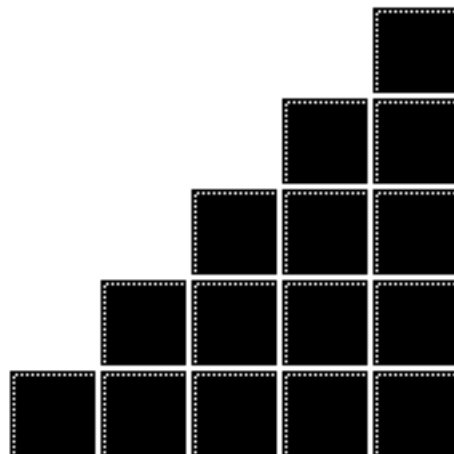
Paul Callan

Albert Kim

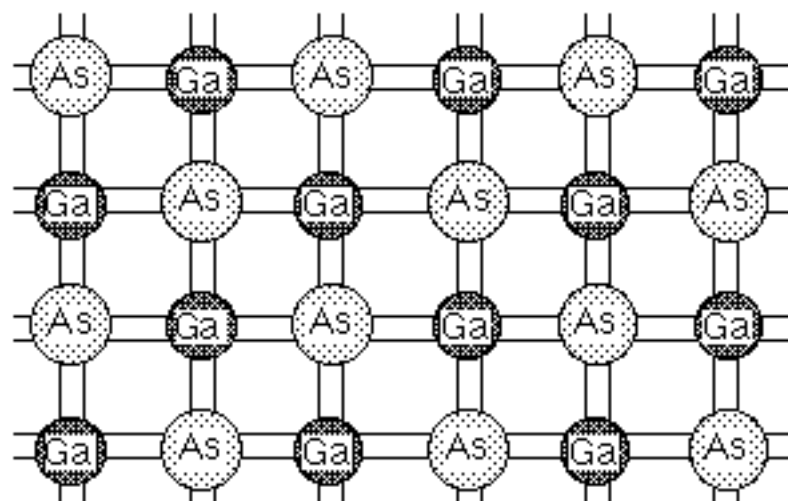
Gordon Conference

Proctor Academy

7 June 1998



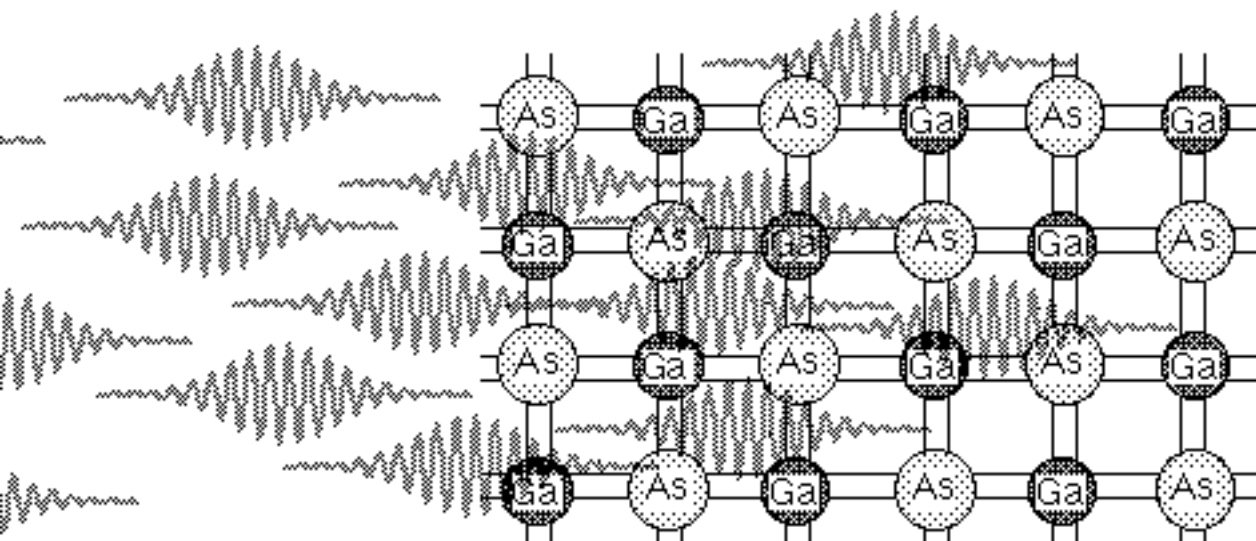
INTRODUCTION



how do femtosecond pulses alter a solid?



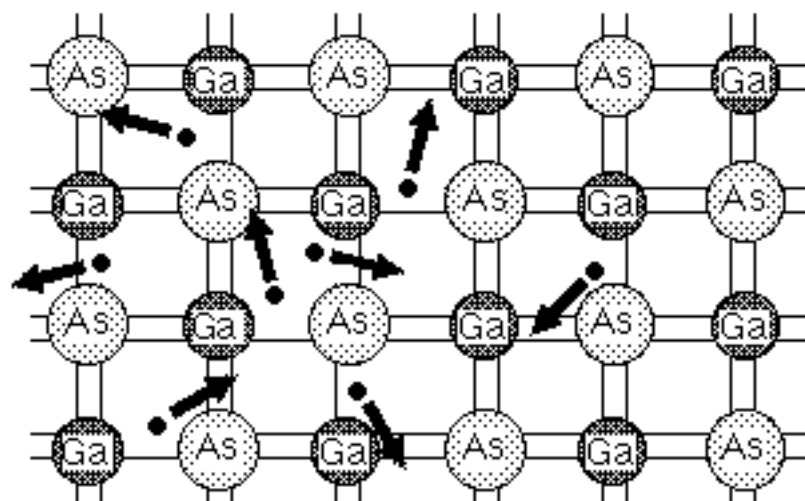
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photons excite valence electrons...



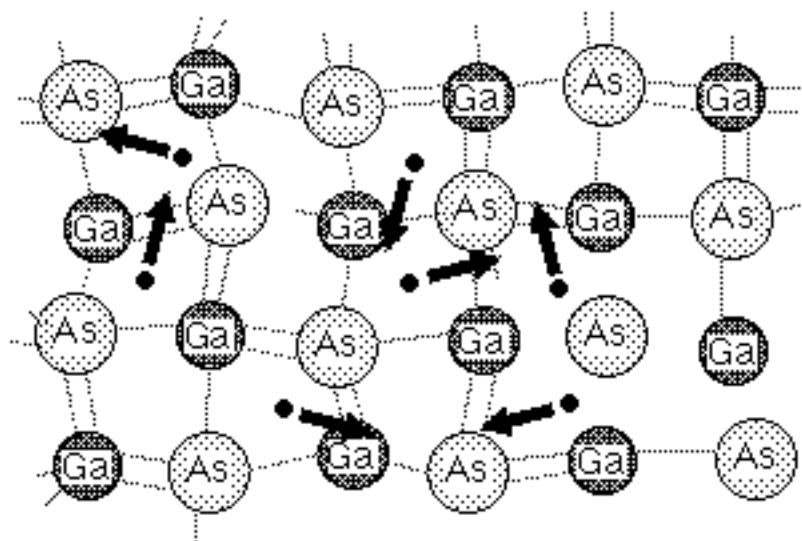
INTRODUCTION



...and create free electrons...



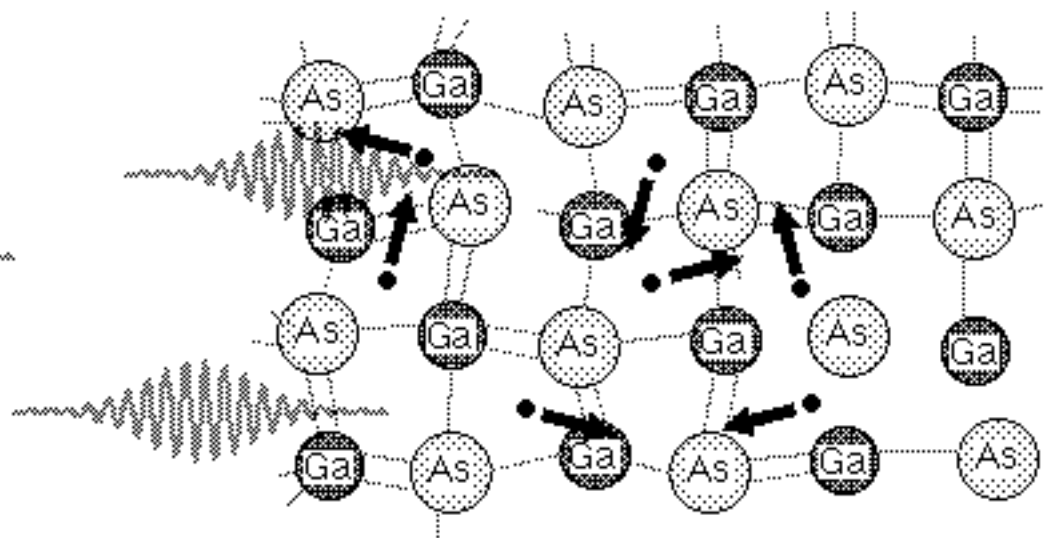
INTRODUCTION



... causing electronic & structural changes...



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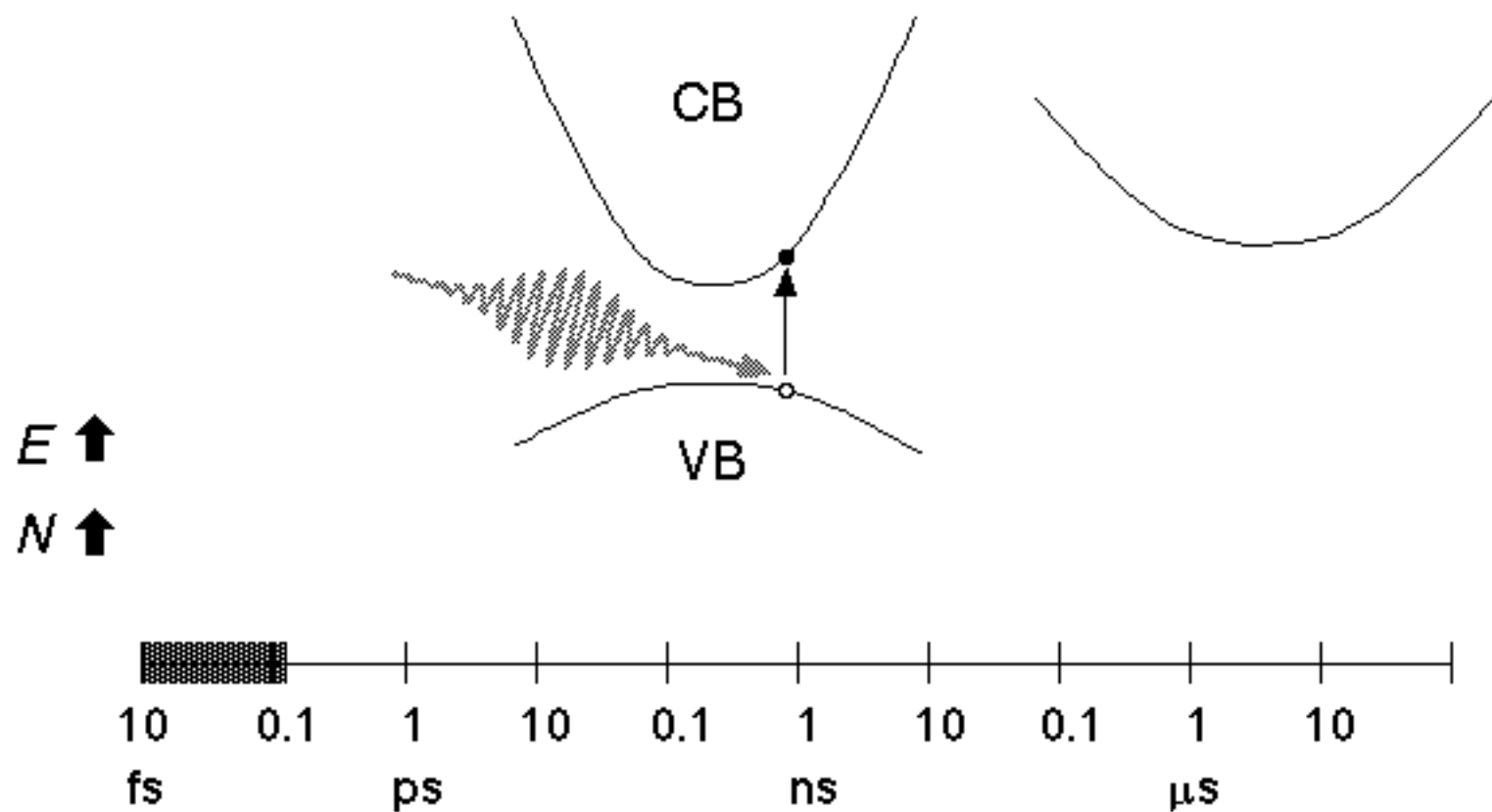


... which we measure with another pulse...



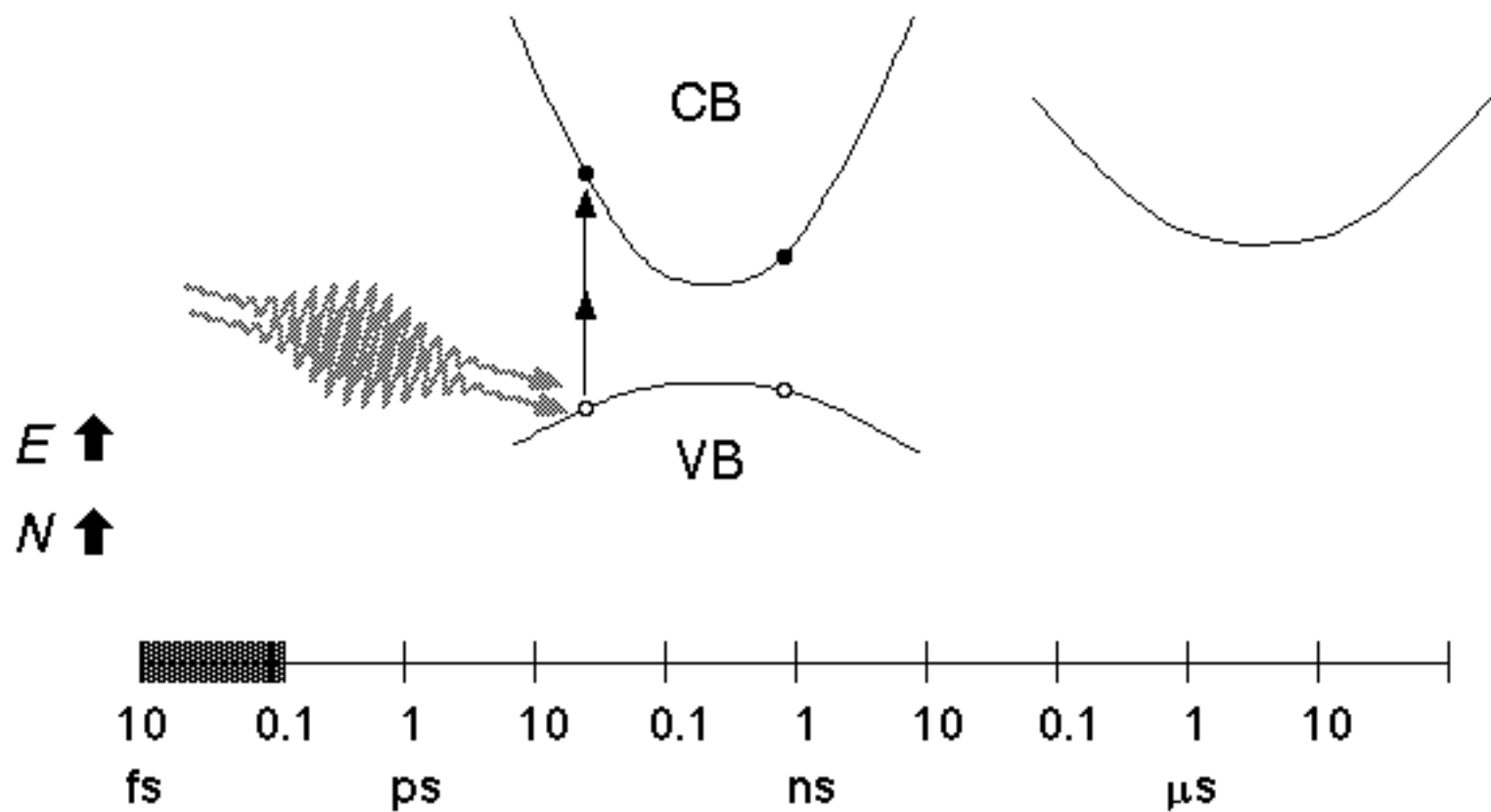
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single photon excitation



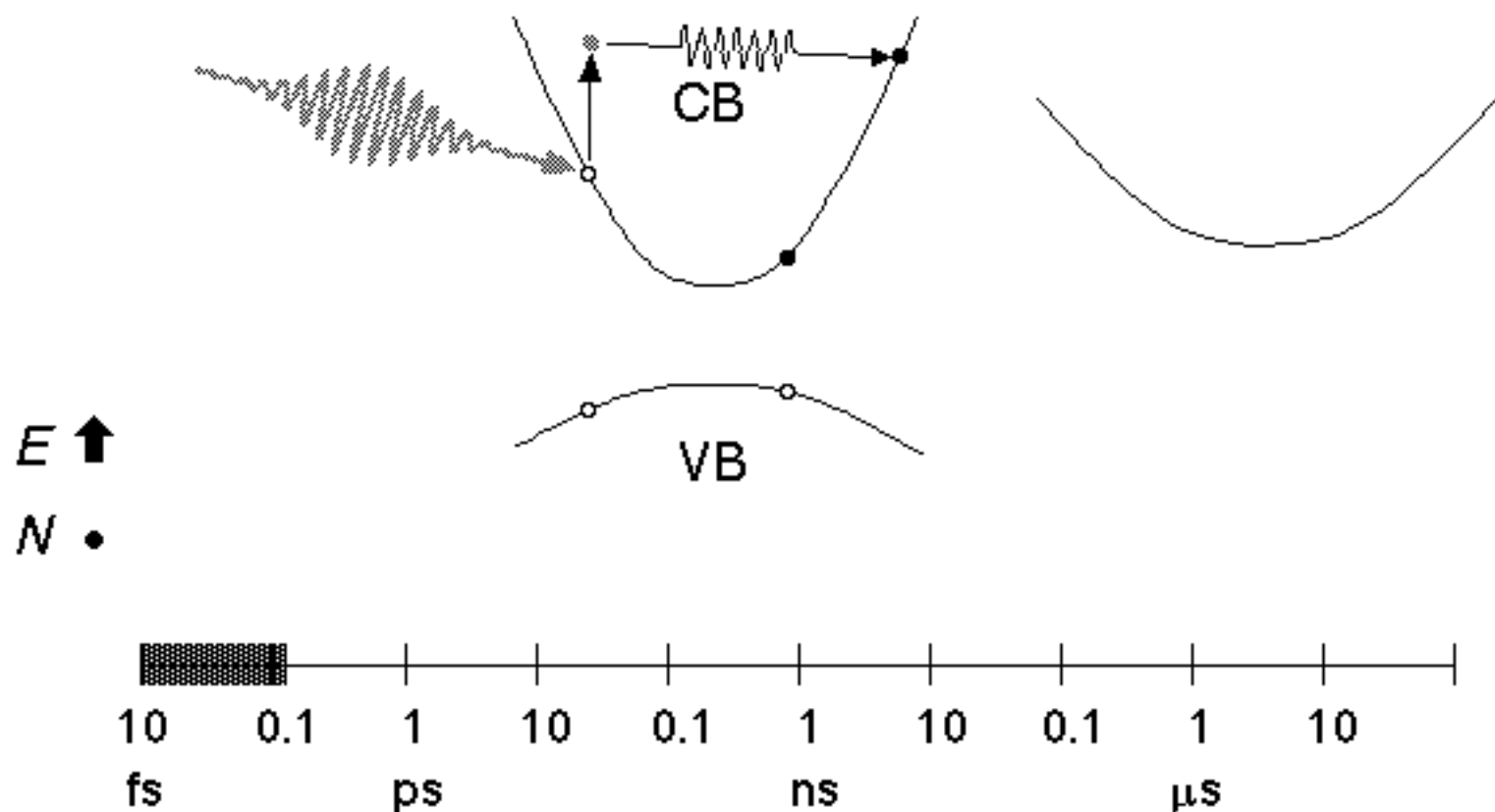
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multi photon excitation



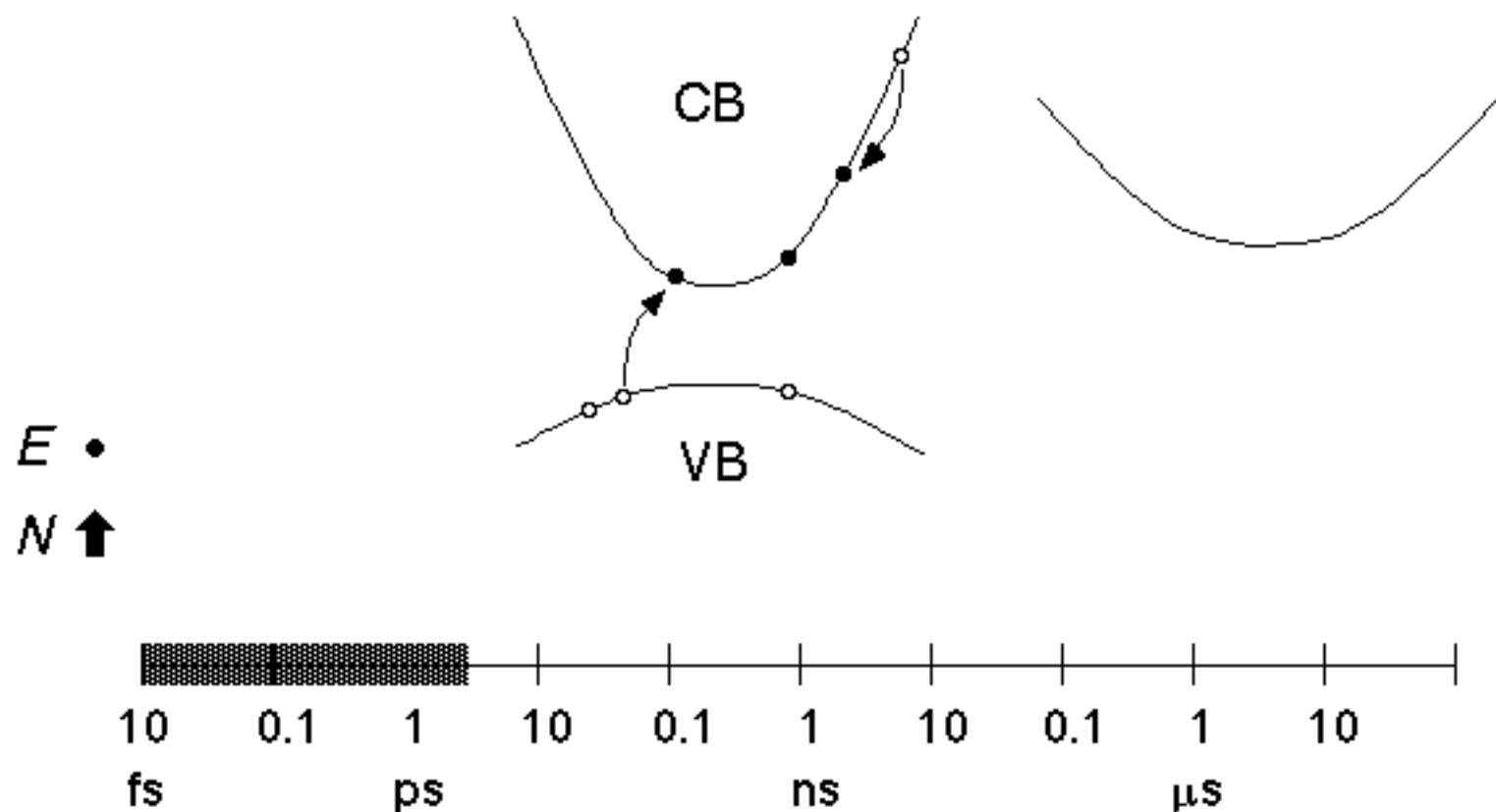
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free carrier absorption



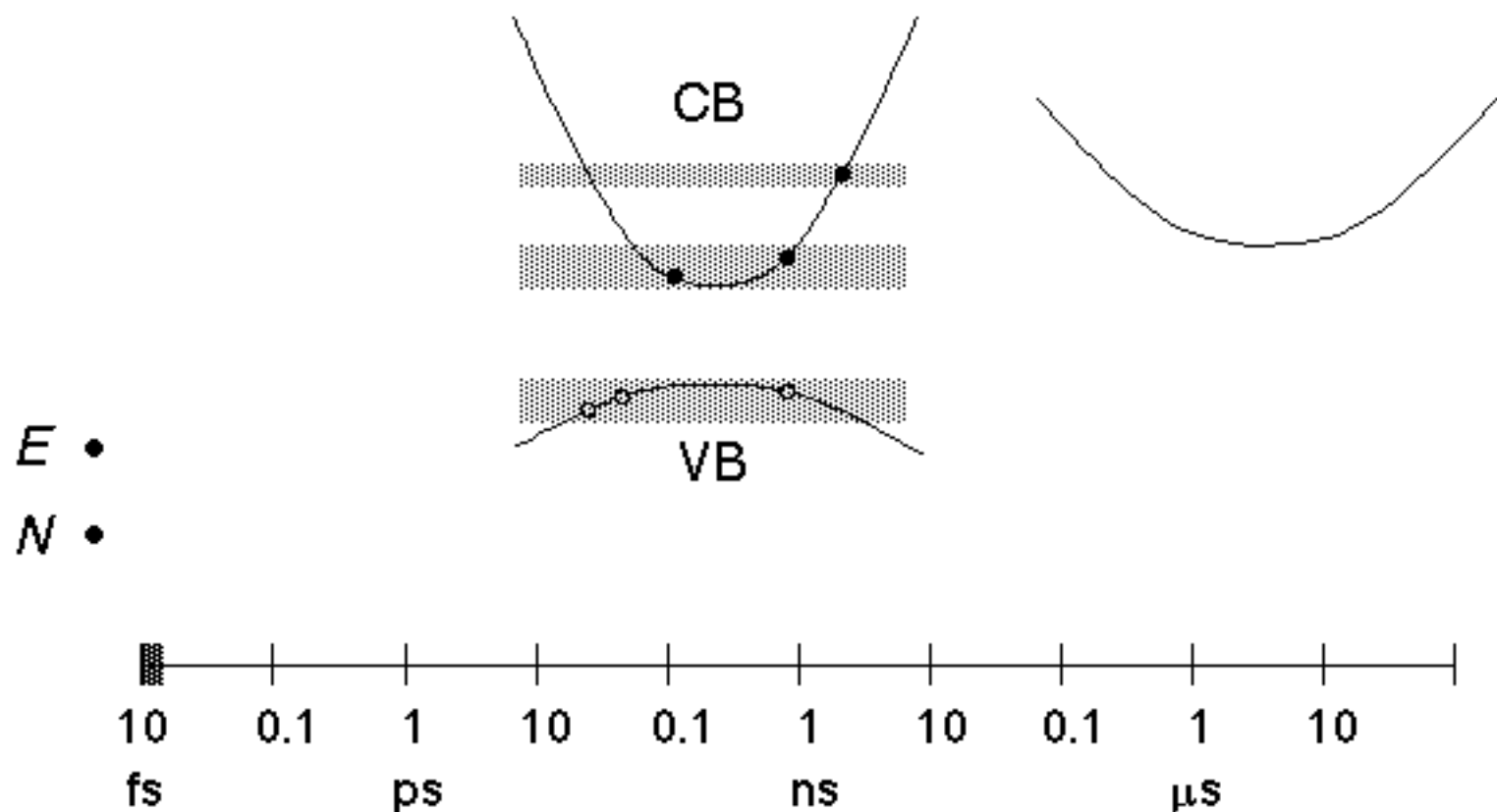
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impact ionization



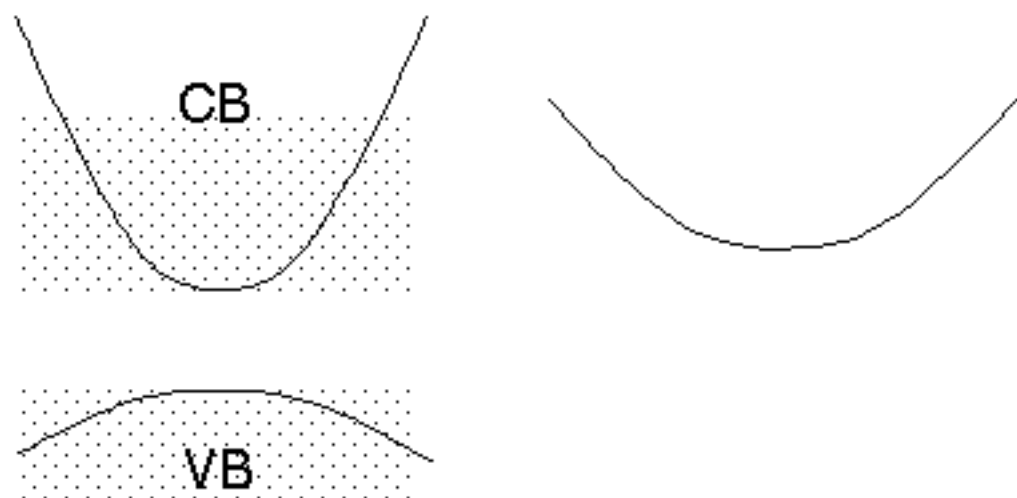
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carrier-carrier scattering



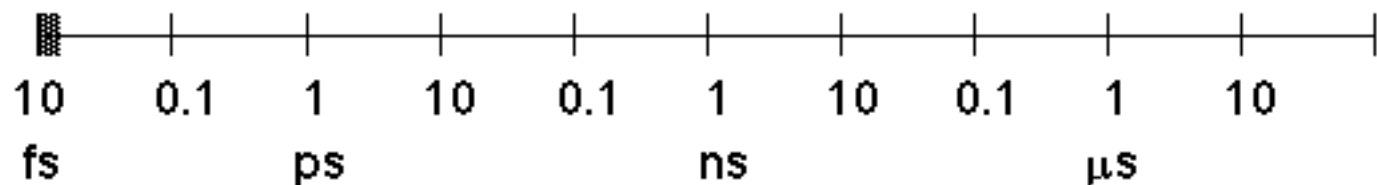
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carrier-carrier scattering



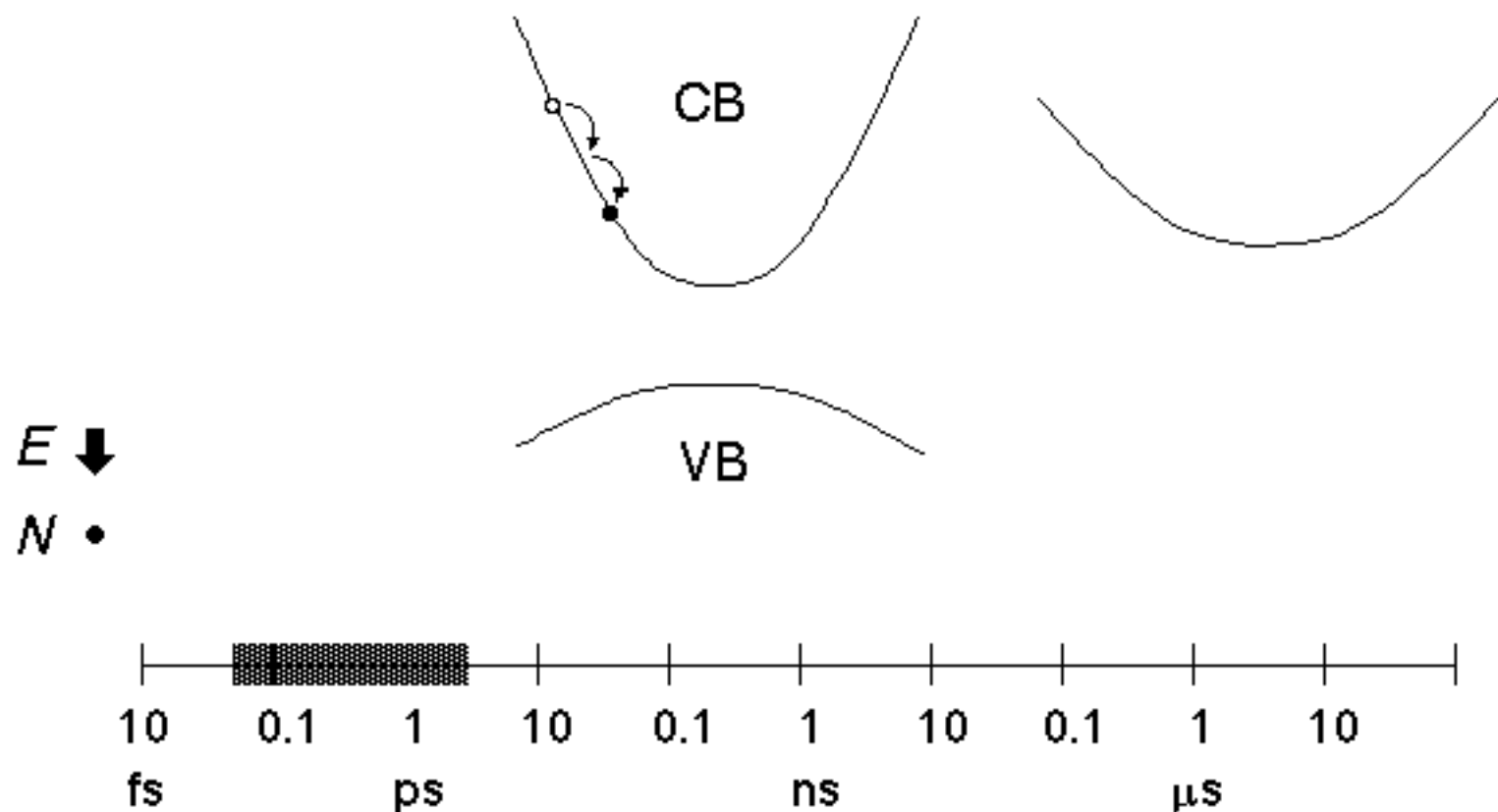
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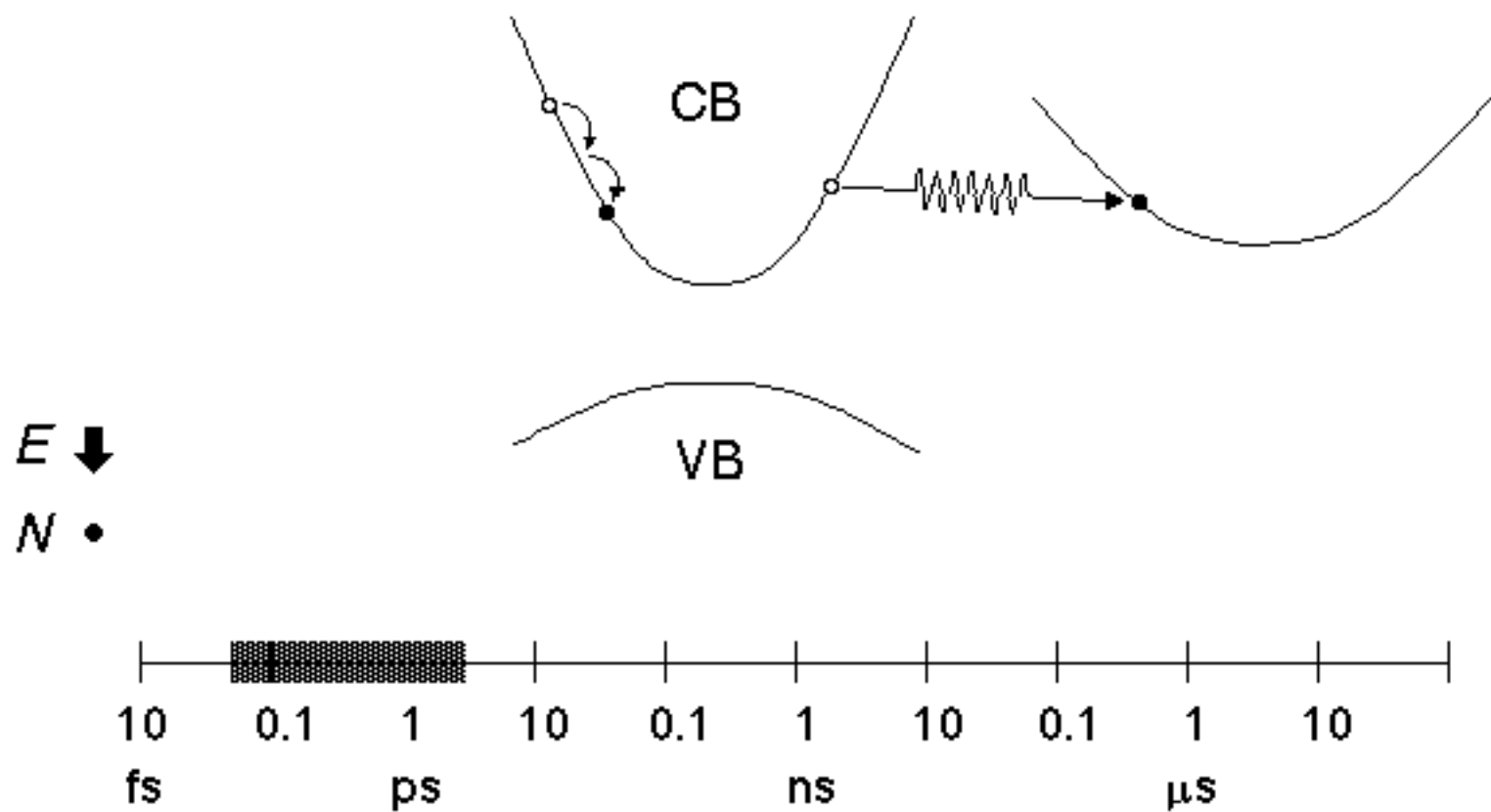
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carrier-phonon scattering



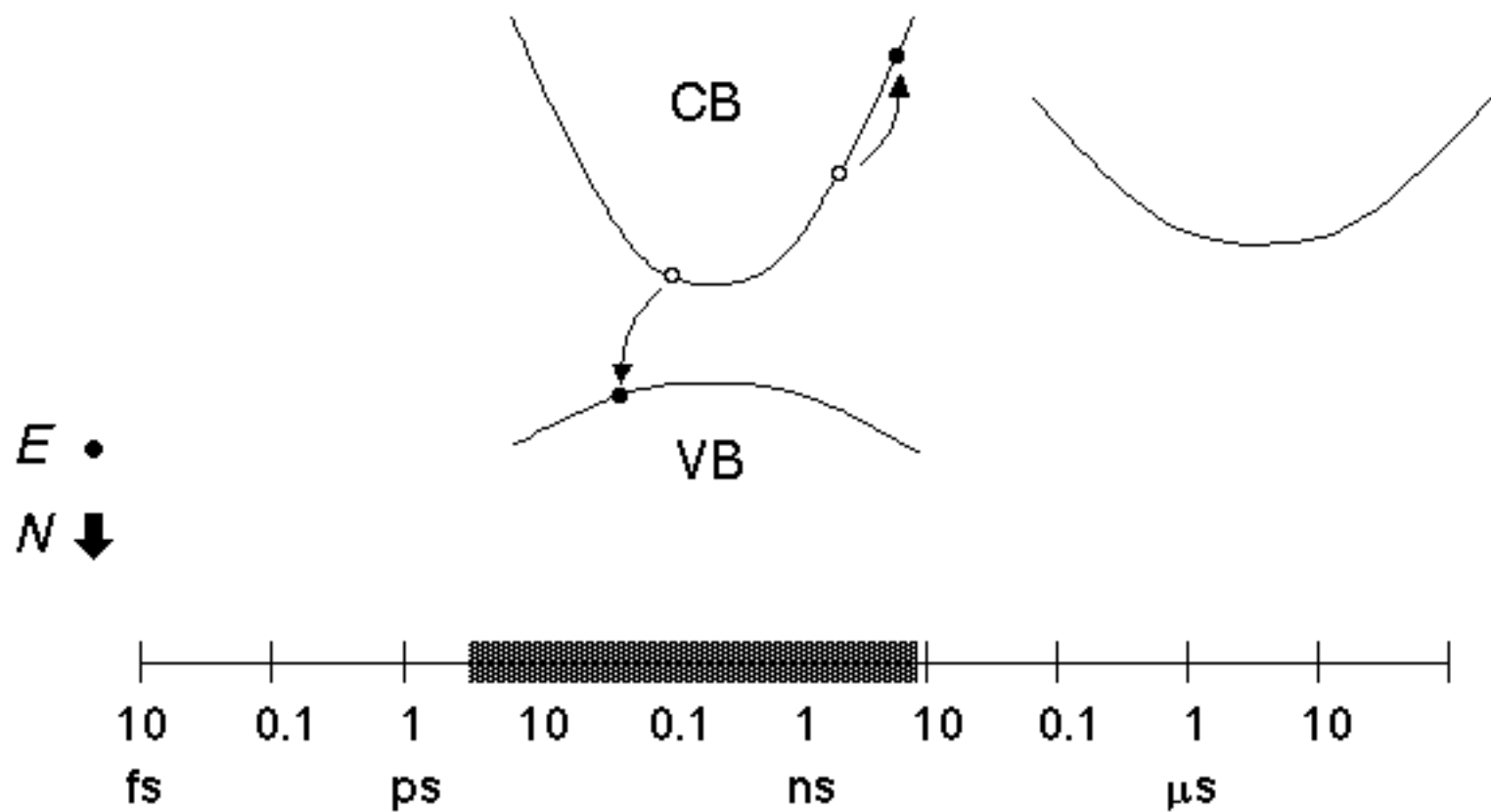
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carrier-phonon scattering



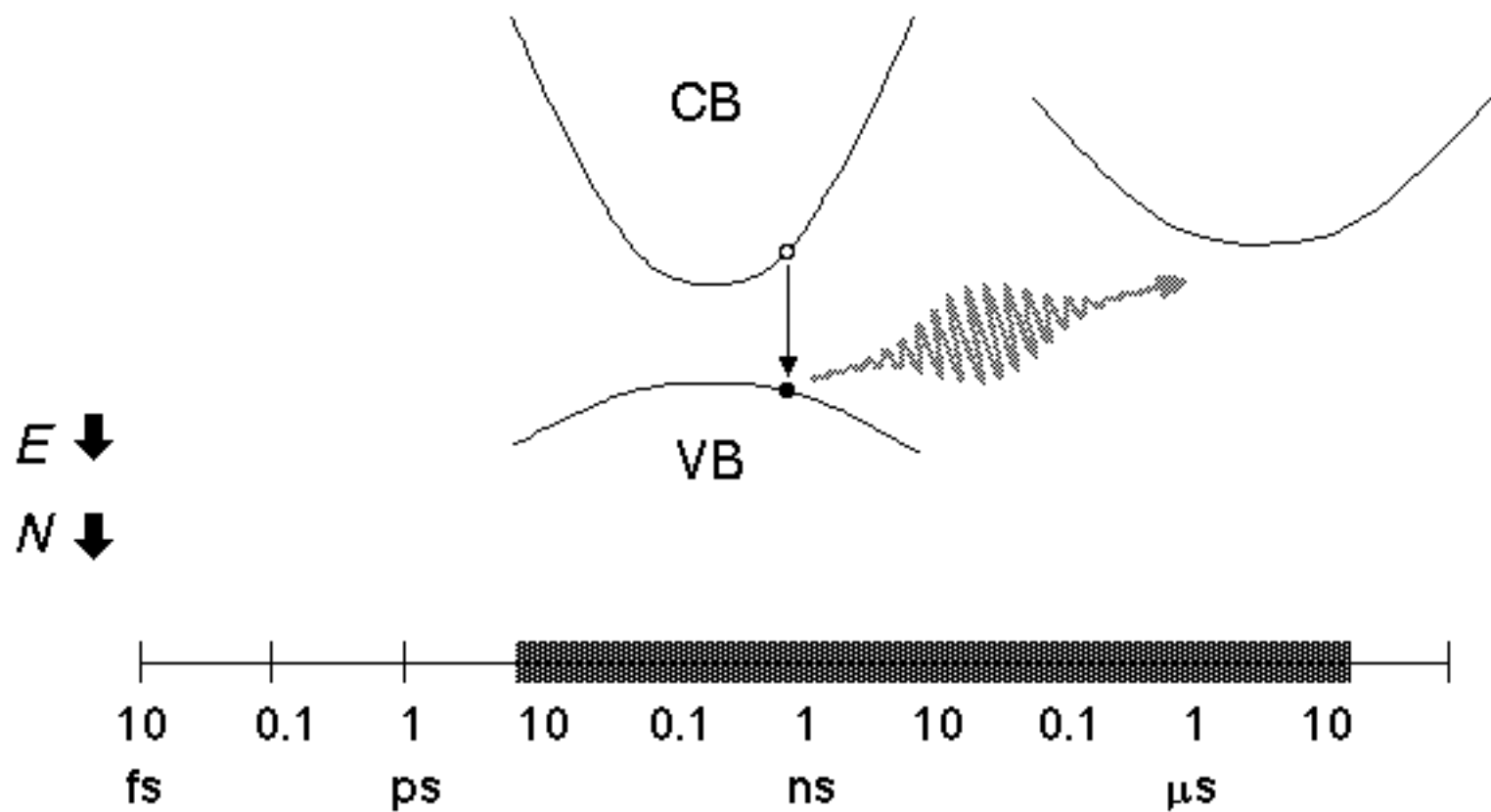
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Auger recombination



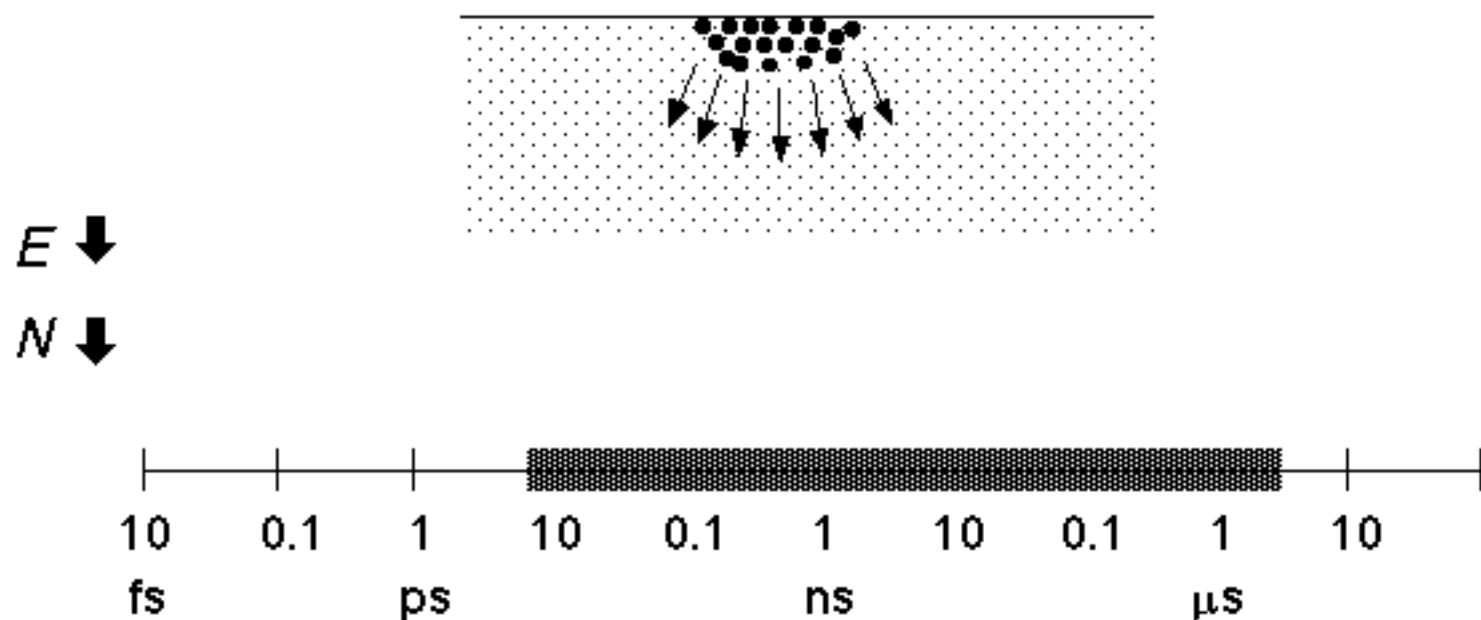
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radiative recombination



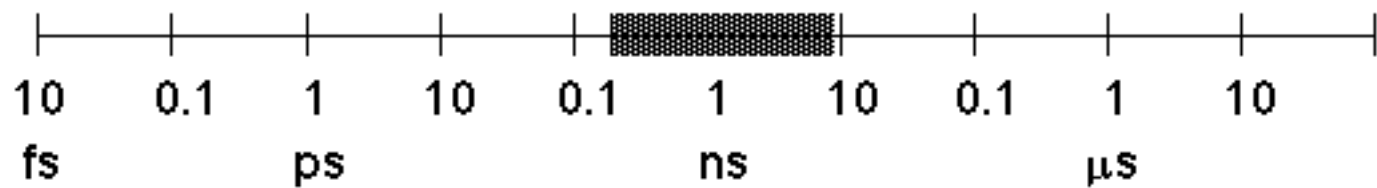
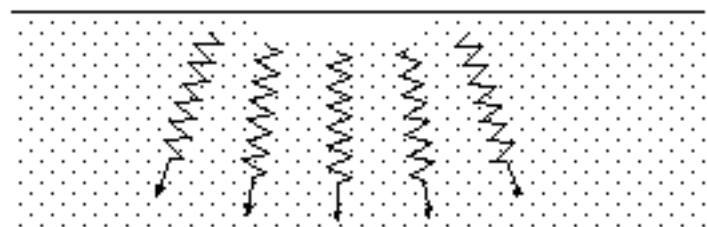
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carrier diffusion



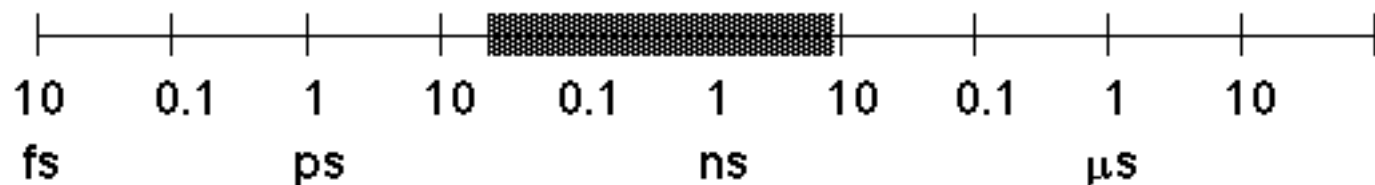
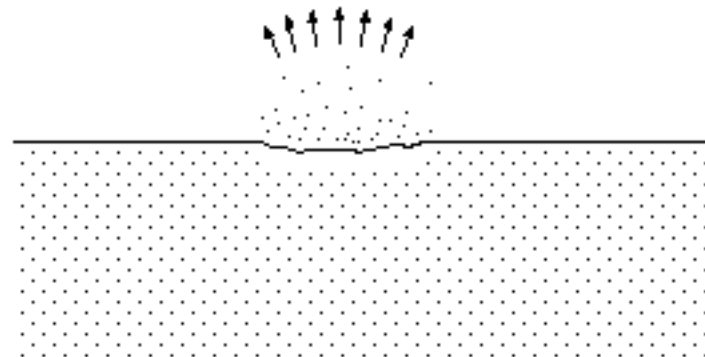
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thermal diffusion



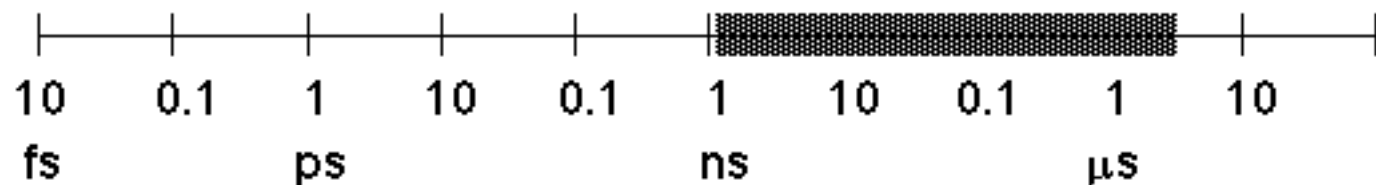
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ablation & evaporation

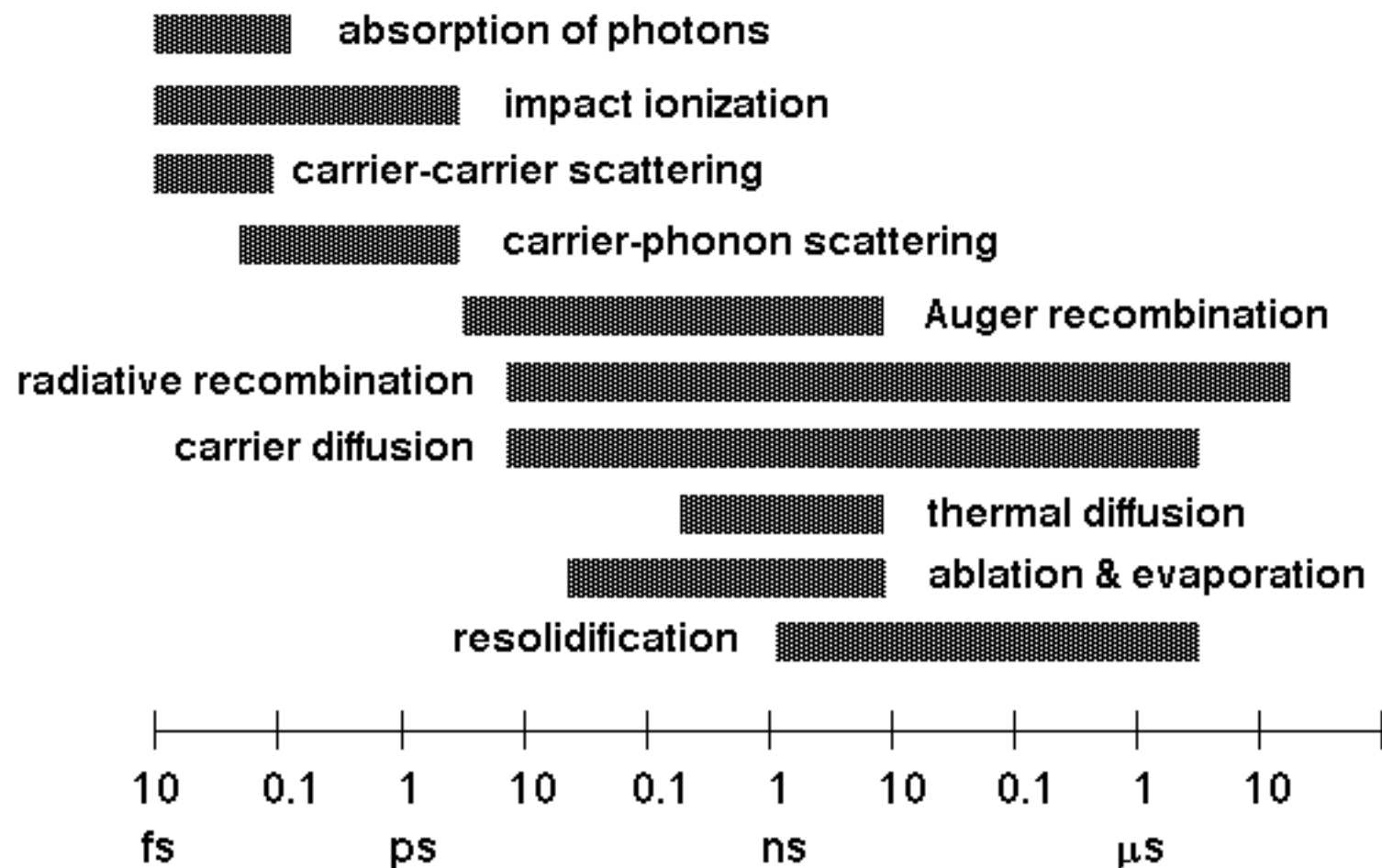


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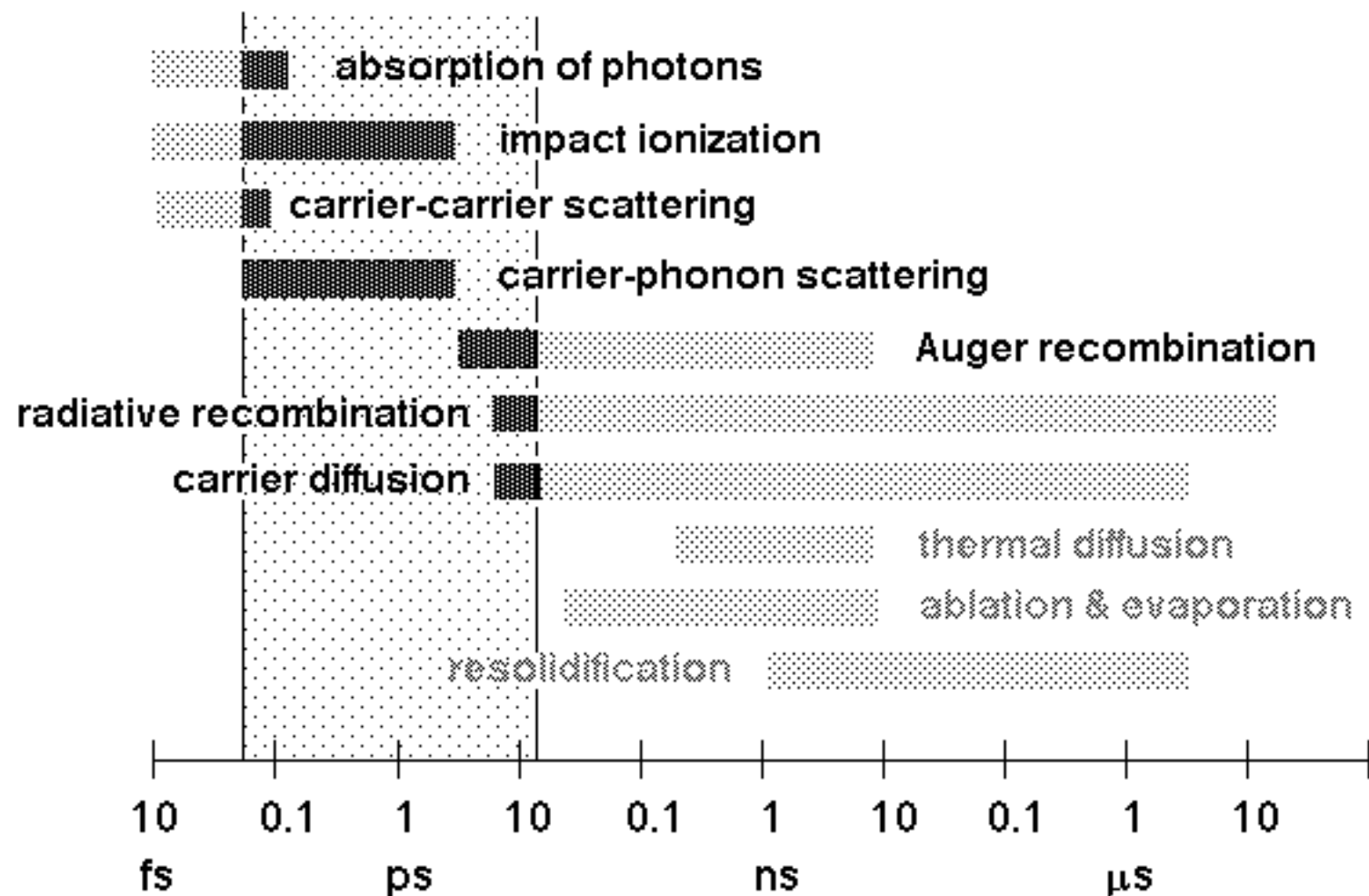
resolidification



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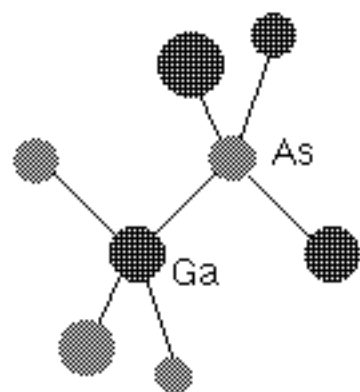


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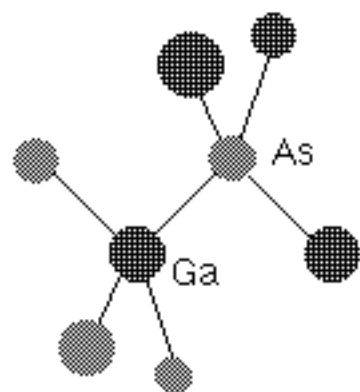
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structure



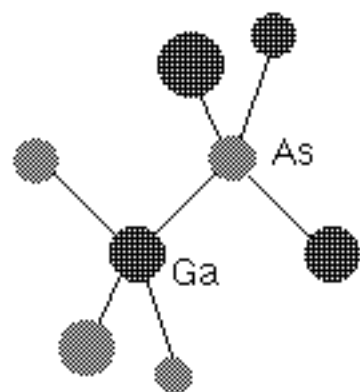
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structure

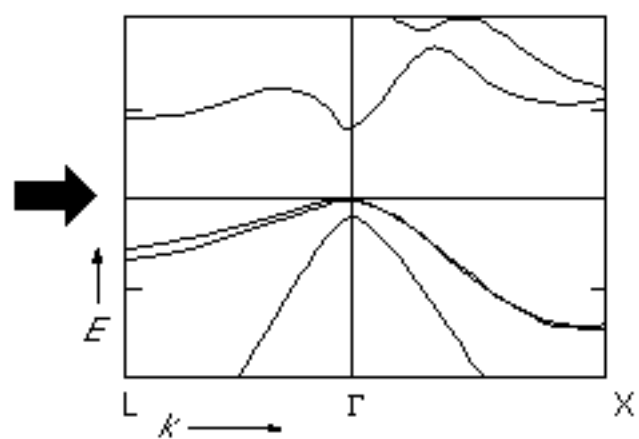


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structure

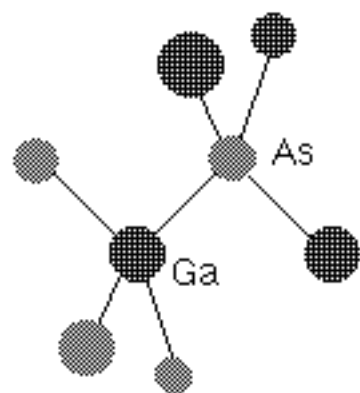


bandstructure

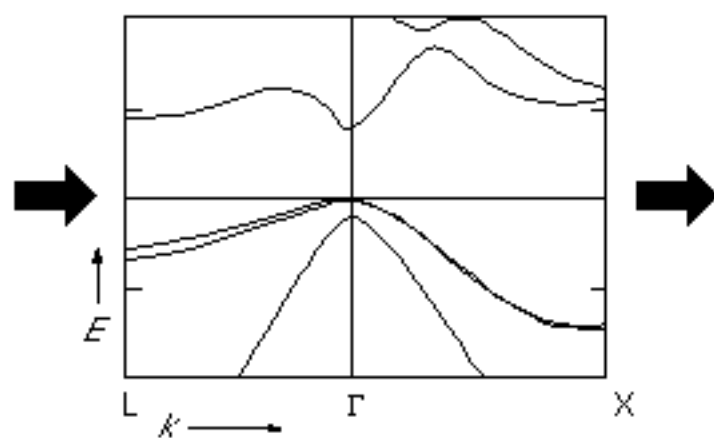


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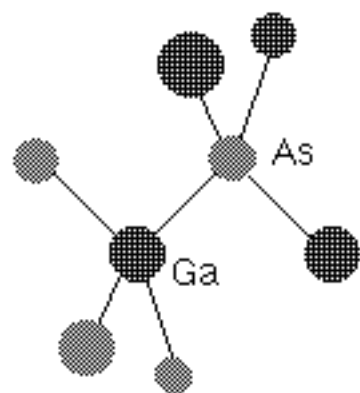


bandstructure

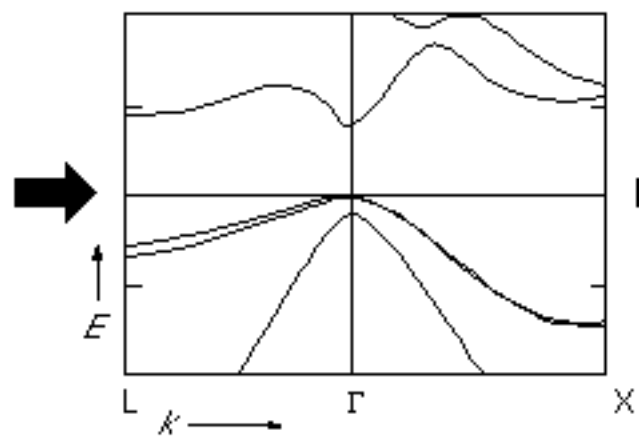


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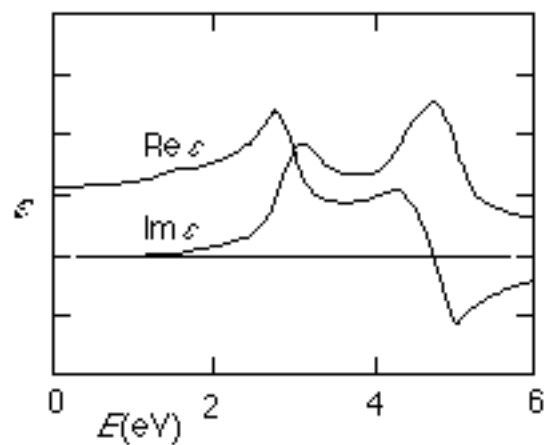
structure



bandstructure

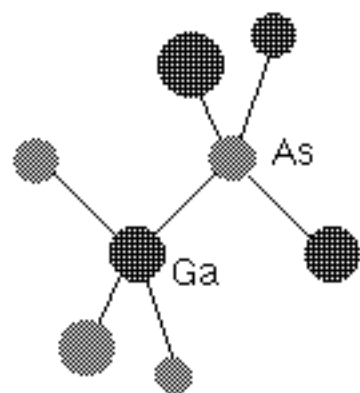


dielectric function

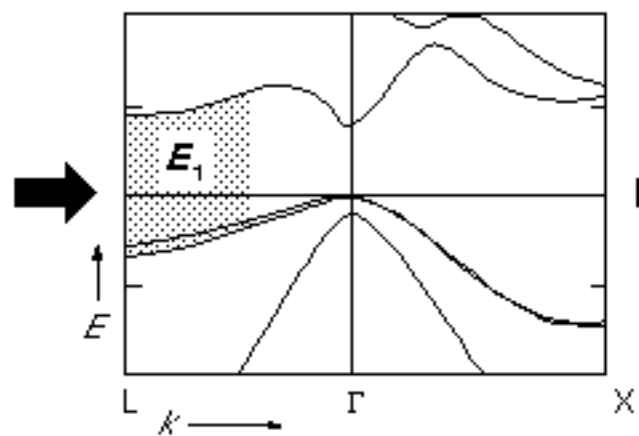


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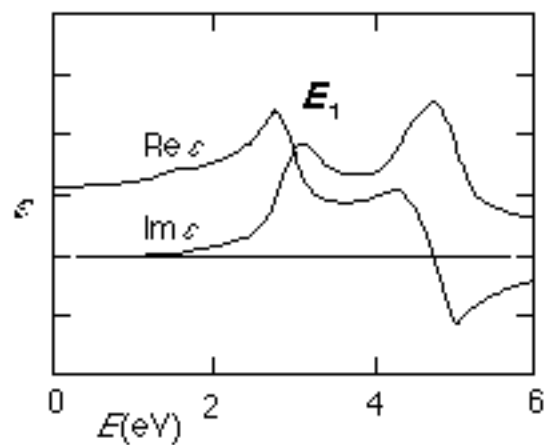
structure



bandstructure

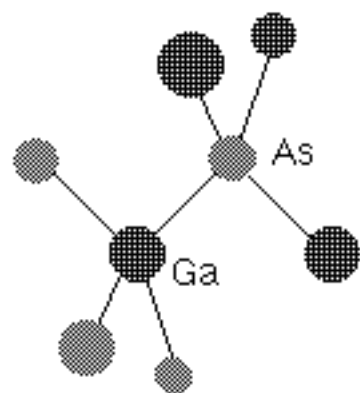


dielectric function

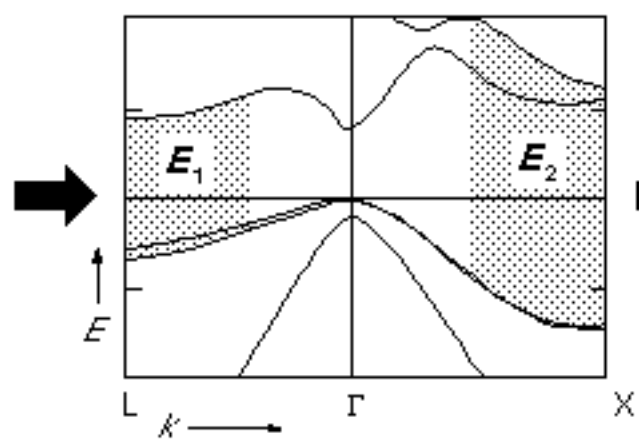


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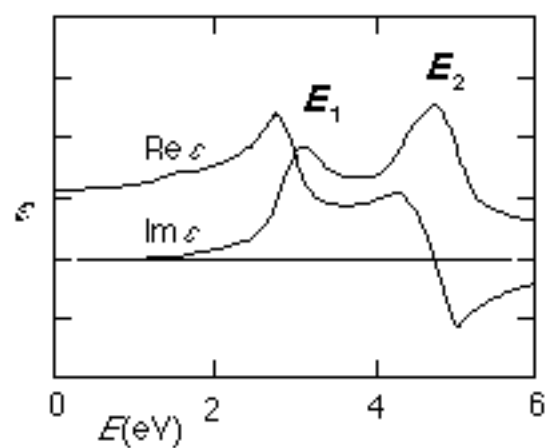
structure



bandstructure

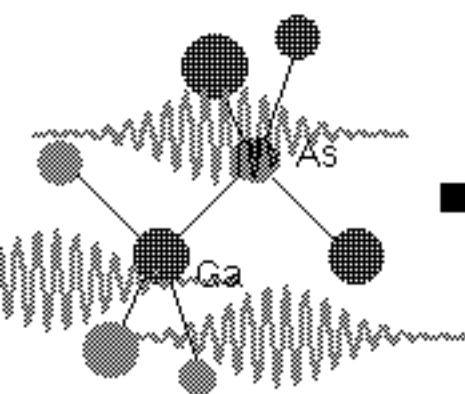


dielectric function



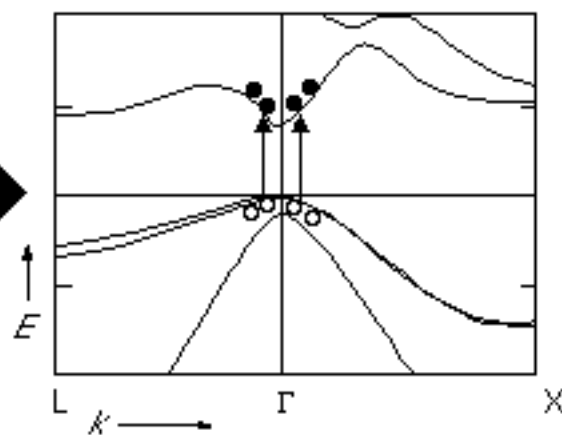
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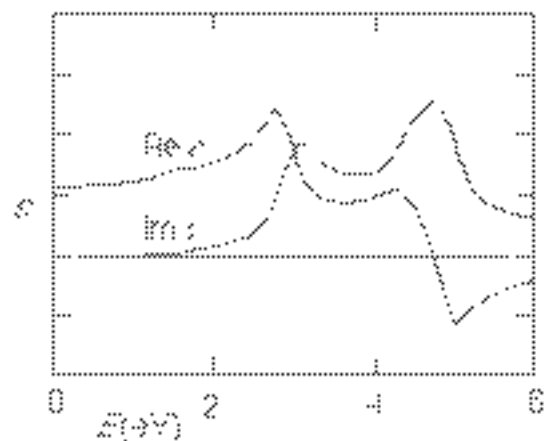
70 fs, 10^{16} W/m²

bandstructure



$10^{20} - 10^{22}$ cm⁻³

dielectric function



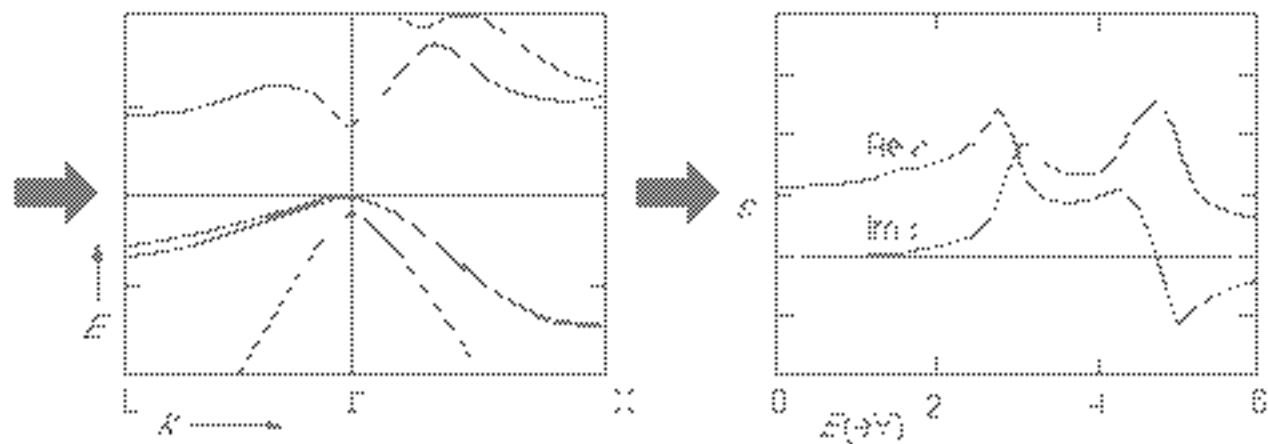
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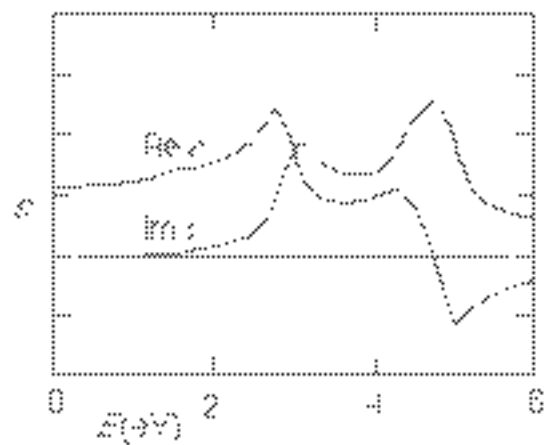
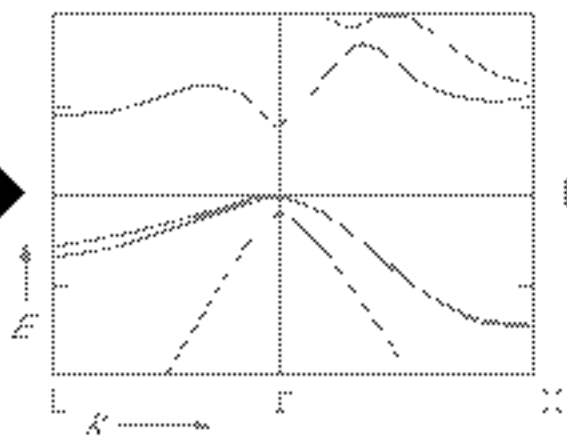
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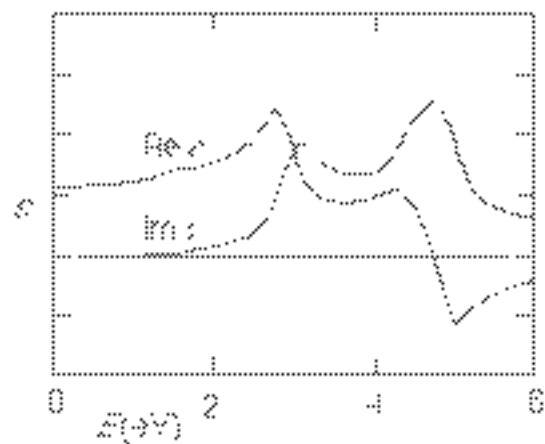
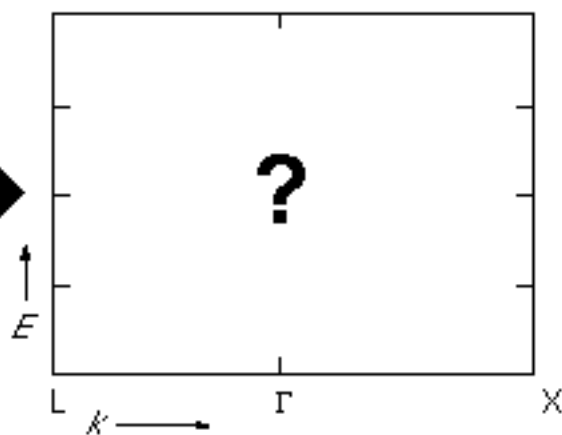


bandstructure

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dielectric function



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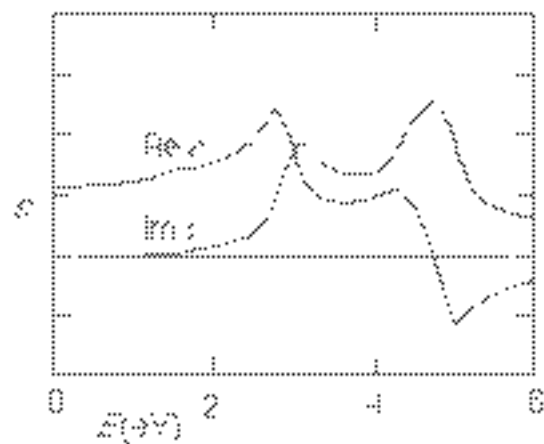
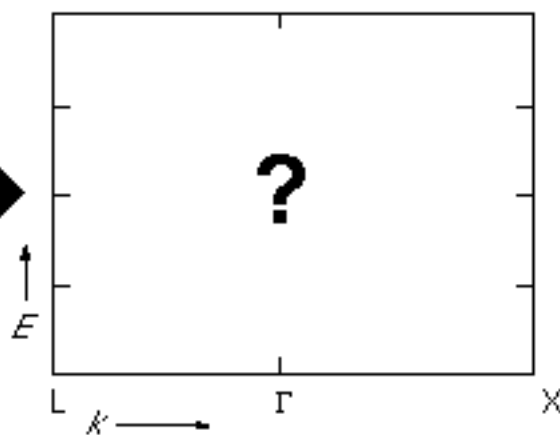


bandstructure

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dielectric function



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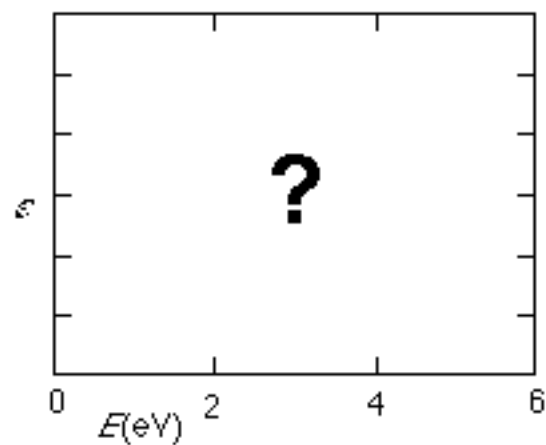
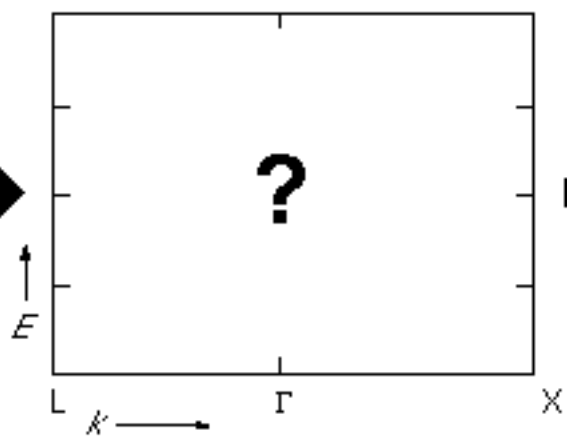
bandstructure

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dielectric function

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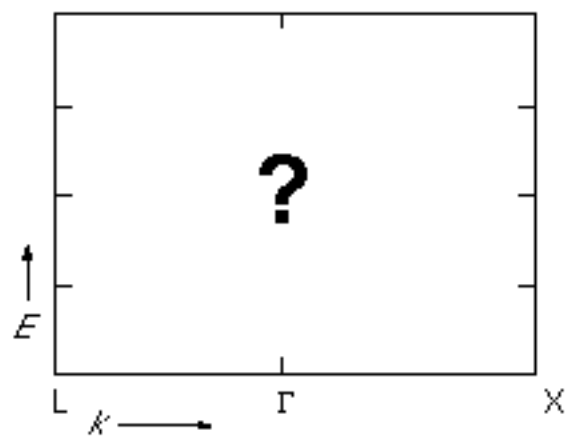


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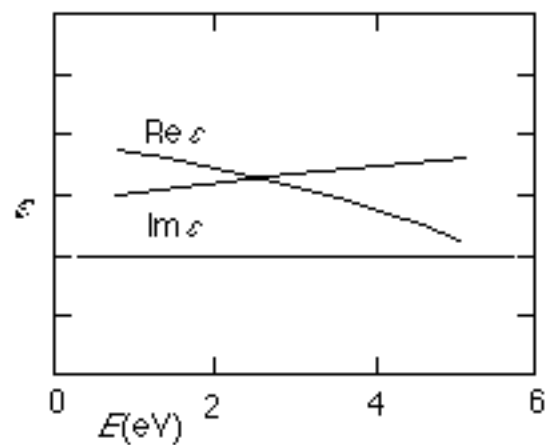
structure

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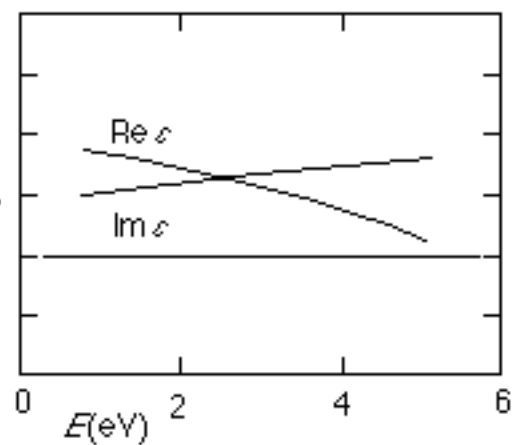
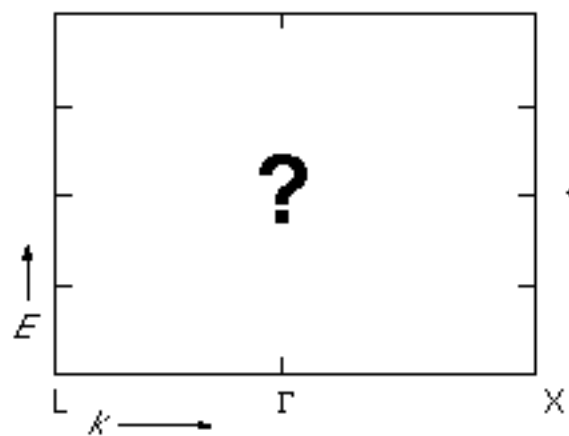
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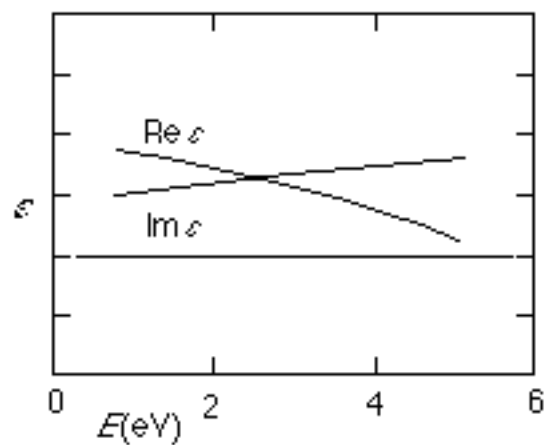
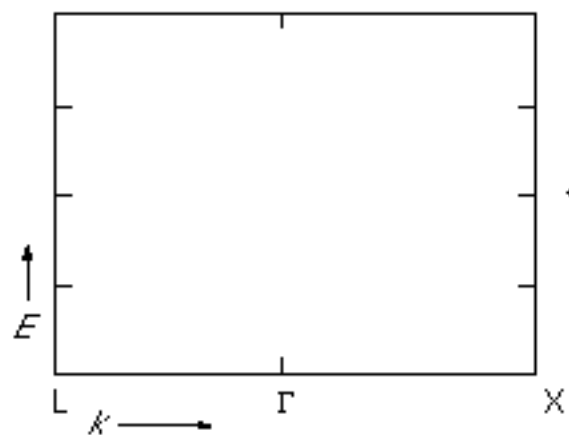
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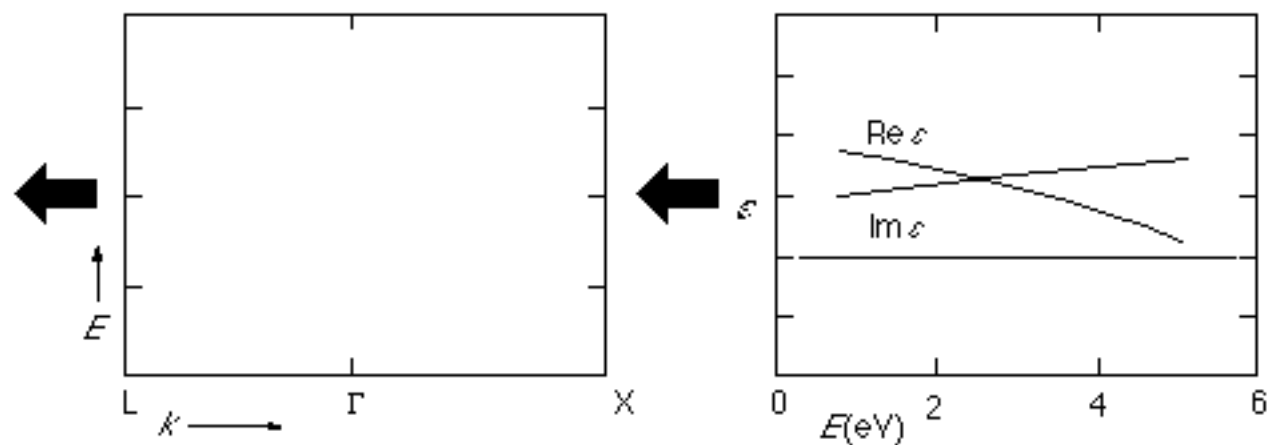
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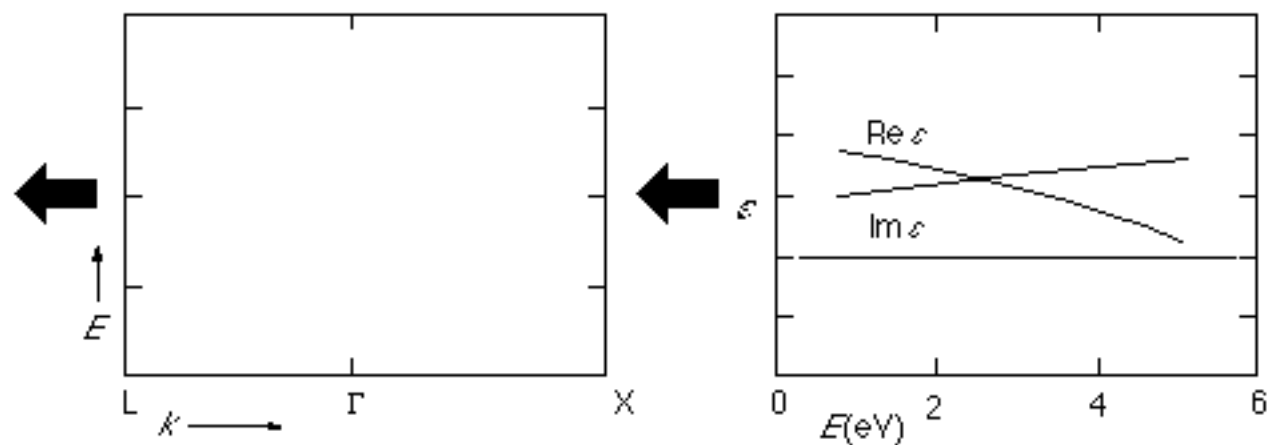


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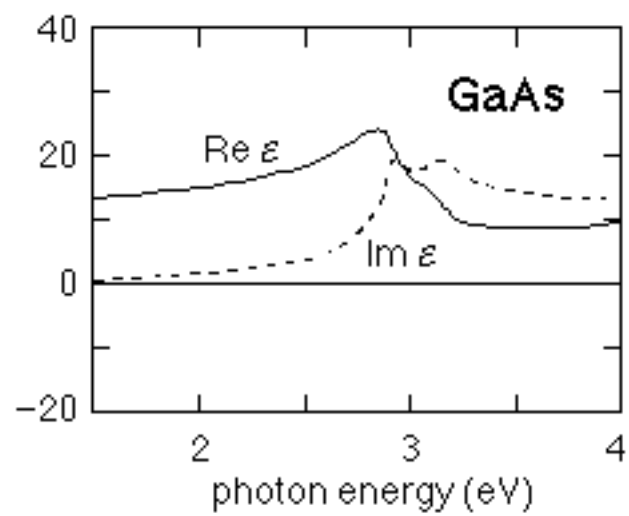
structure

bandstructure

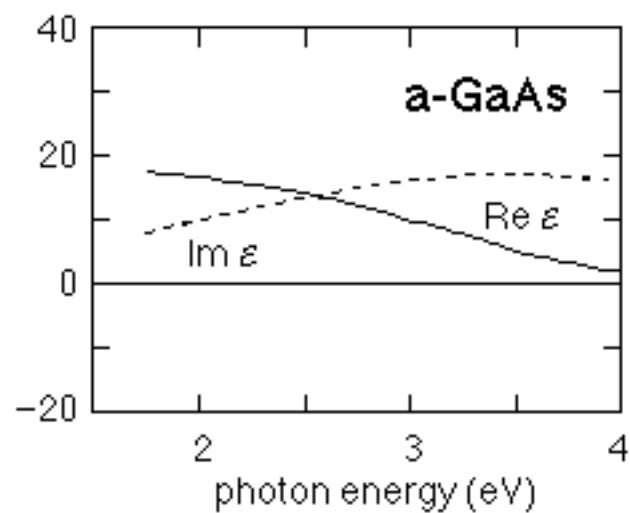
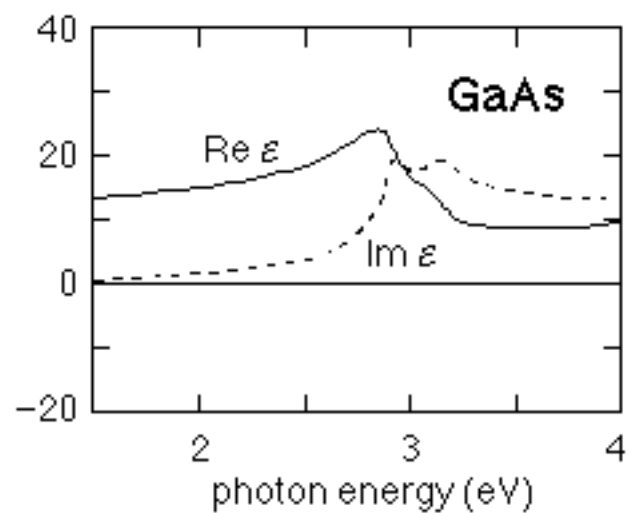
dielectric function



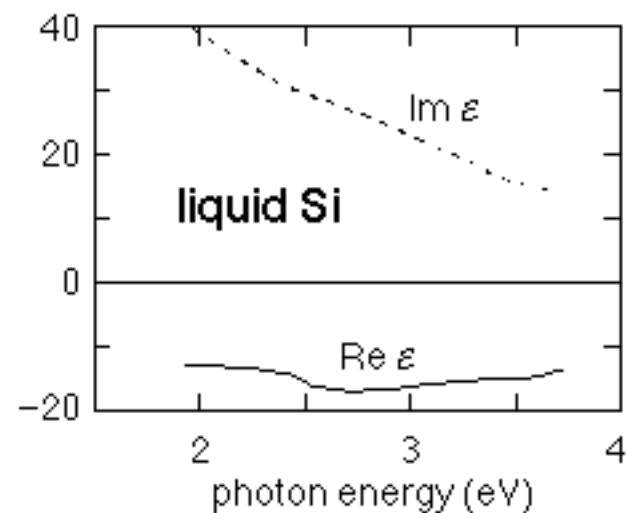
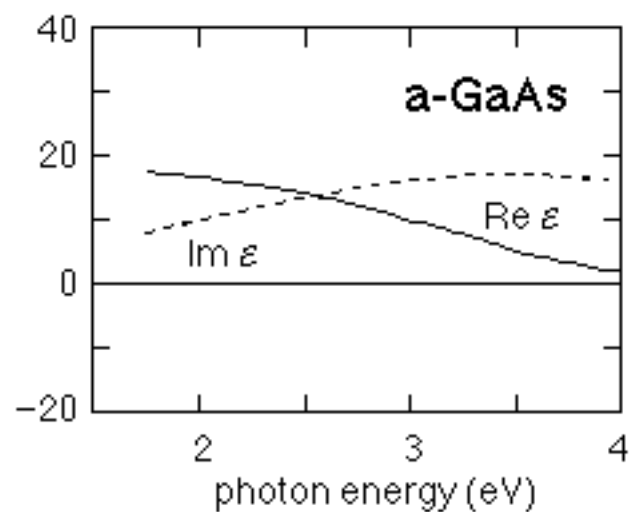
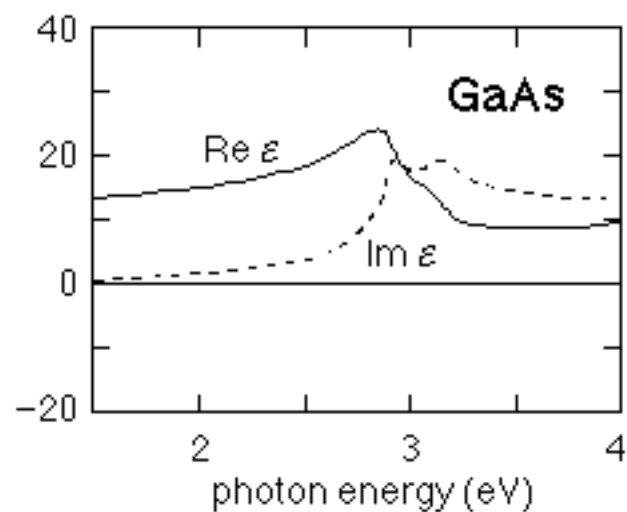
DIELECTRIC FUNCTION



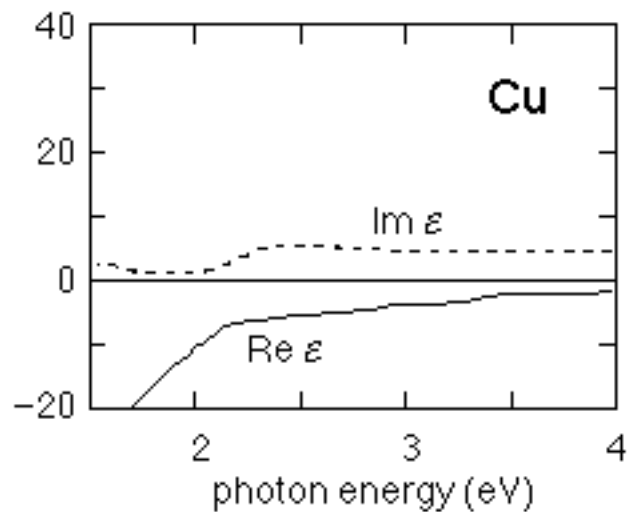
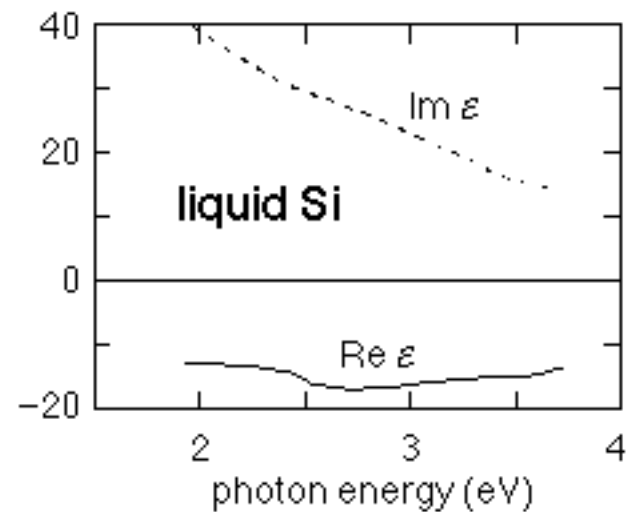
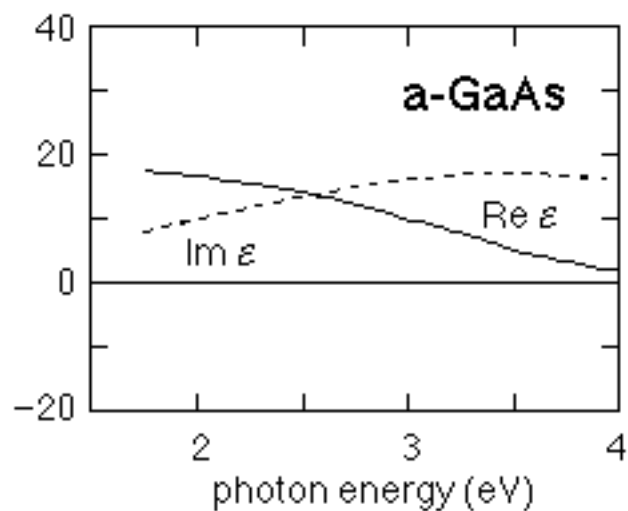
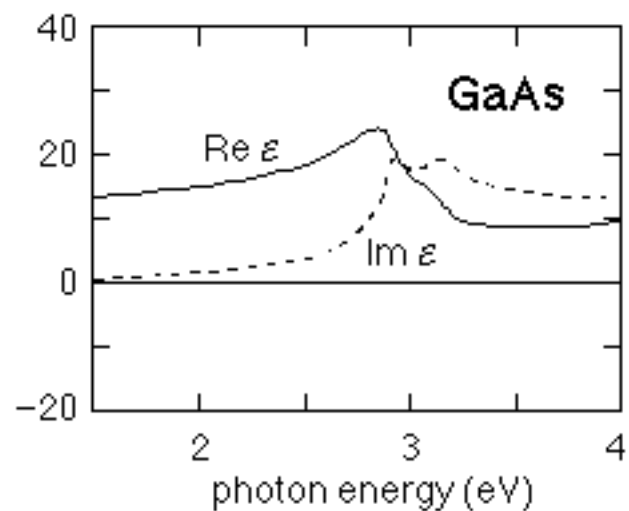
DIELECTRIC FUNCTION



DIELECTRIC FUNCTION



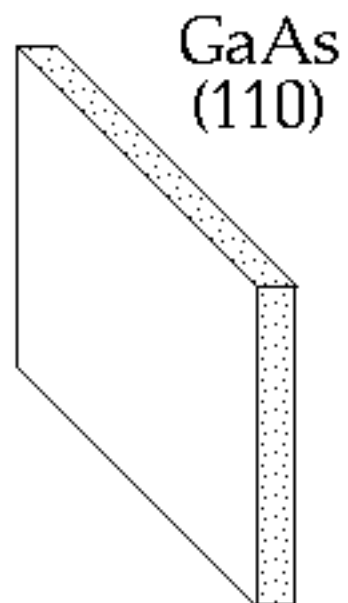
DIELECTRIC FUNCTION



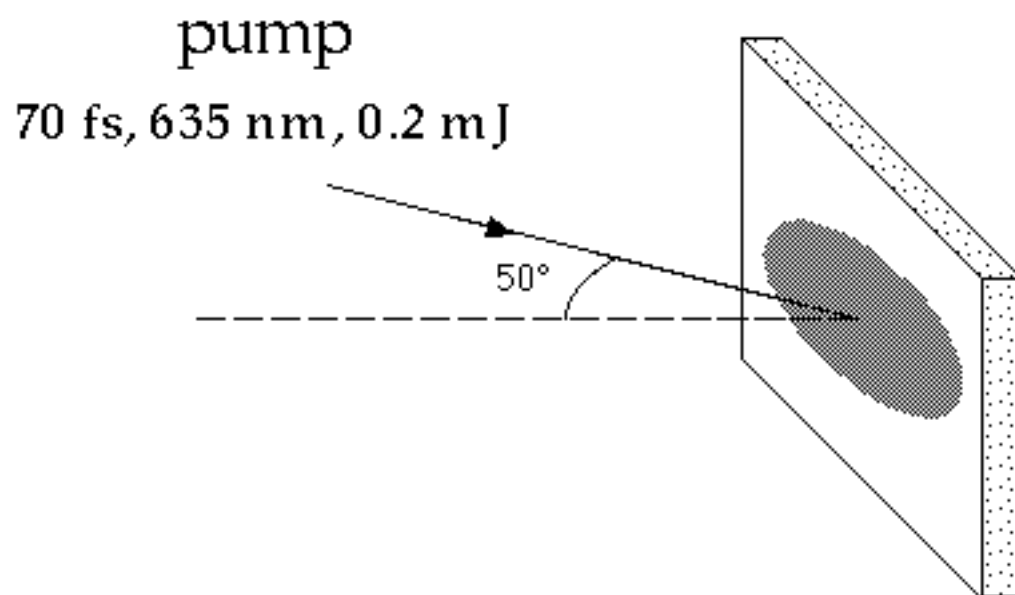
- ① Method
- ② Results
- ③ Discussion



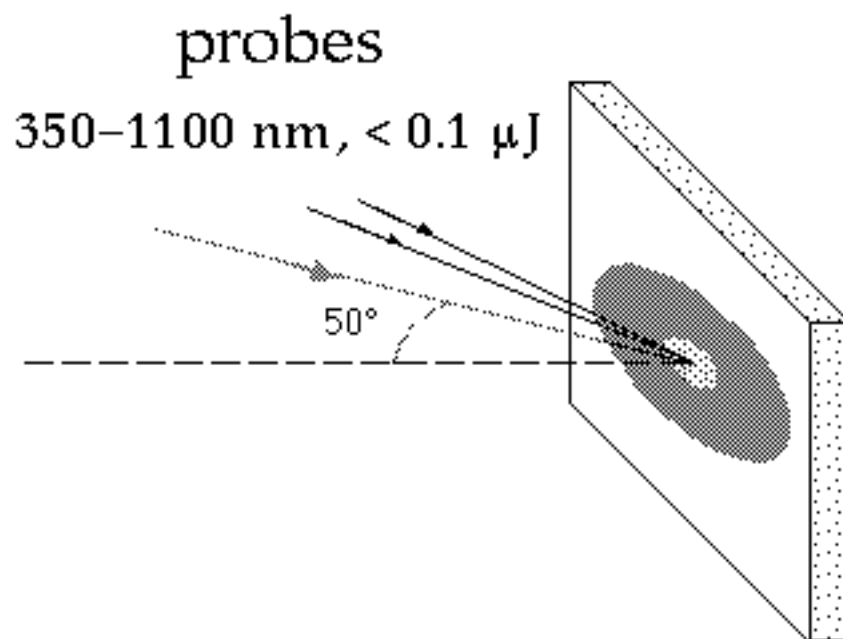
PUMP & PROBE MEASUREMENT



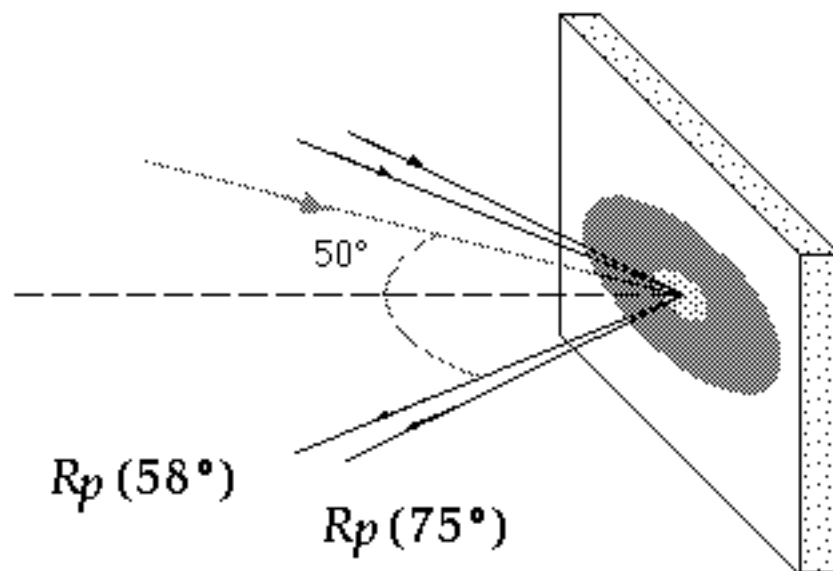
PUMP & PROBE MEASUREMENT



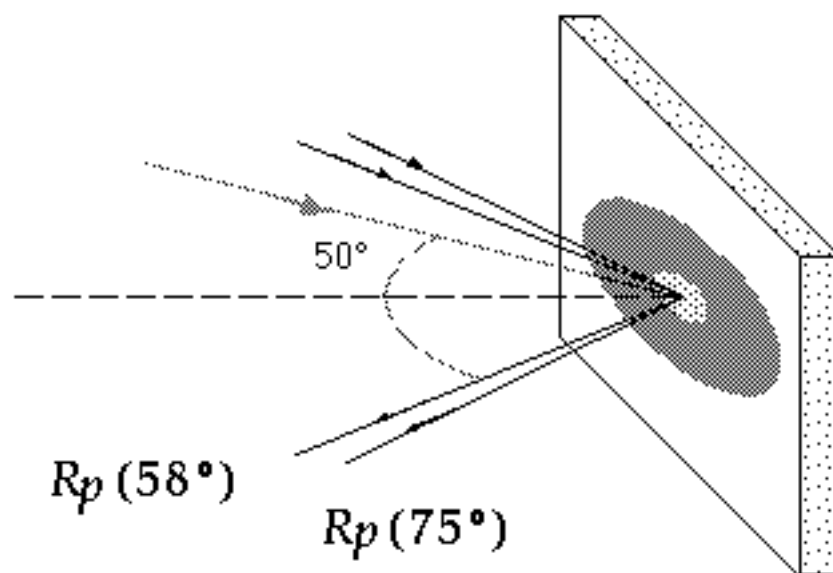
PUMP & PROBE MEASUREMENT



PUMP & PROBE MEASUREMENT



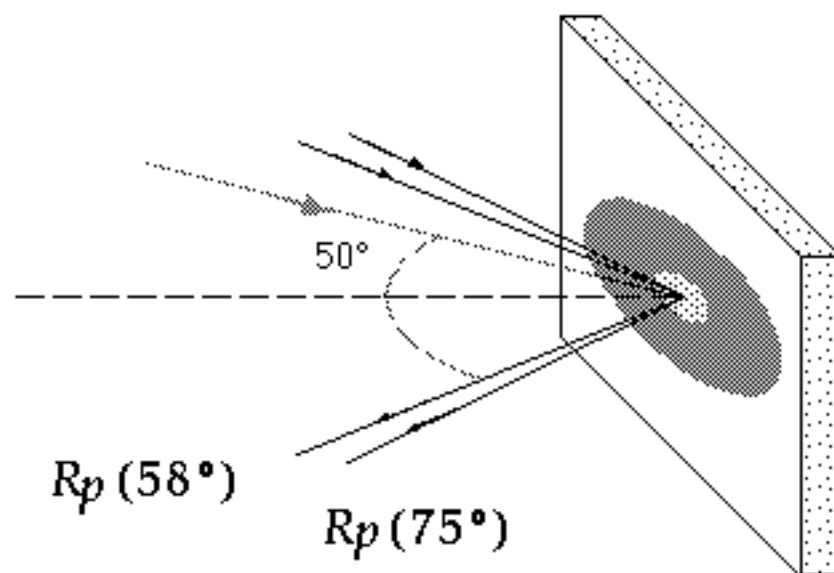
PUMP & PROBE MEASUREMENT



Fresnel
equations



PUMP & PROBE MEASUREMENT



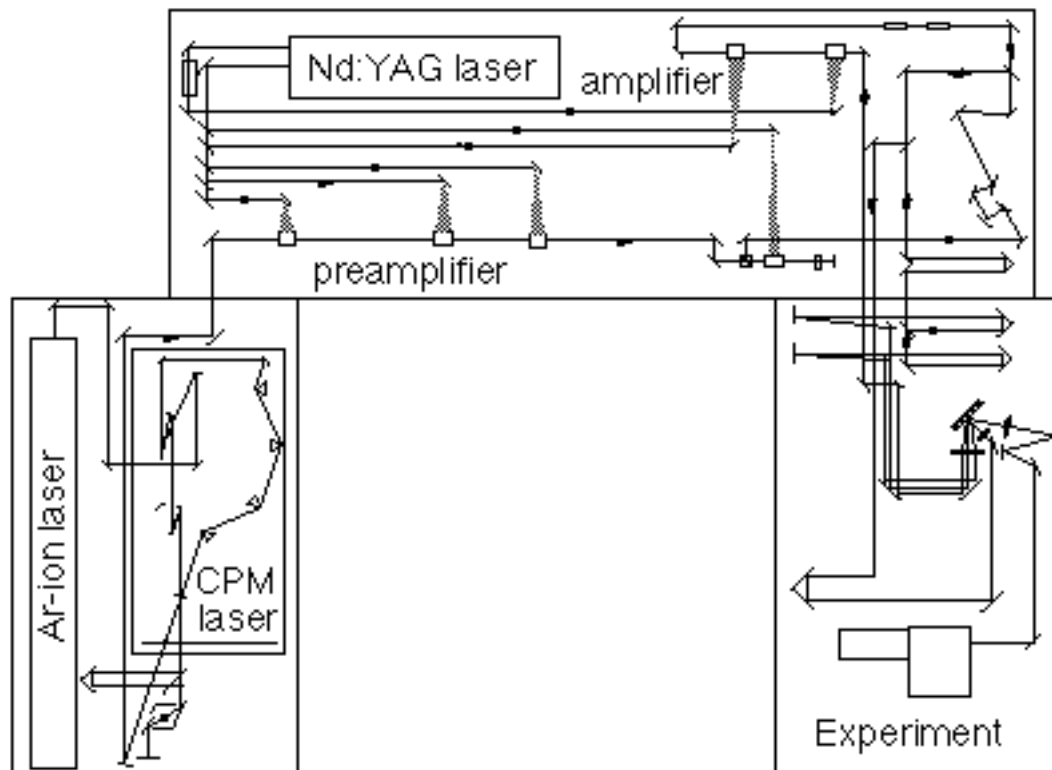
↓
Fresnel
equations



$\text{Re } \epsilon(\omega), \text{Im } \epsilon(\omega)$



SETUP

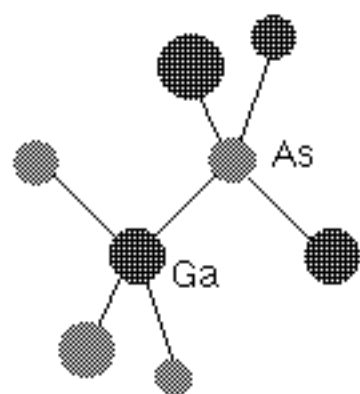


- ① Method
- ② Results
- ③ Discussion

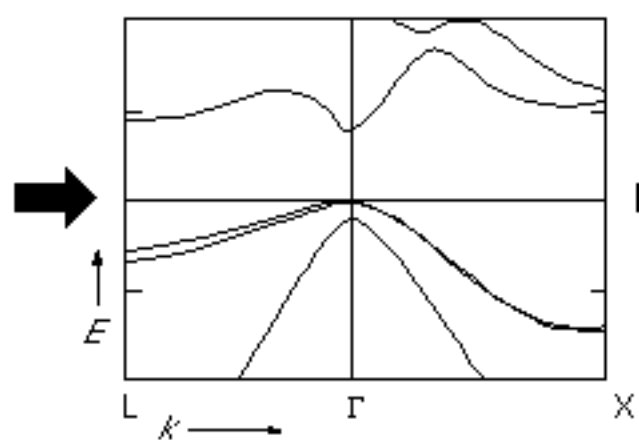


WHAT IS HAPPENING?

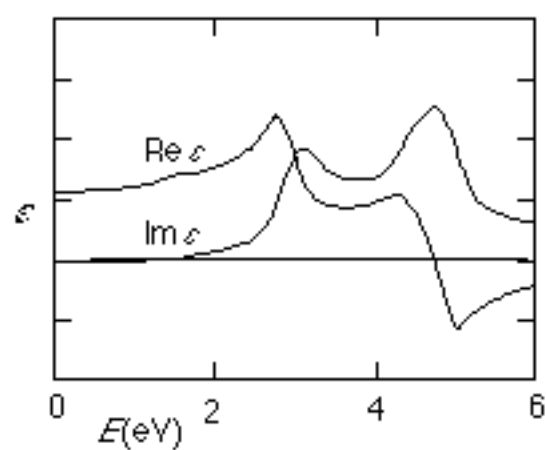
structure



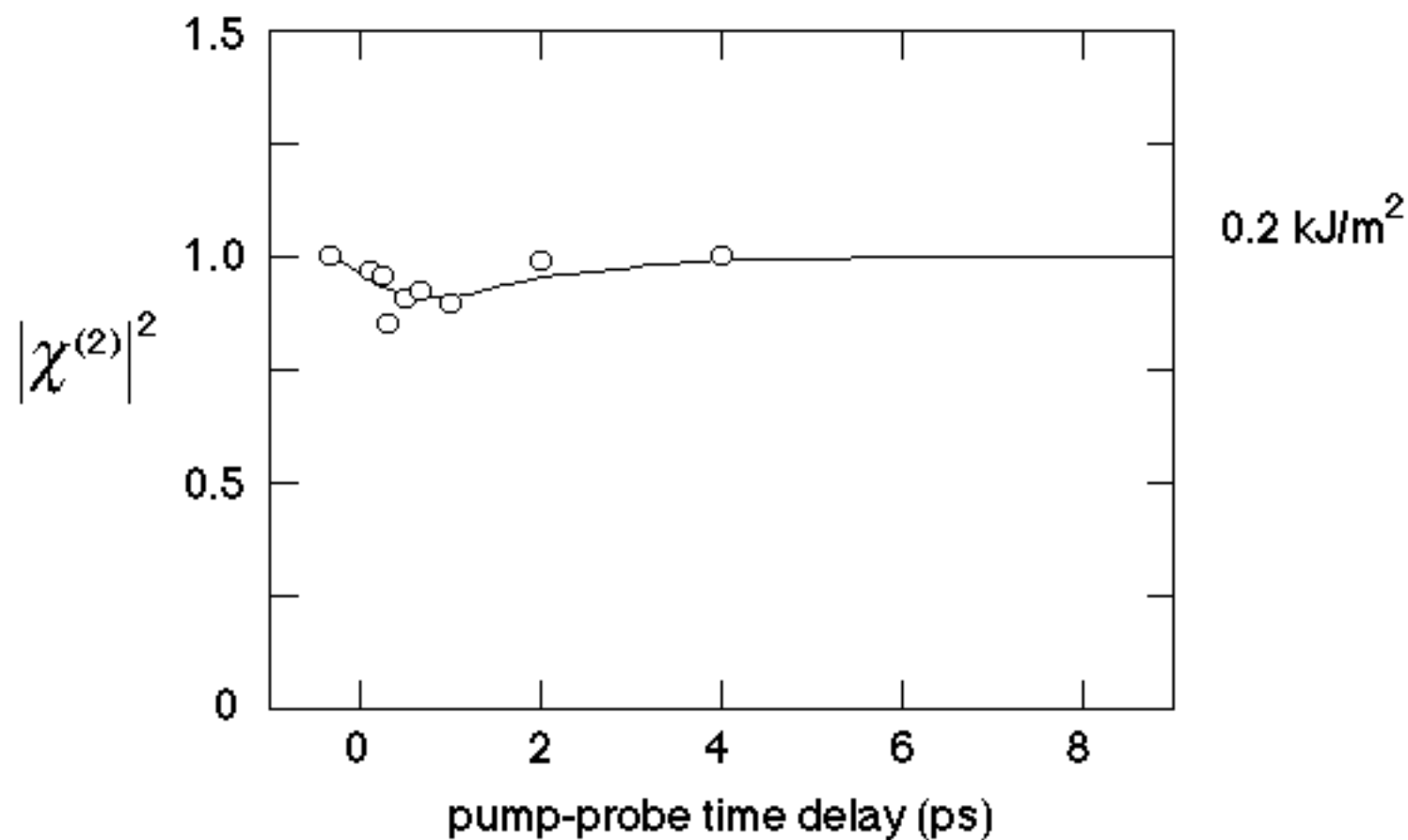
bandstructure



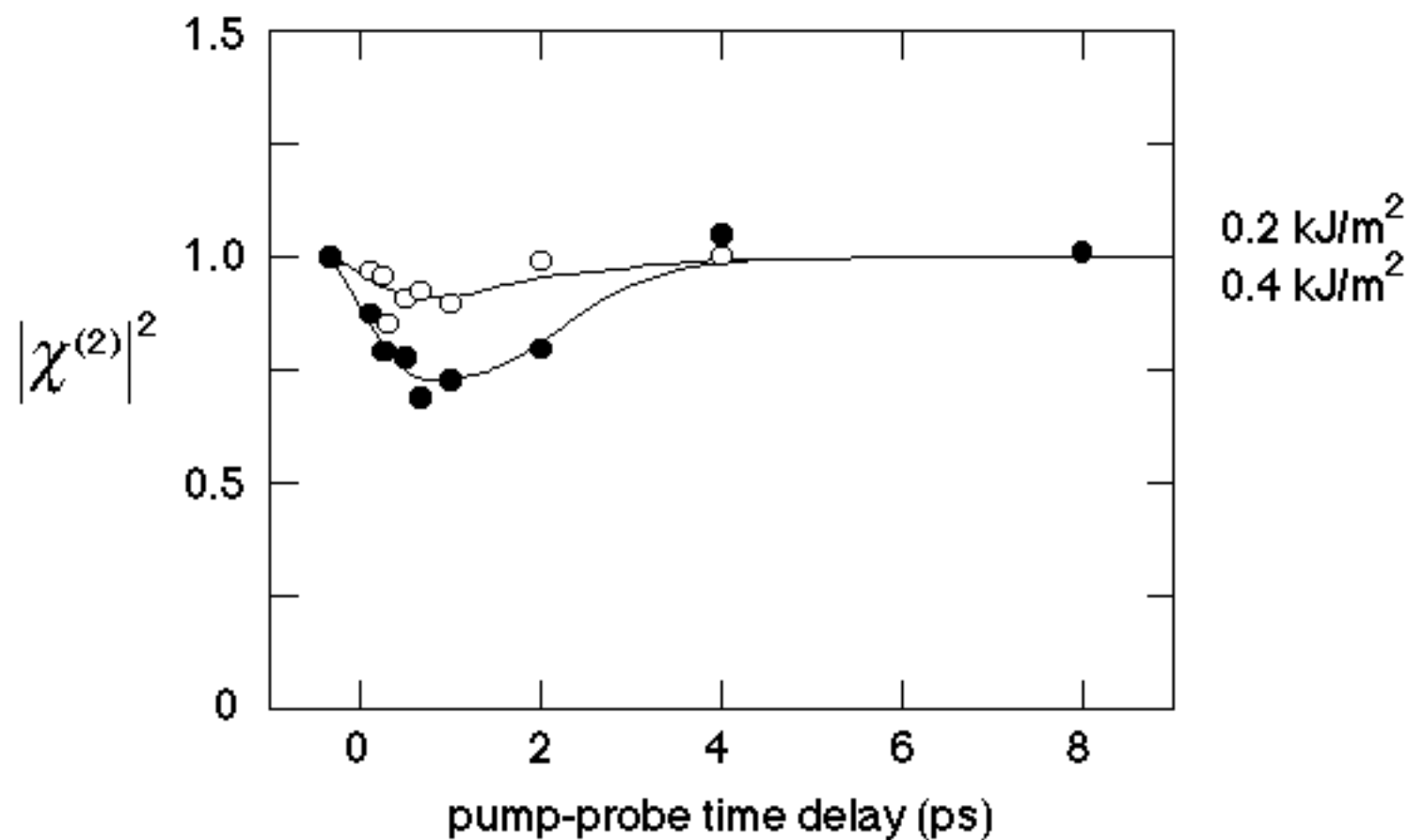
dielectric function



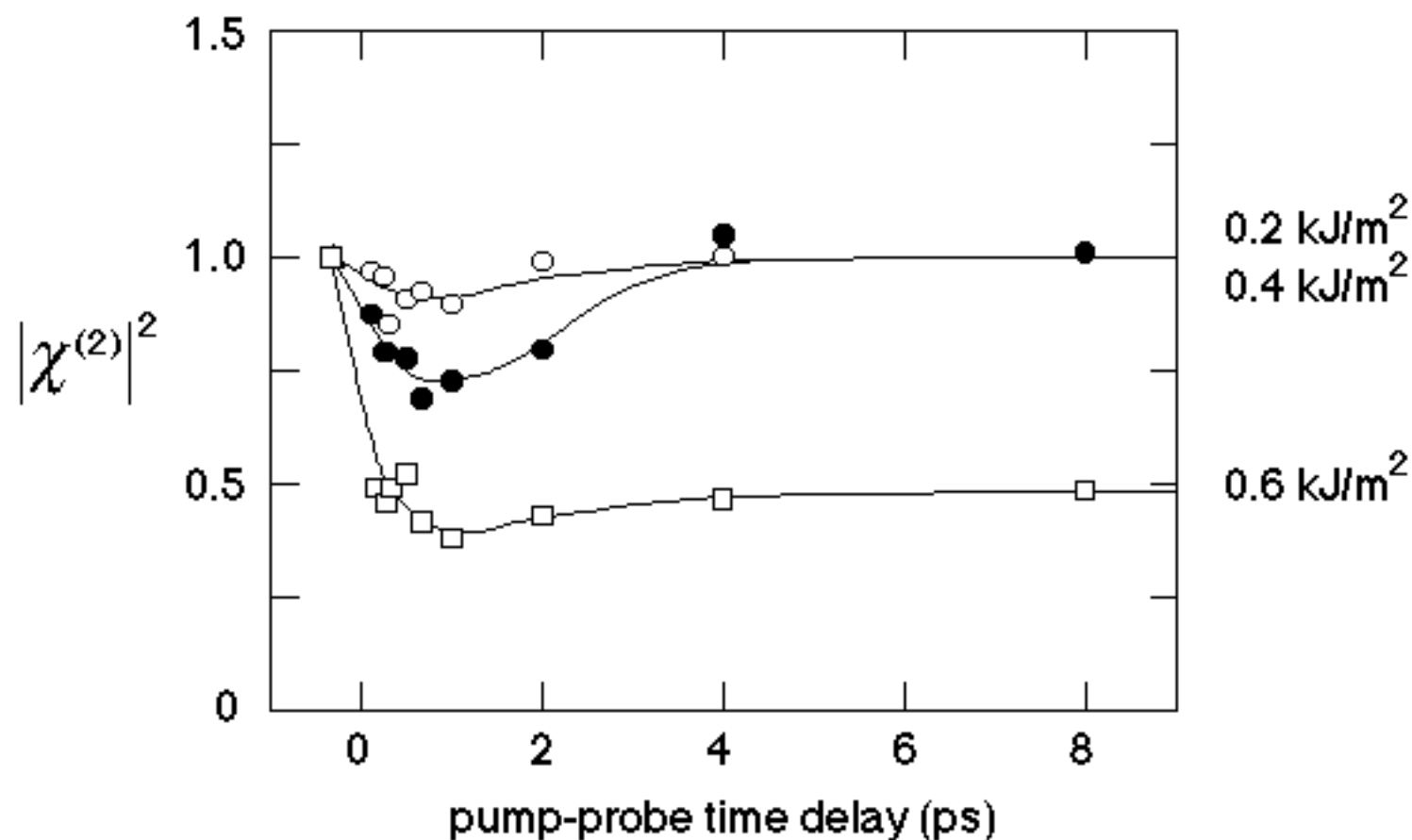
SECOND HARMONIC DATA



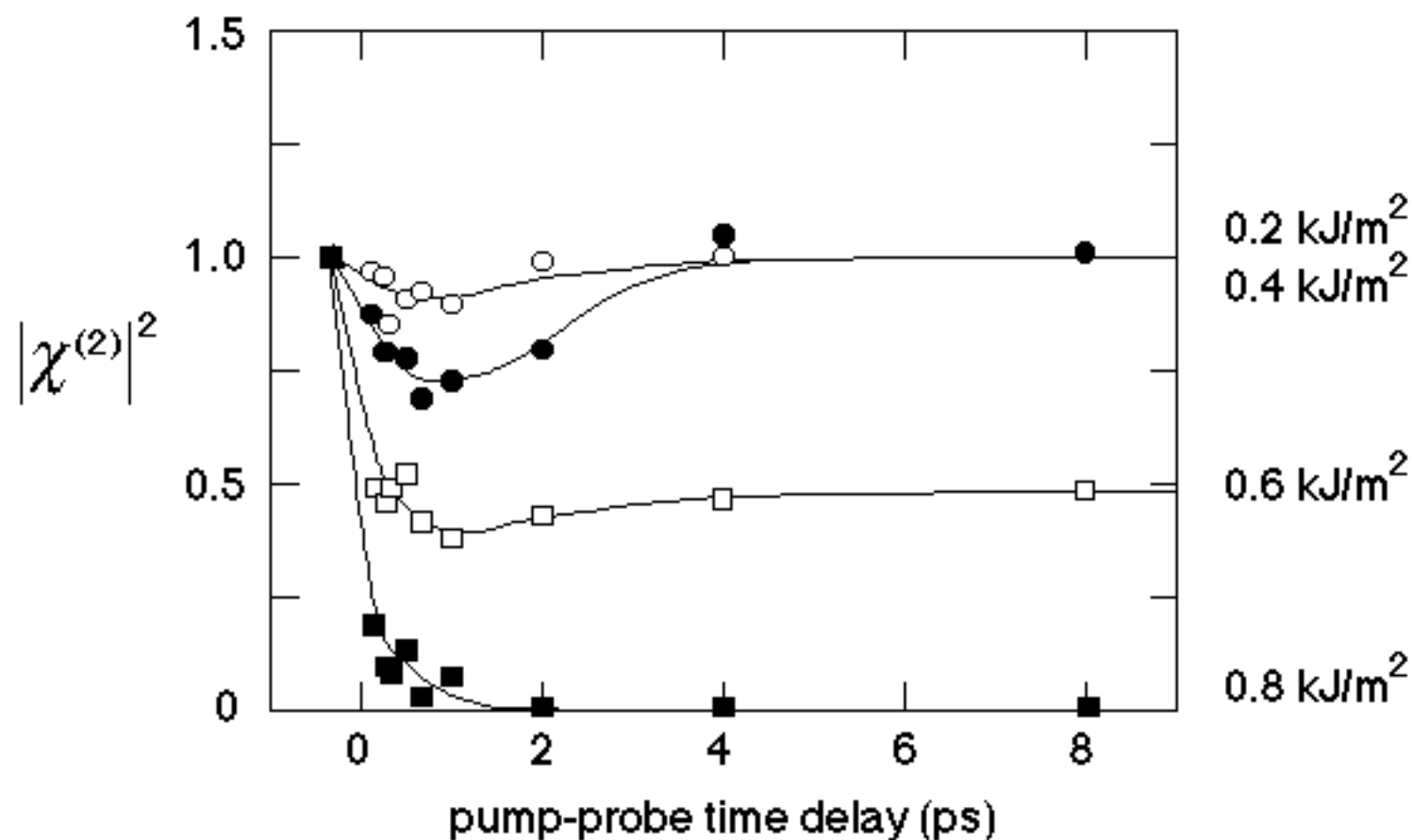
SECOND HARMONIC DATA



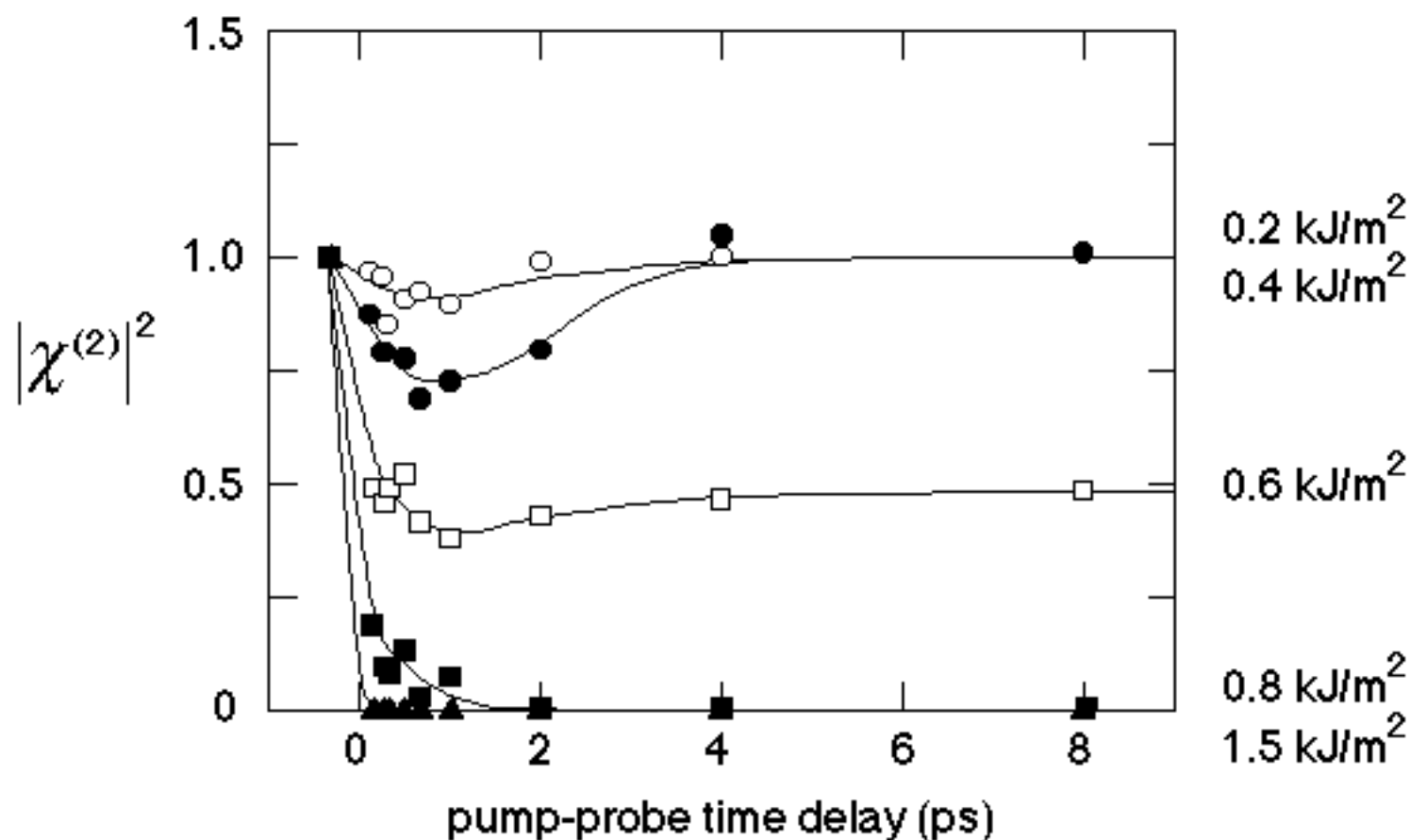
SECOND HARMONIC DATA



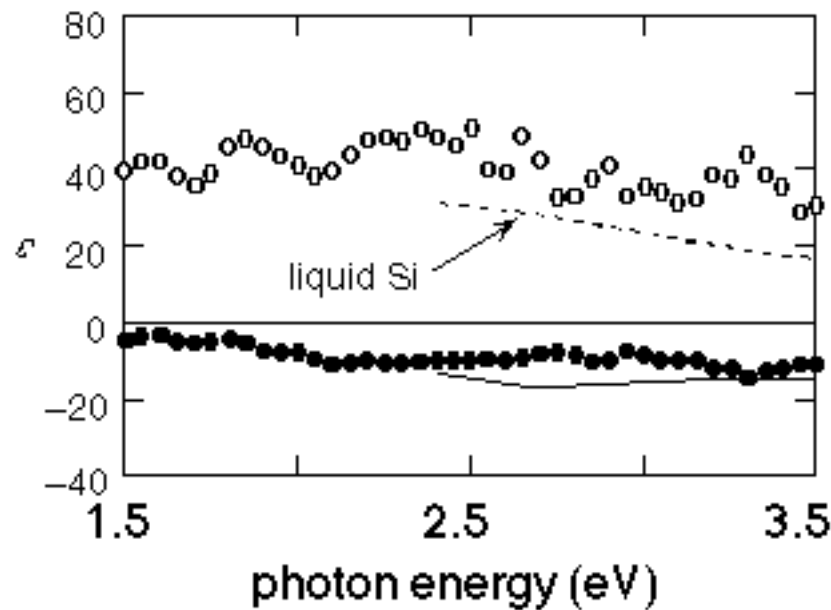
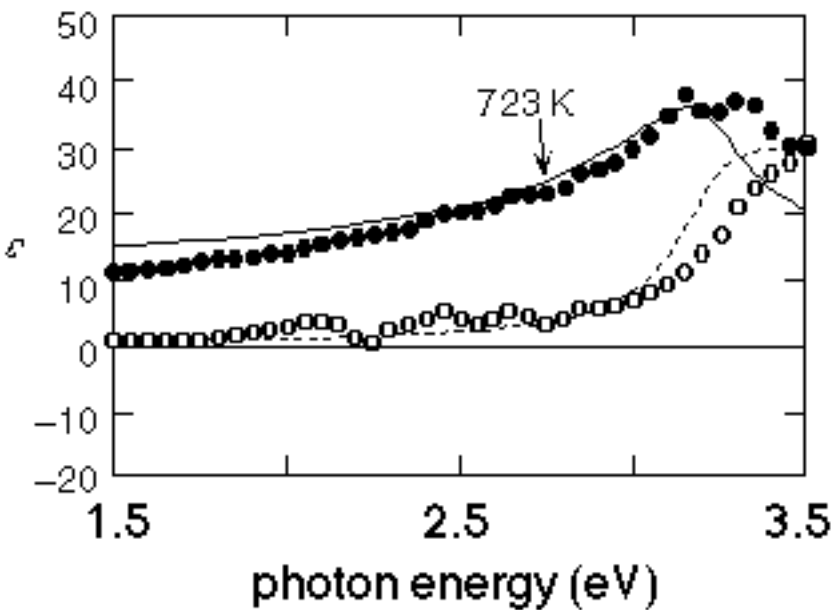
SECOND HARMONIC DATA



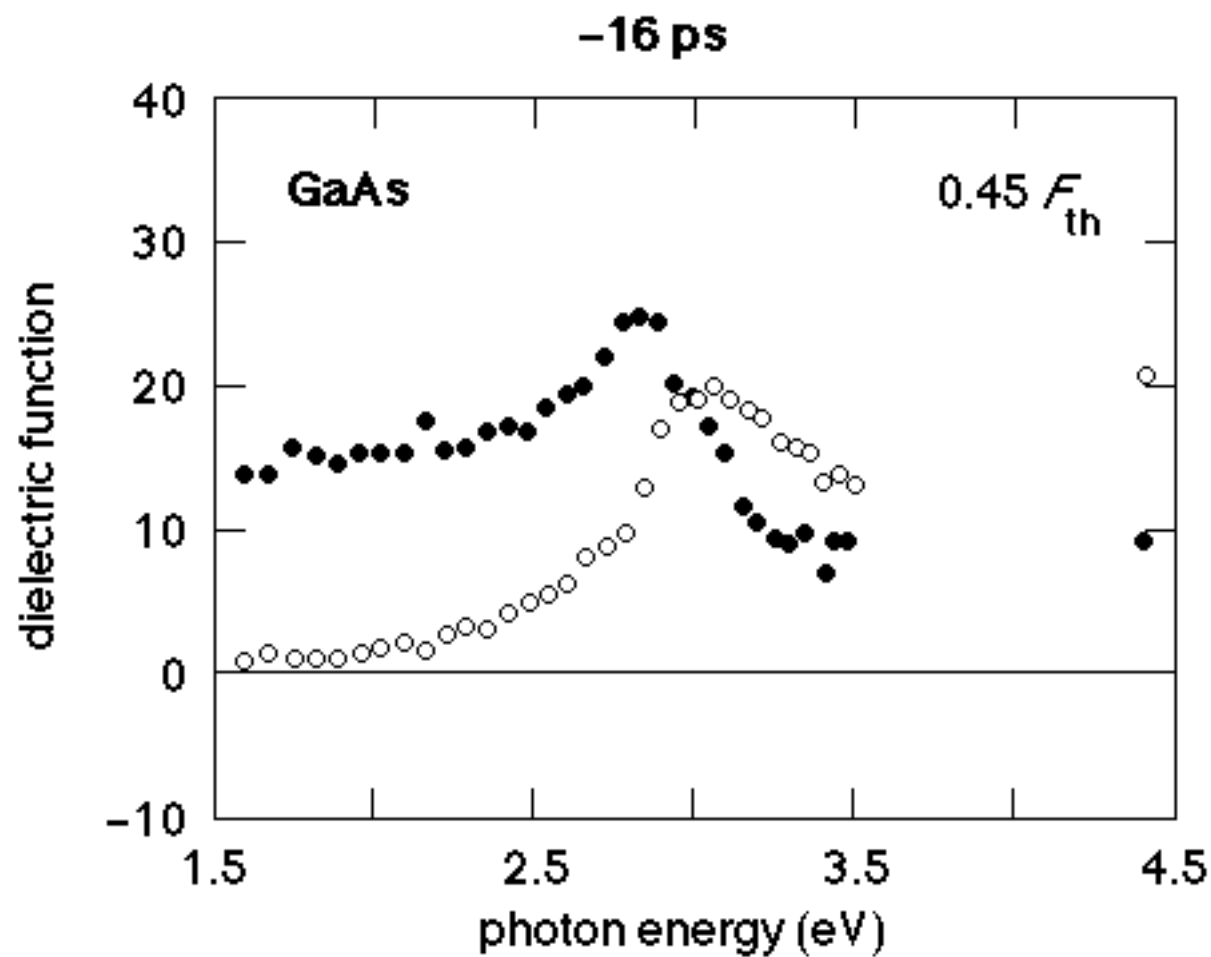
SECOND HARMONIC DATA



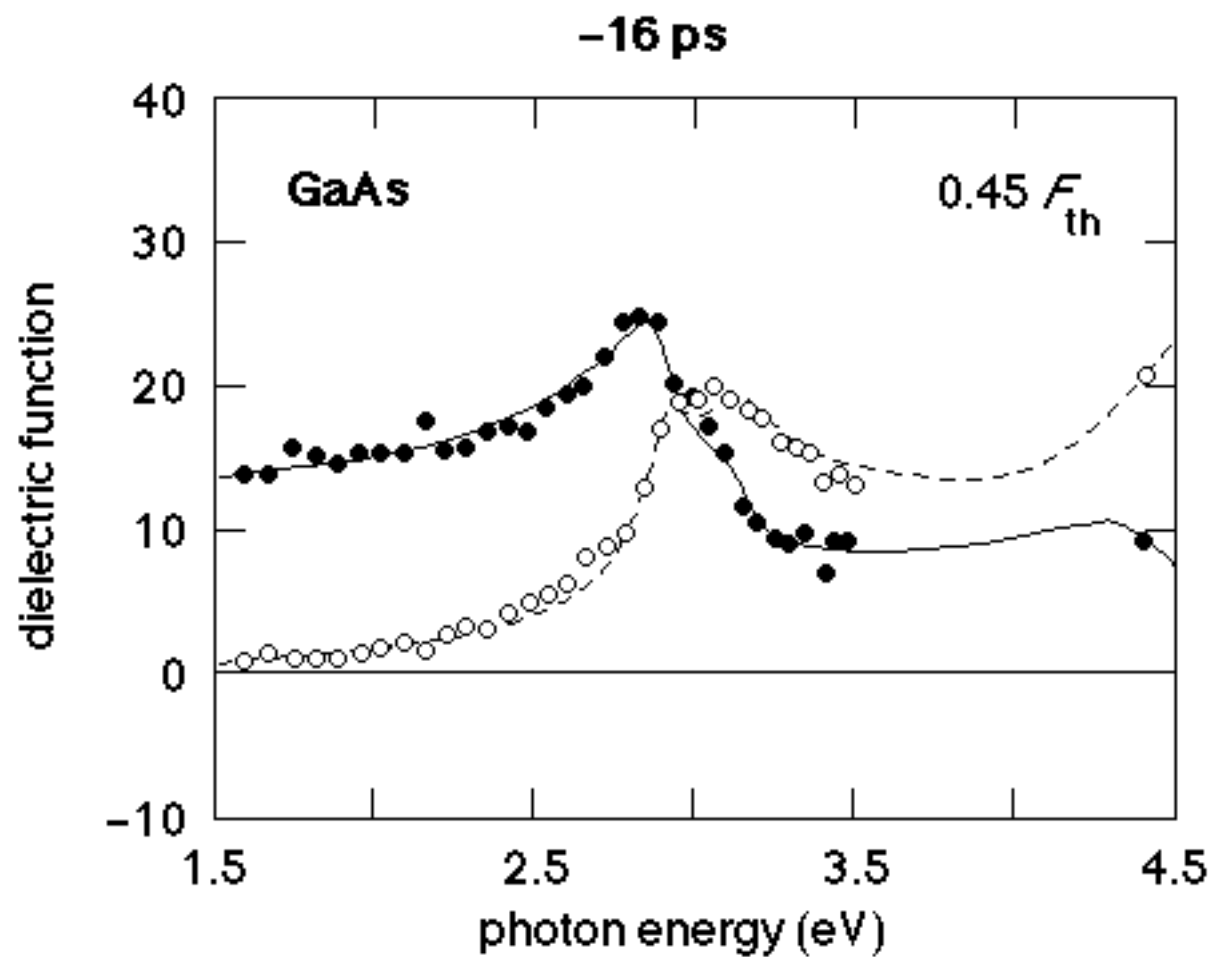
PRELIMINARY RESULTS *Si*



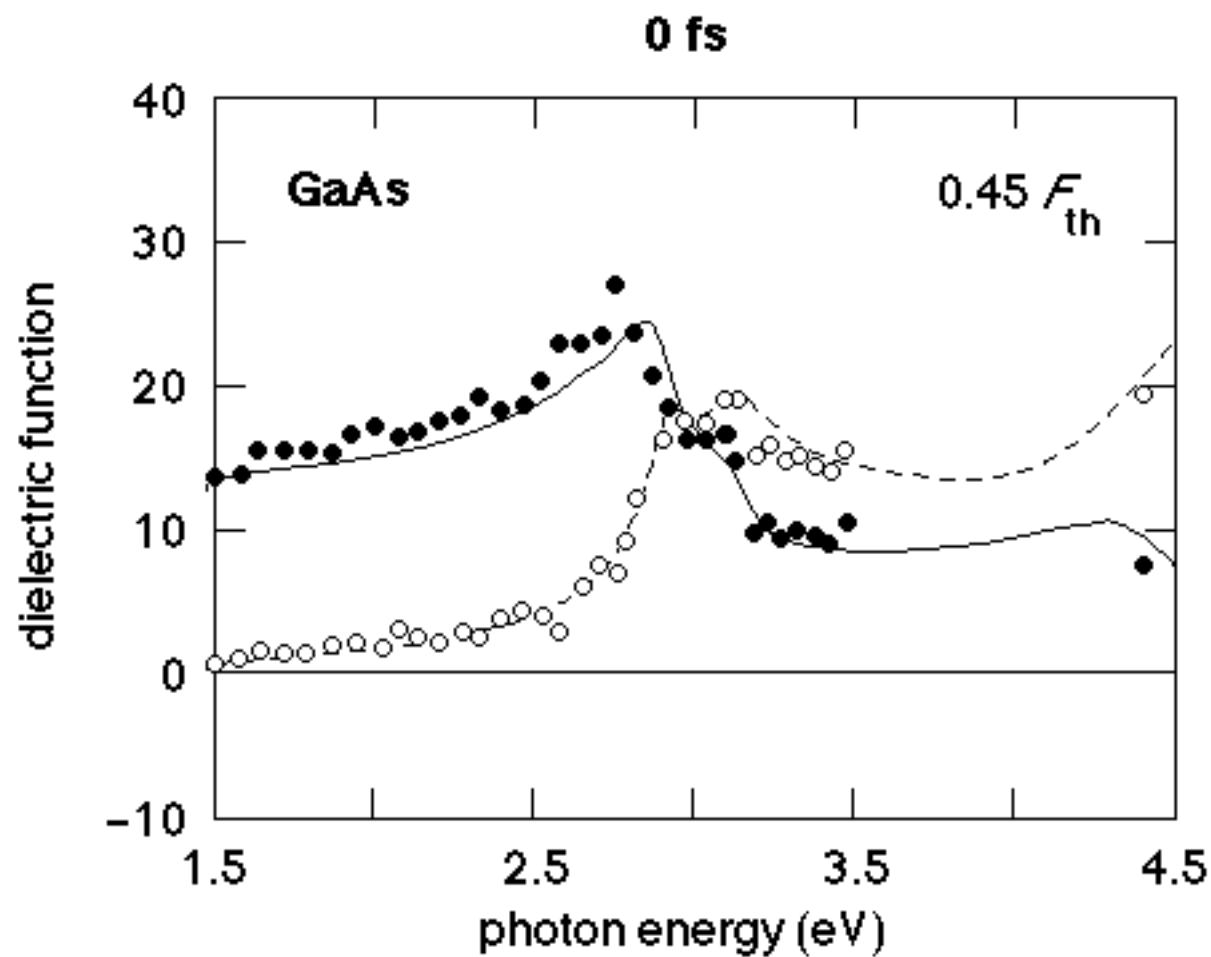
LOW FLUENCE DATA



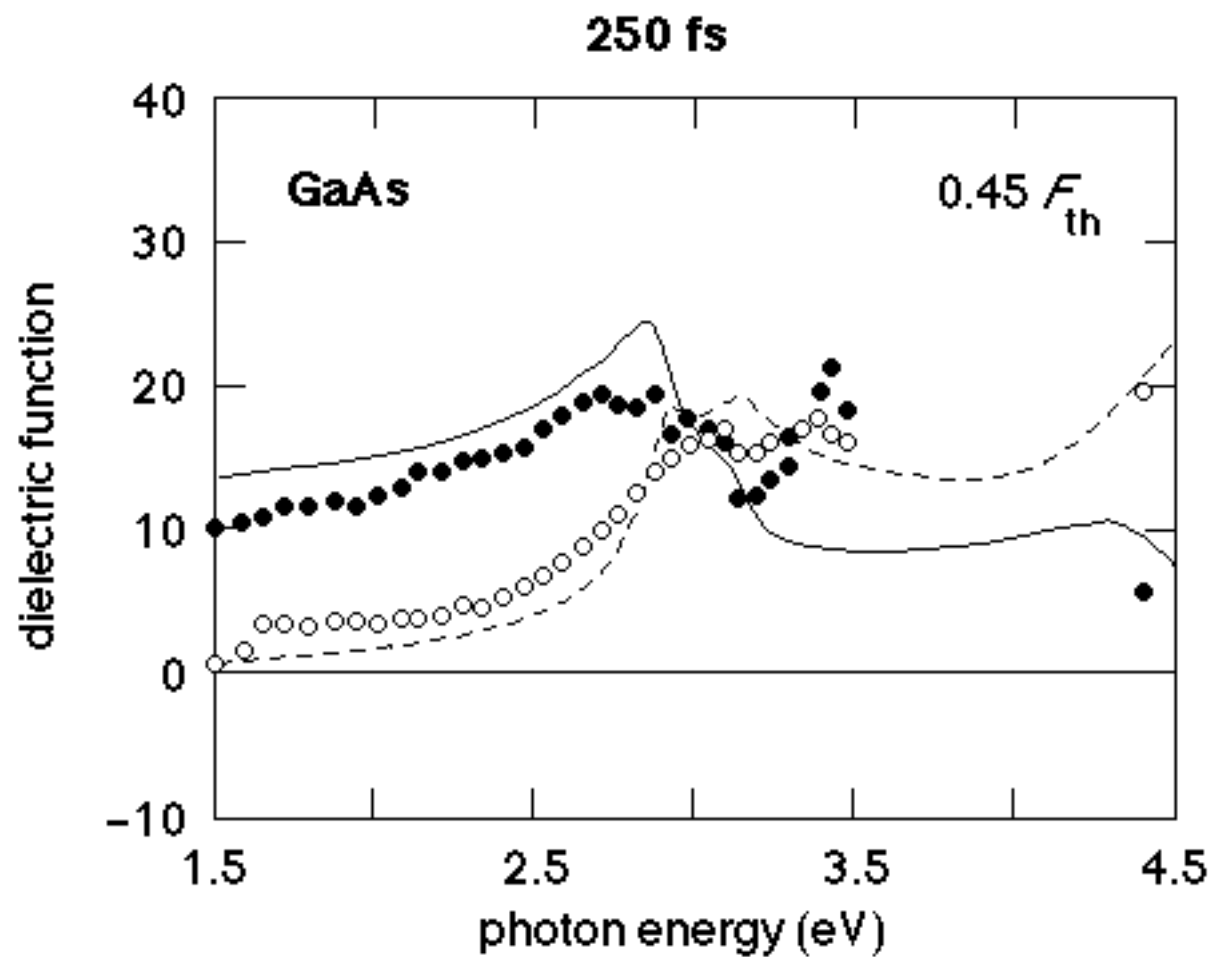
LOW FLUENCE DATA



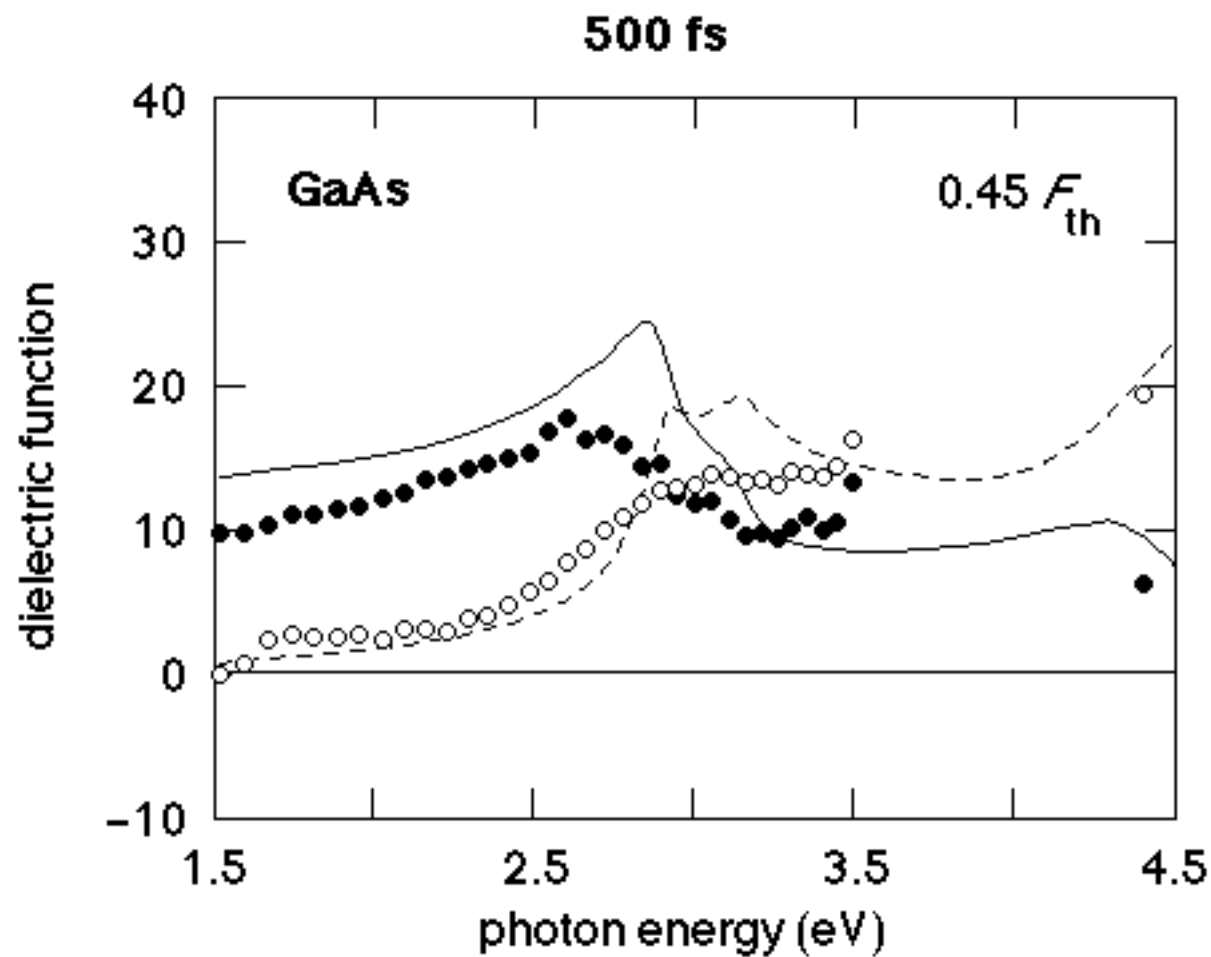
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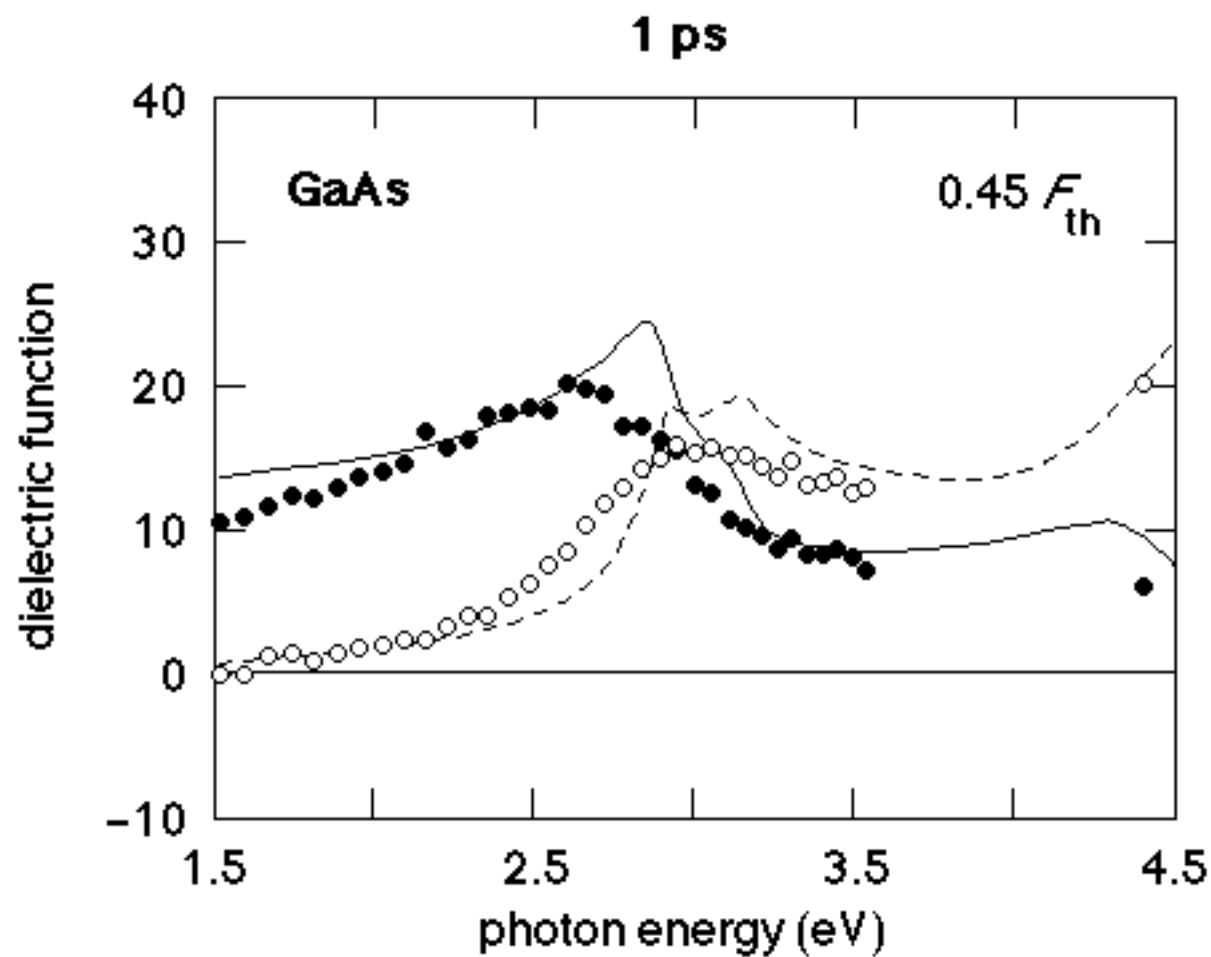
LOW FLUENCE DATA



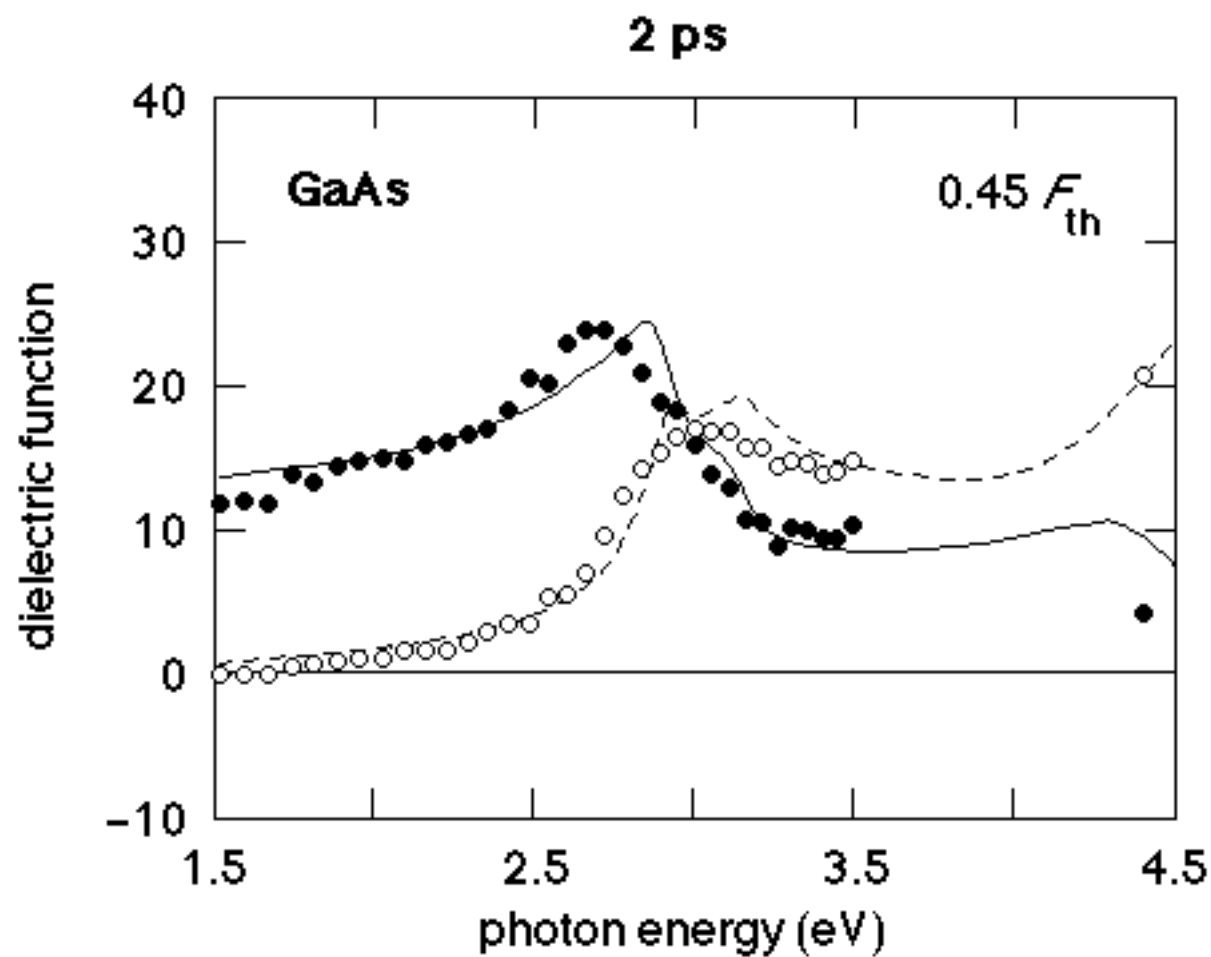
LOW FLUENCE DATA



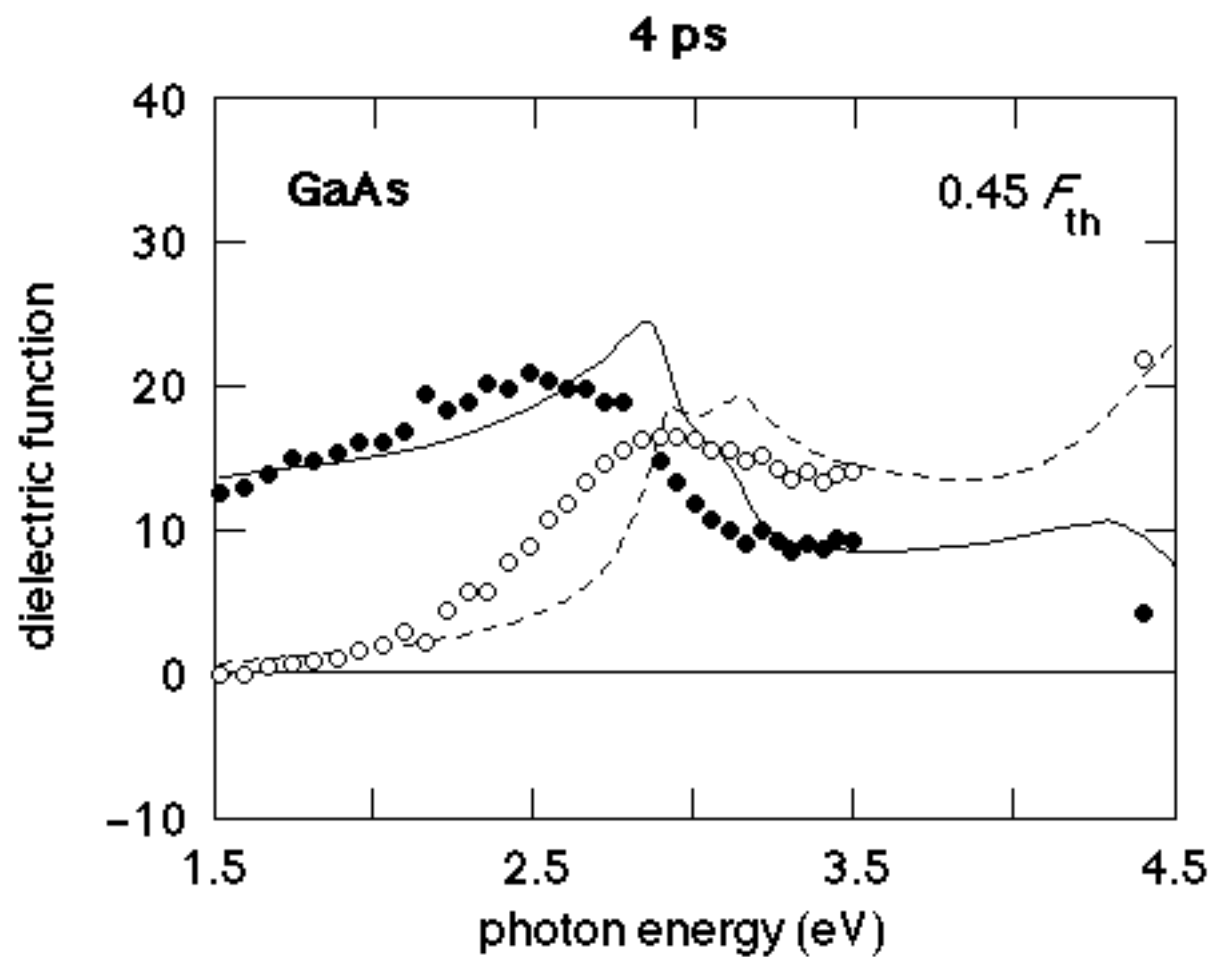
LOW FLUENCE DATA



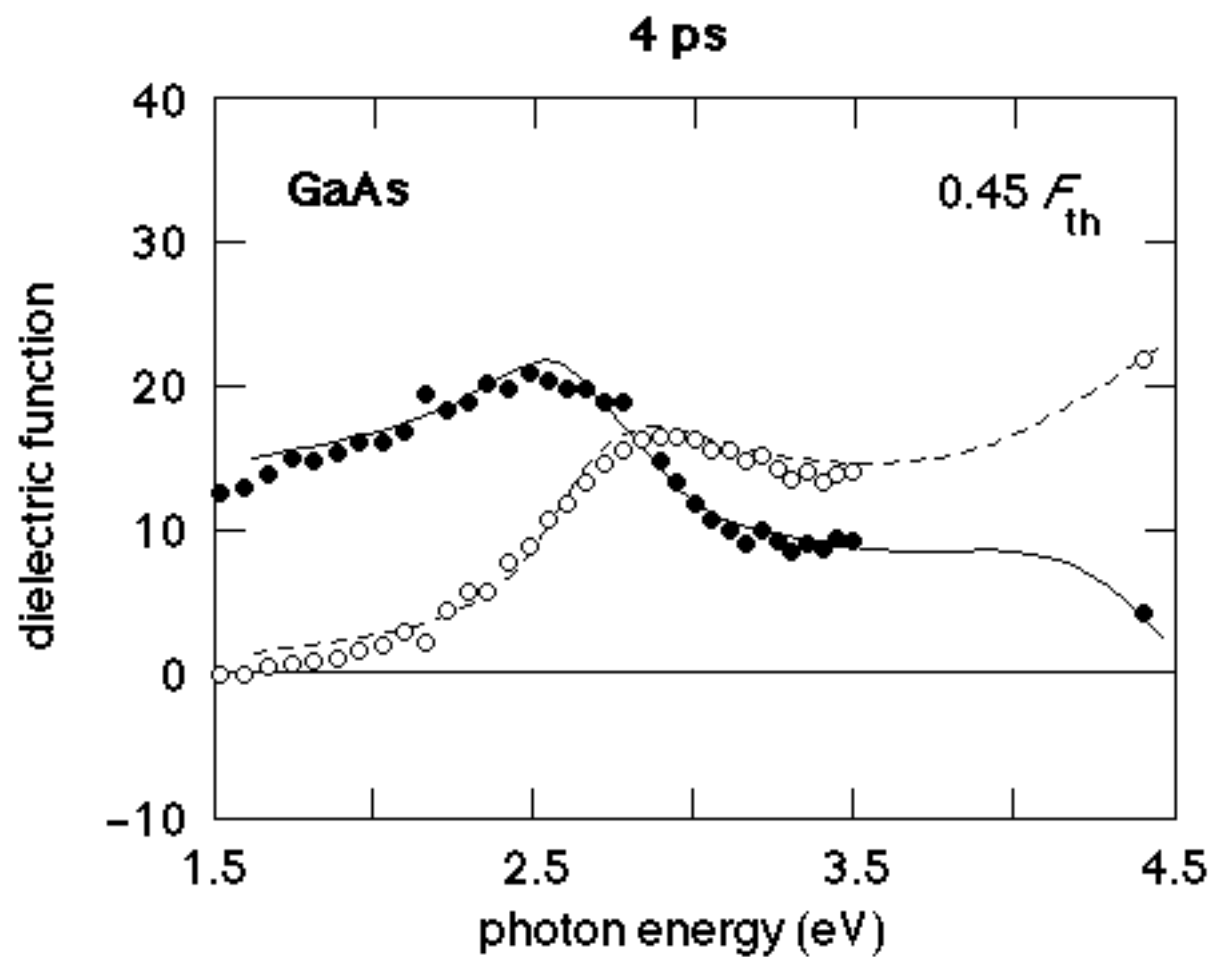
LOW FLUENCE DATA



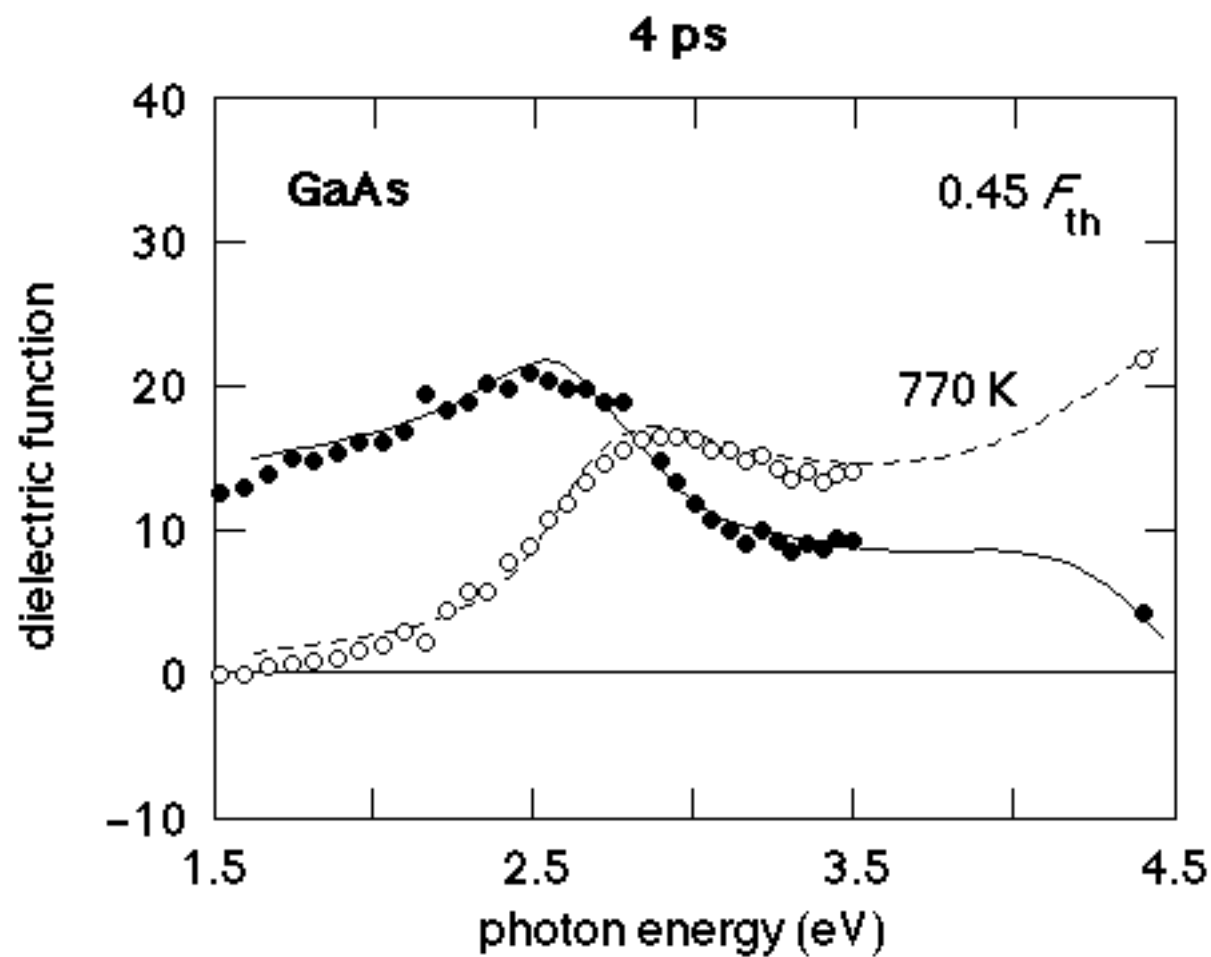
LOW FLUENCE DATA



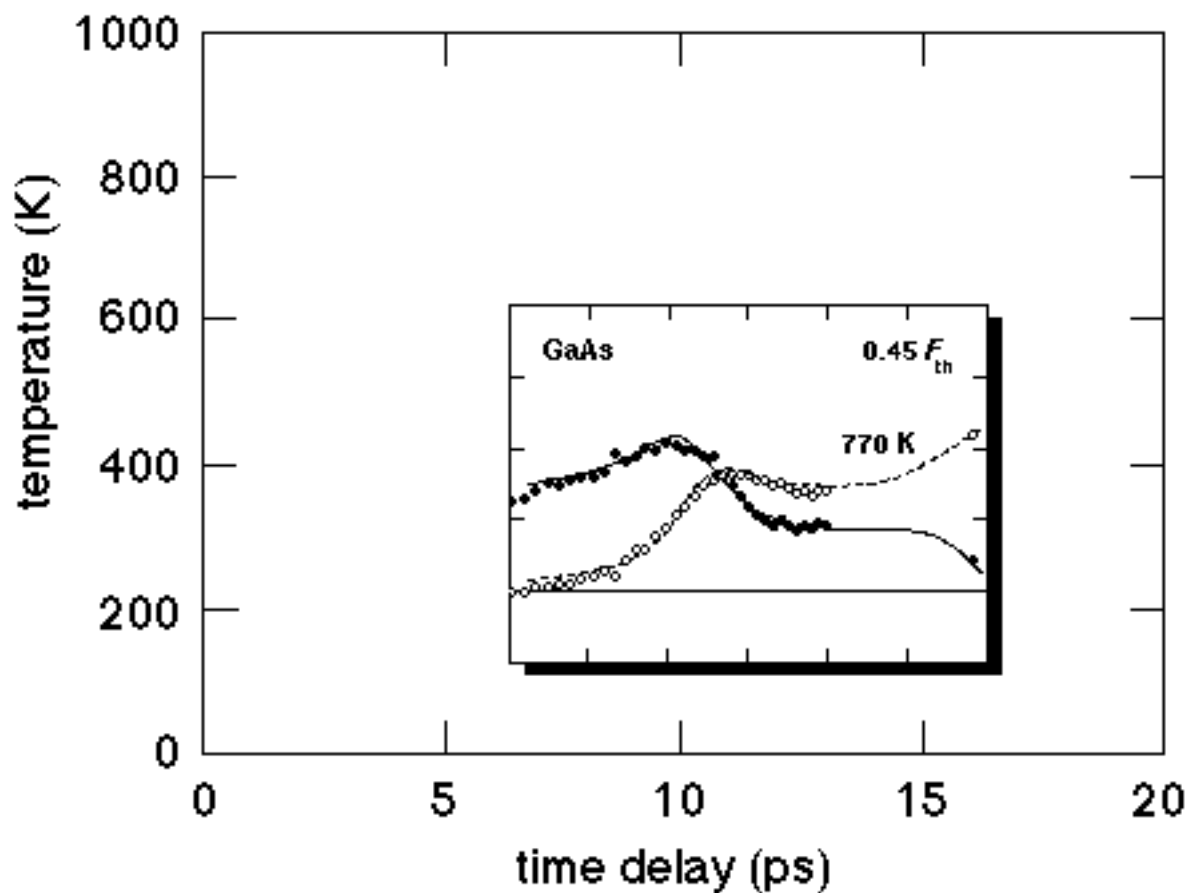
LOW FLUENCE DATA



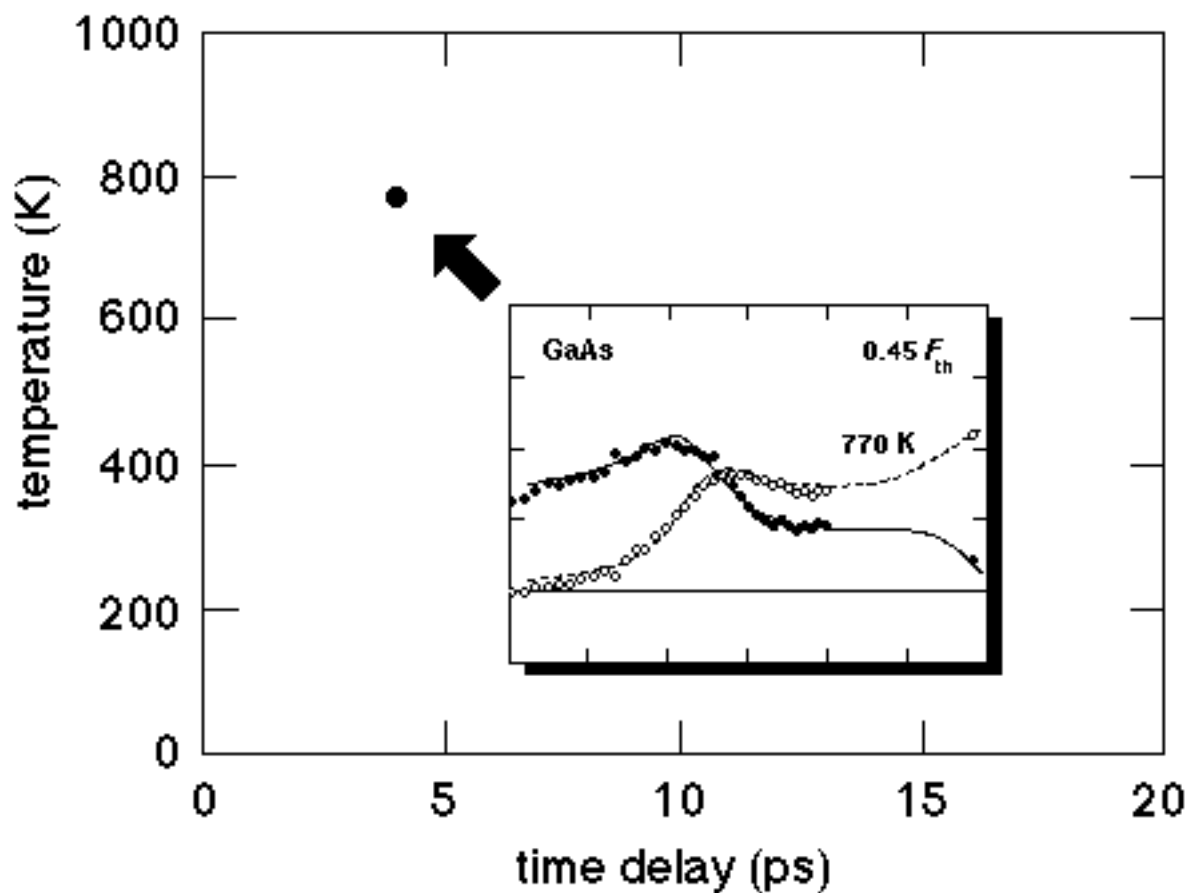
LOW FLUENCE DATA



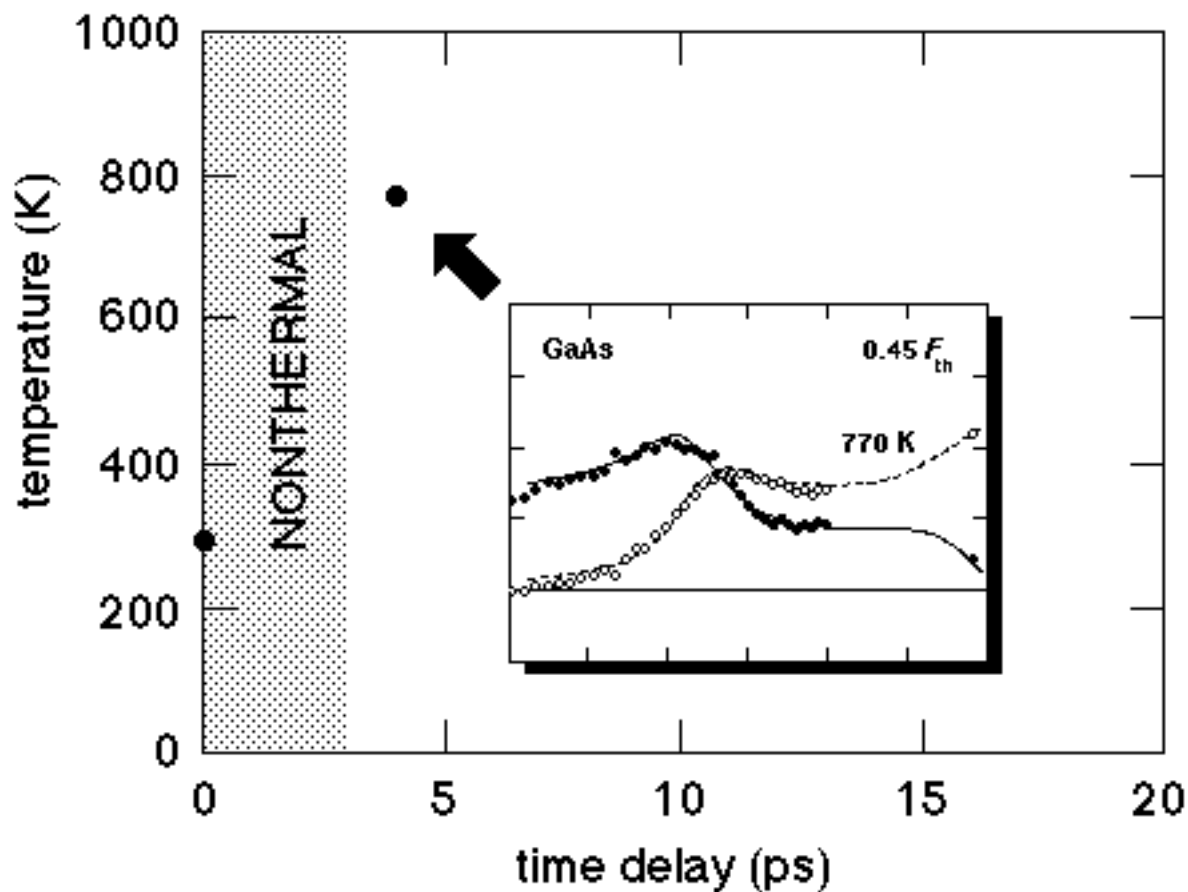
TEMPERATURE



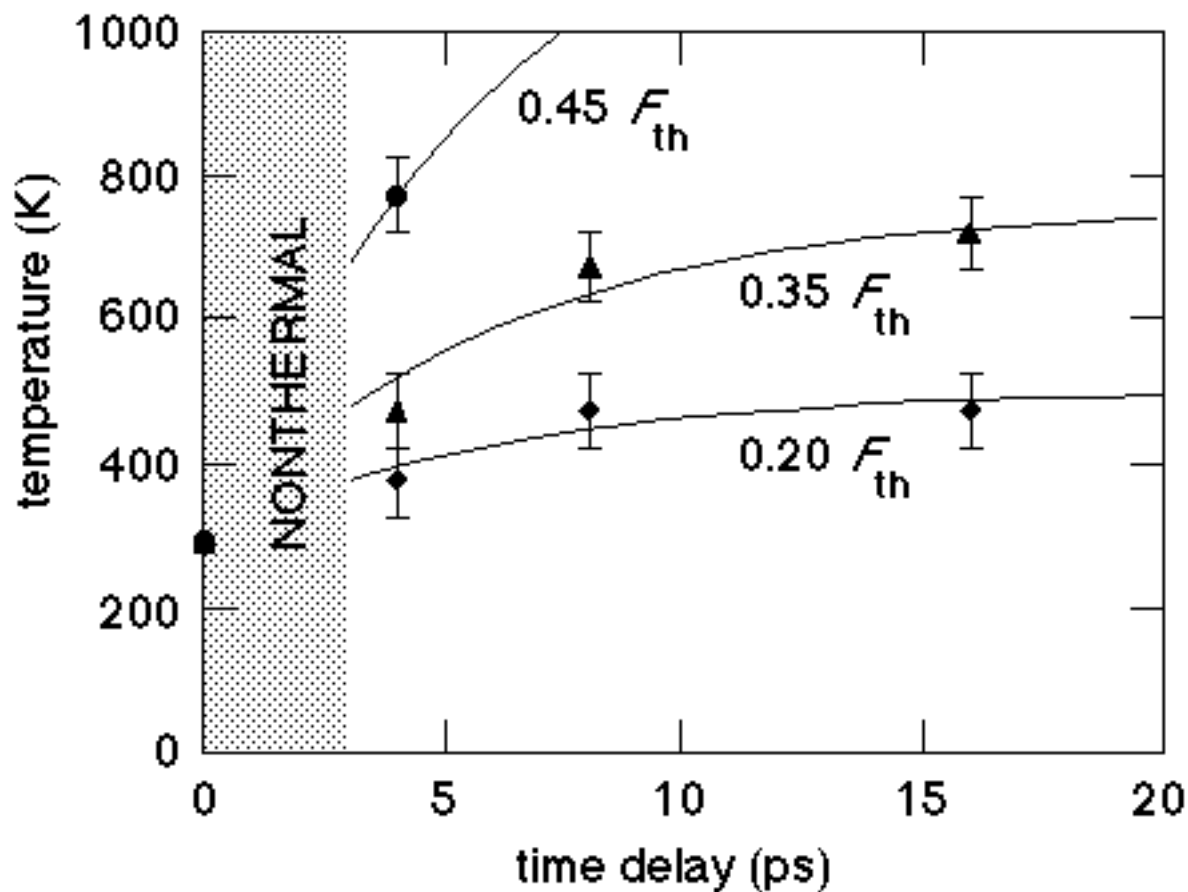
TEMPERATURE



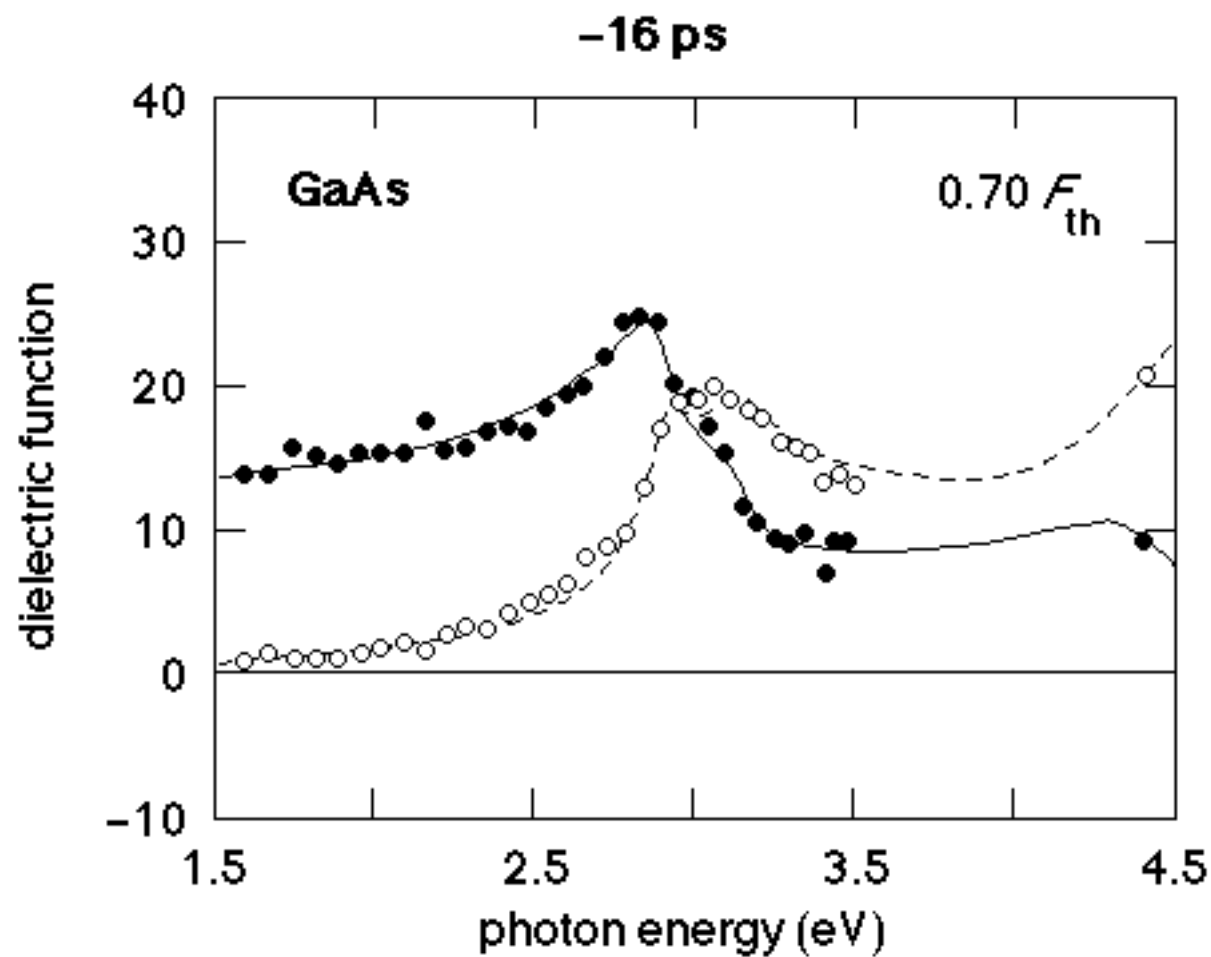
TEMPERATURE



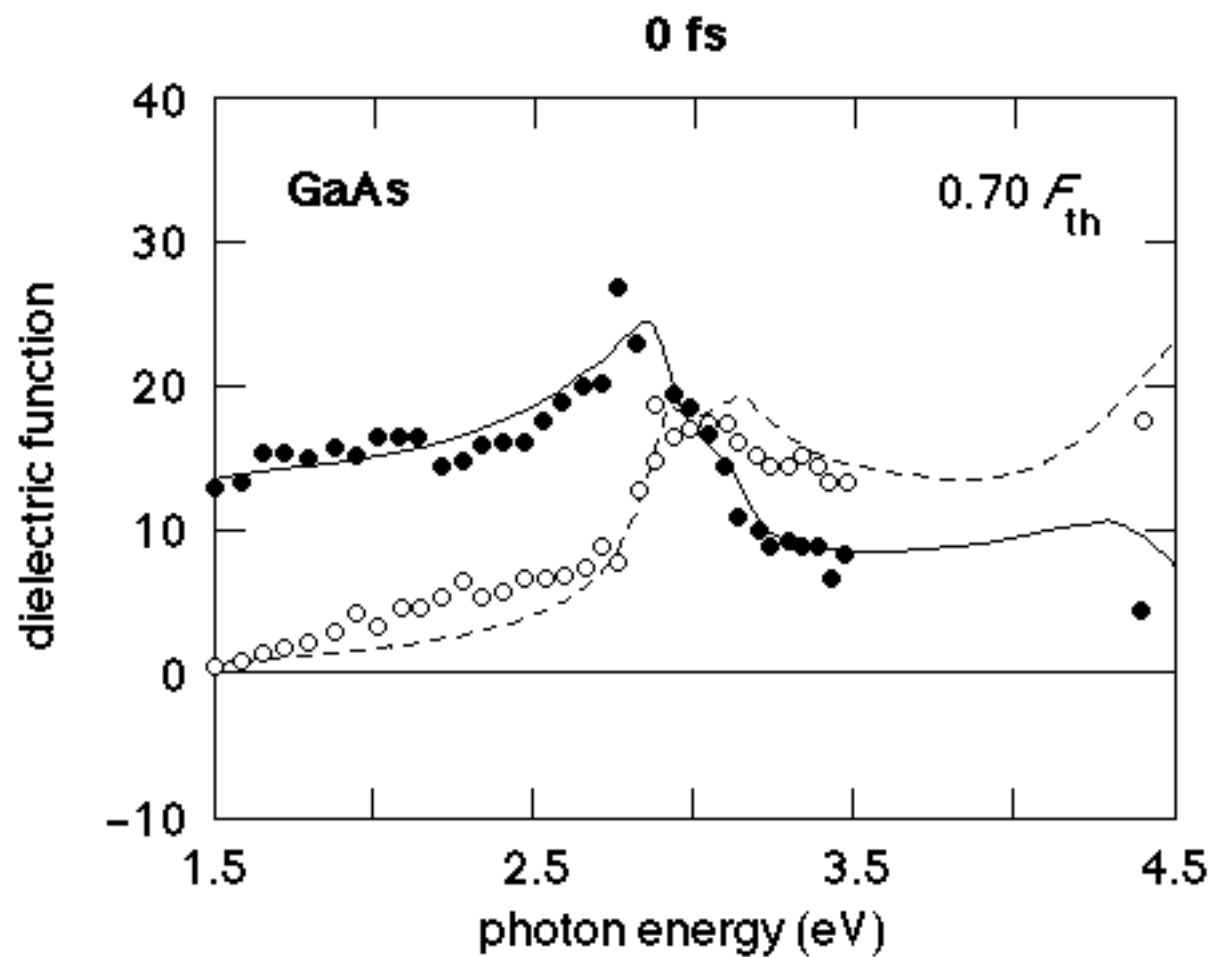
TEMPERATURE



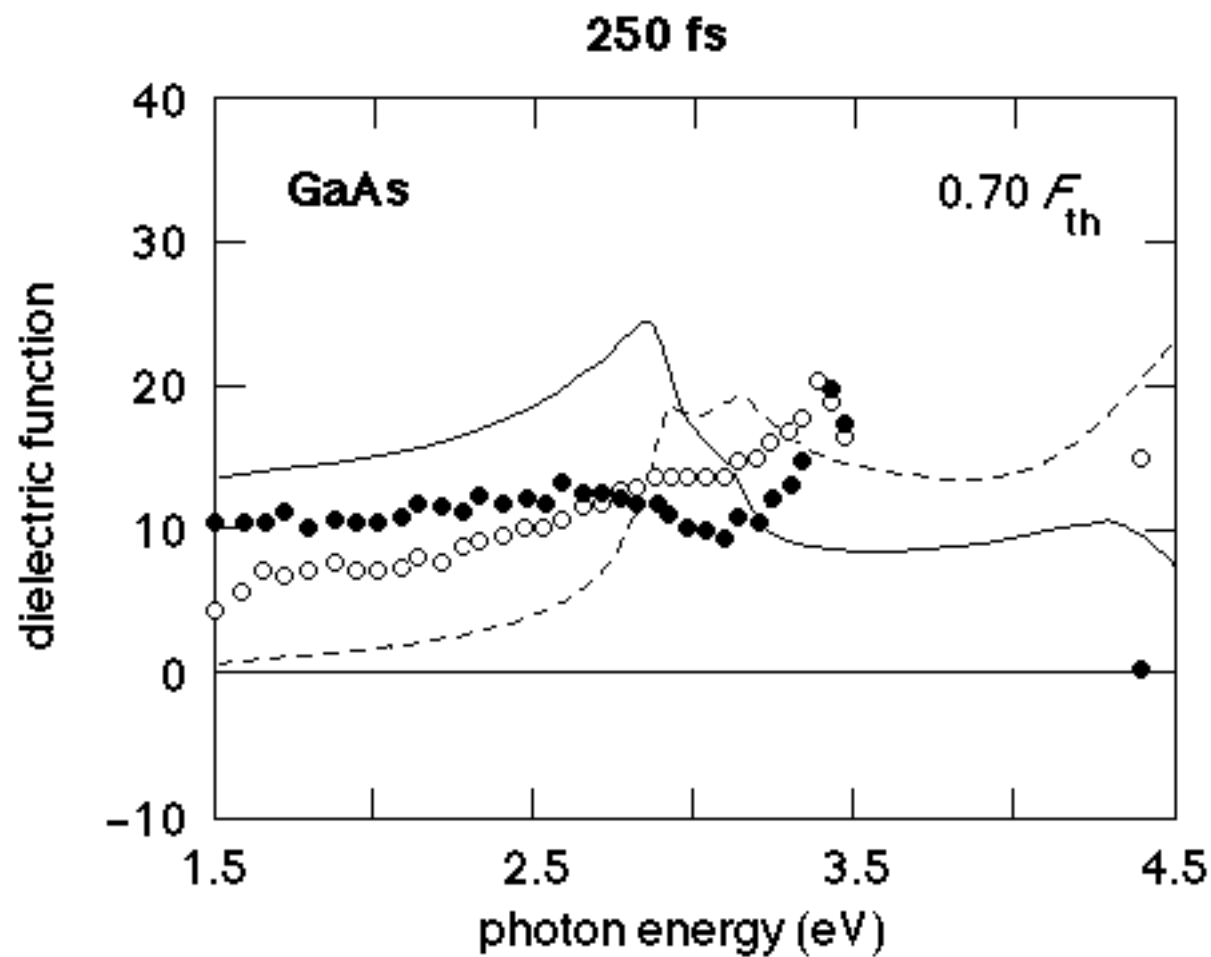
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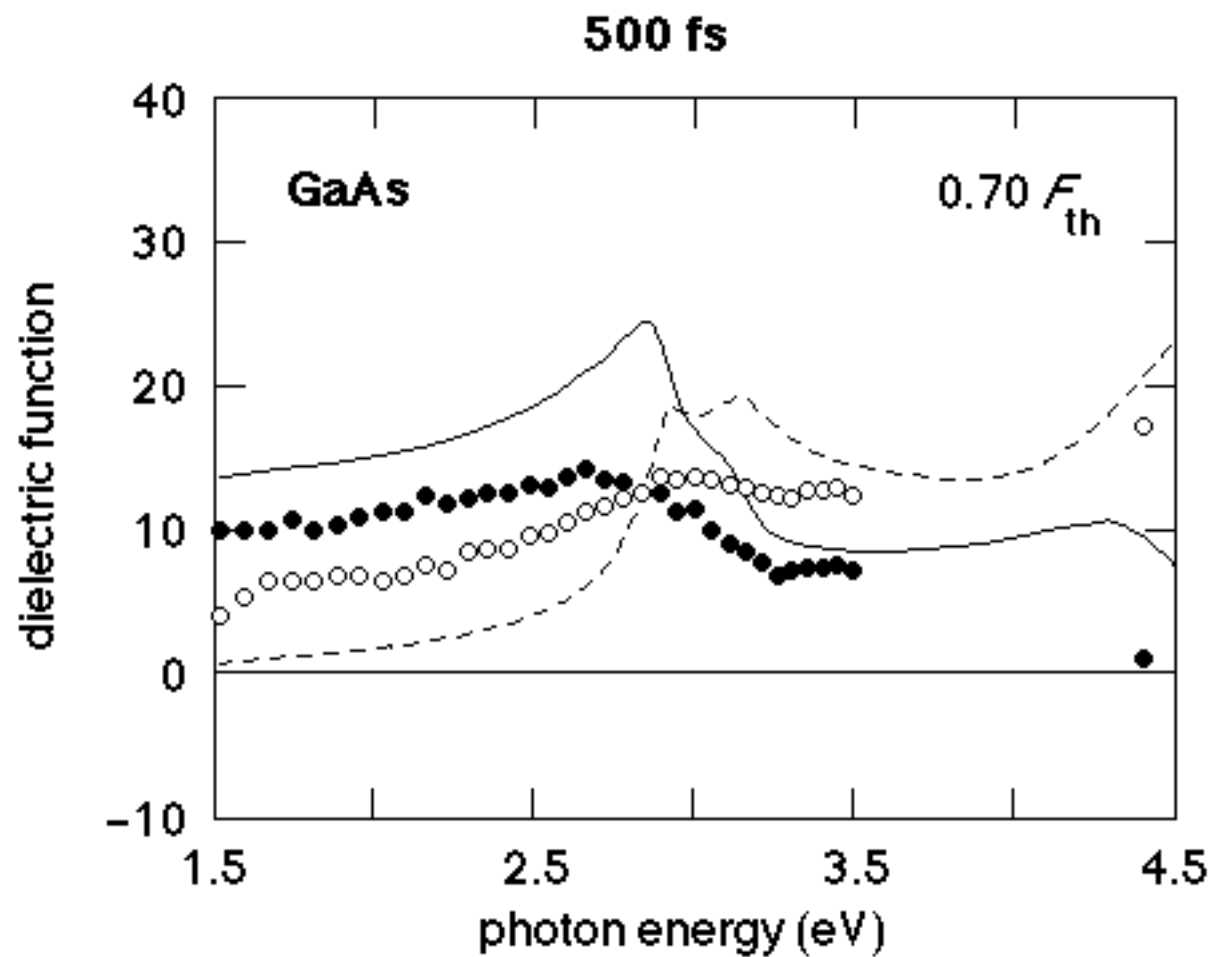
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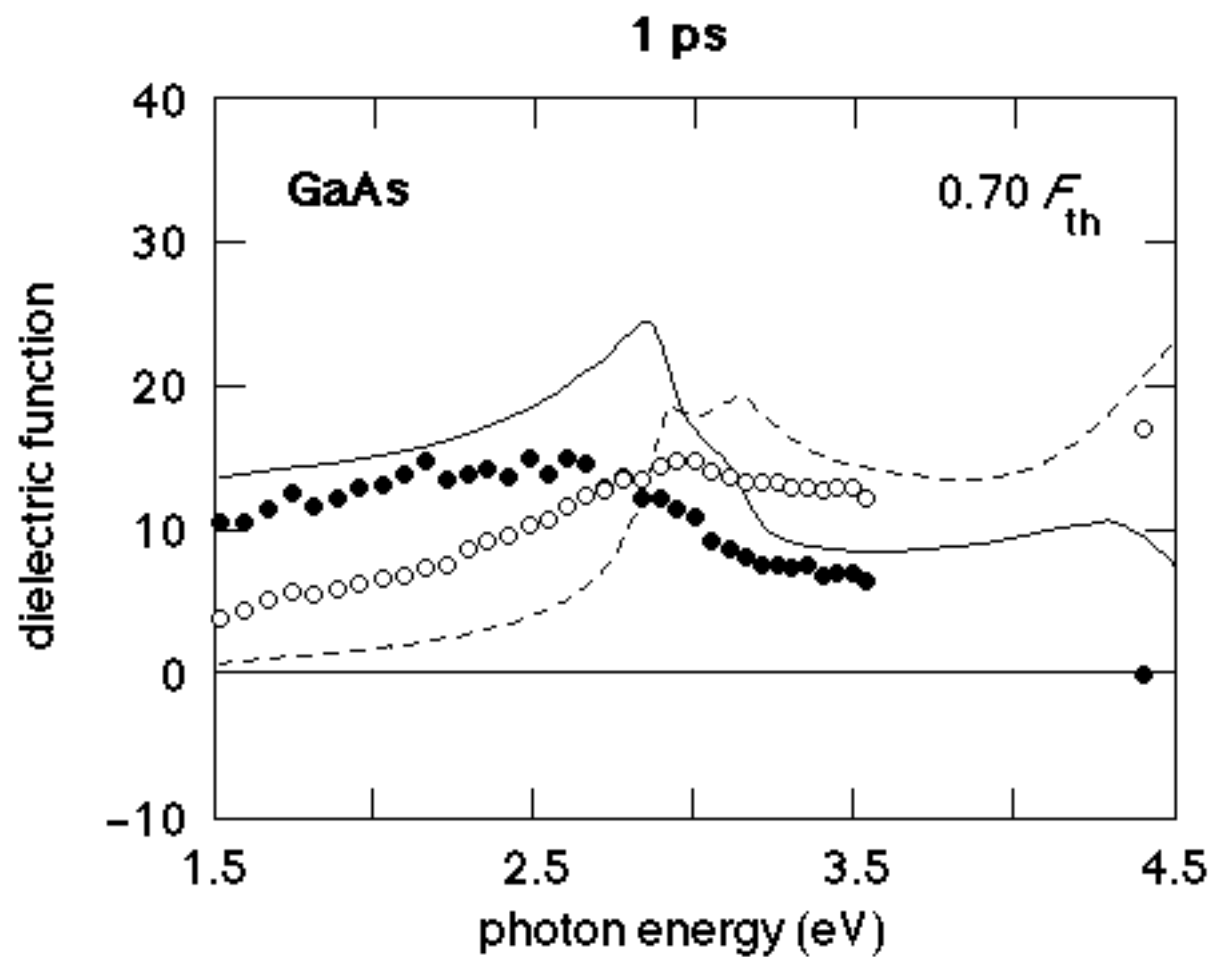
MEDIUM FLUENCE DATA



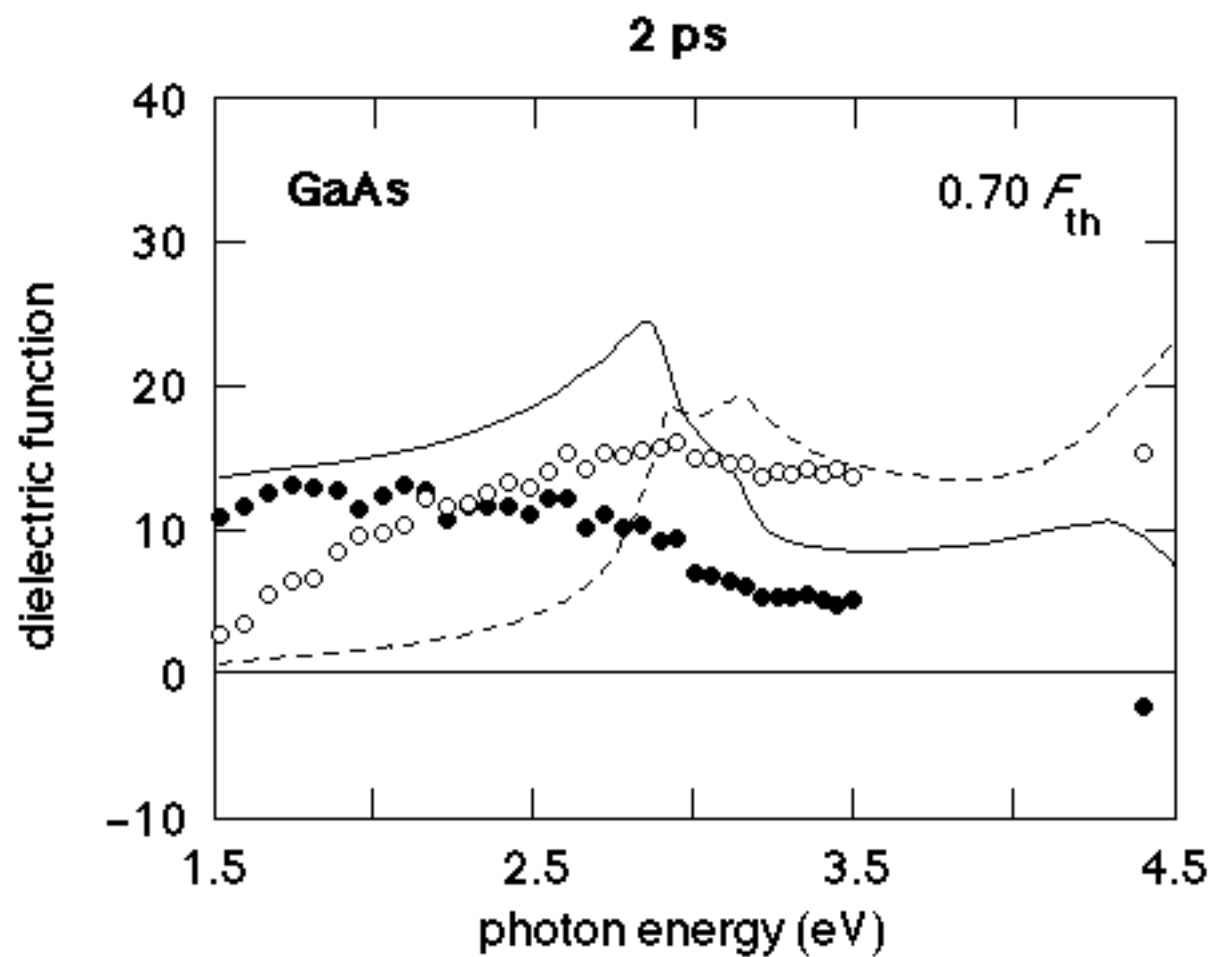
MEDIUM FLUENCE DATA



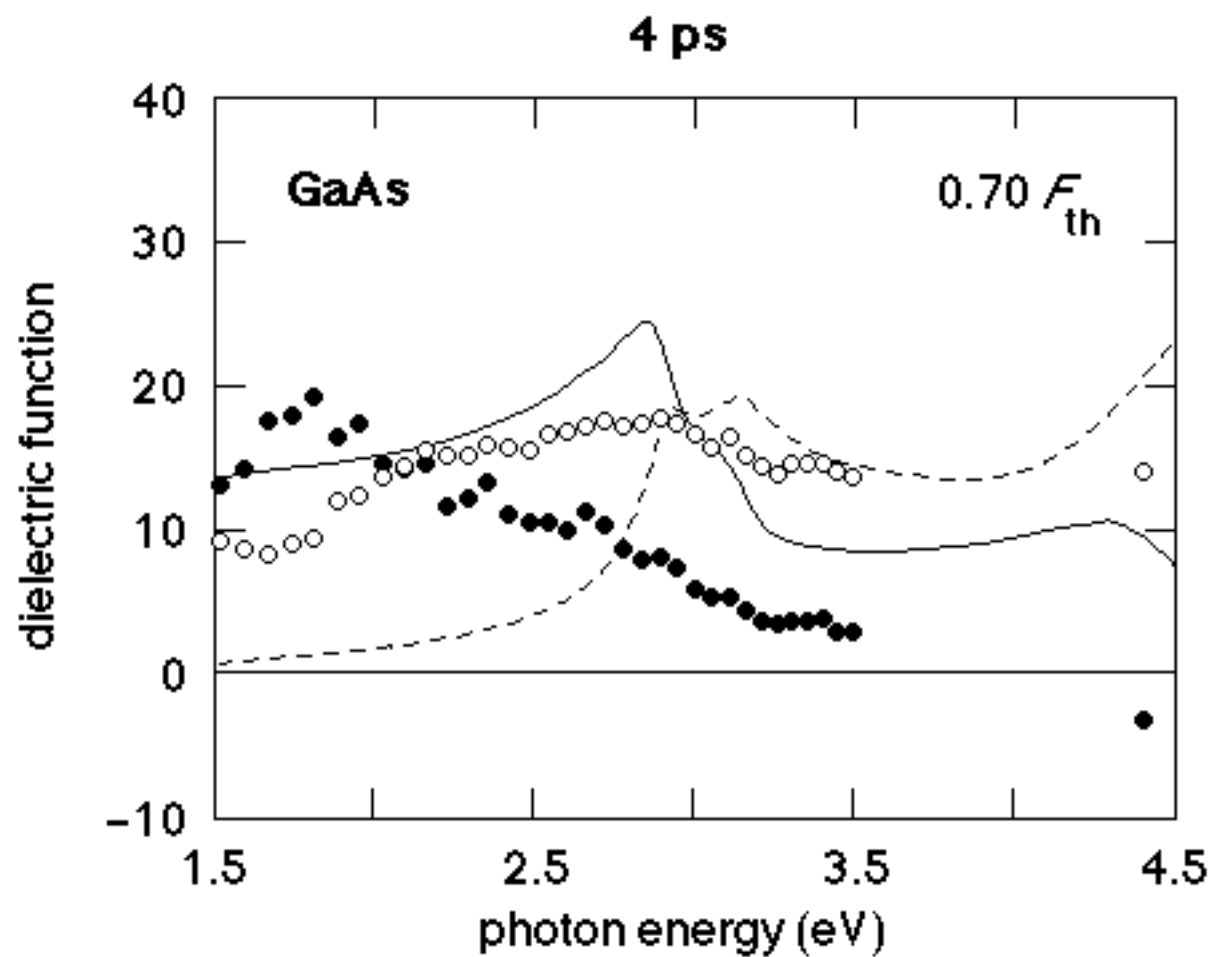
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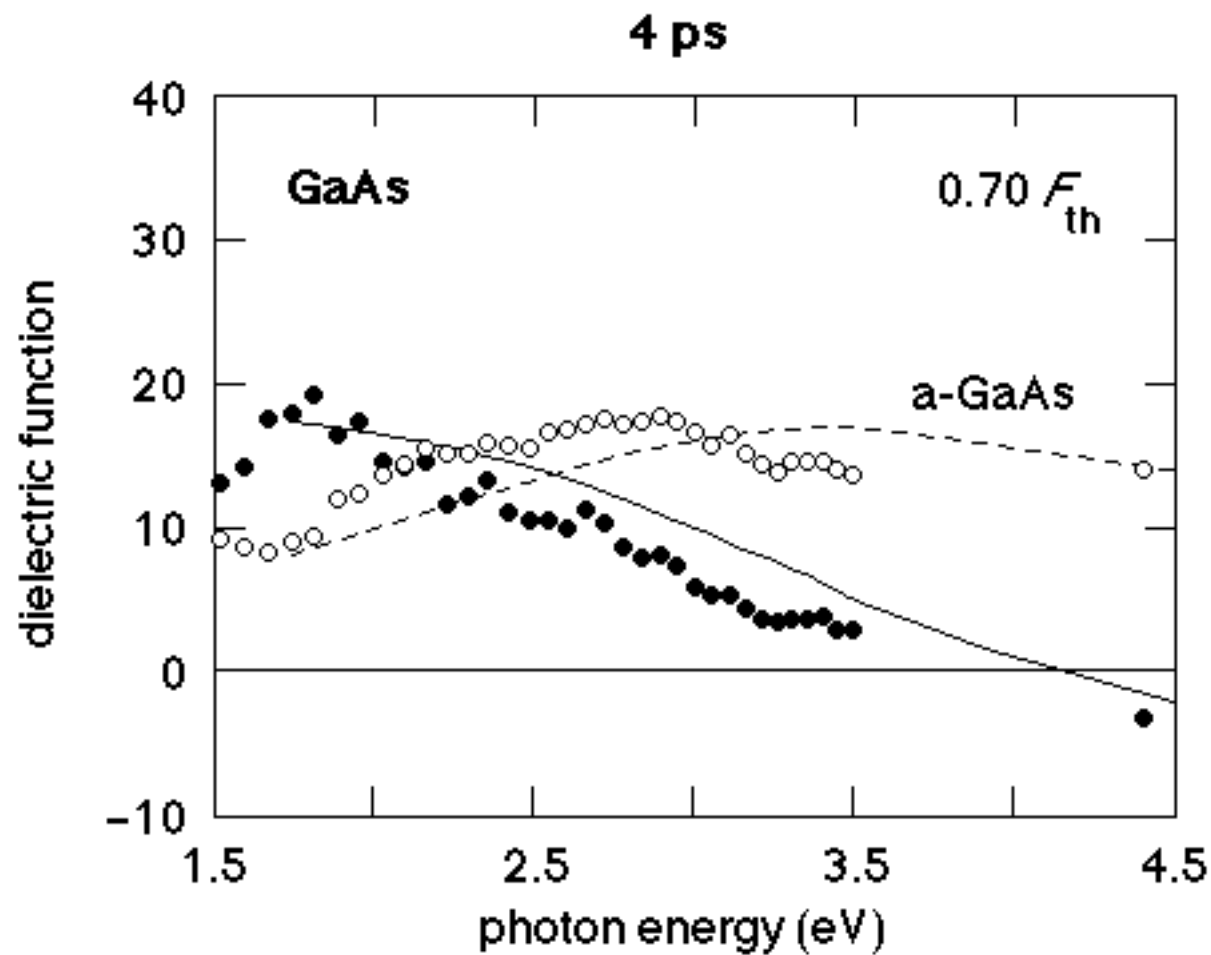
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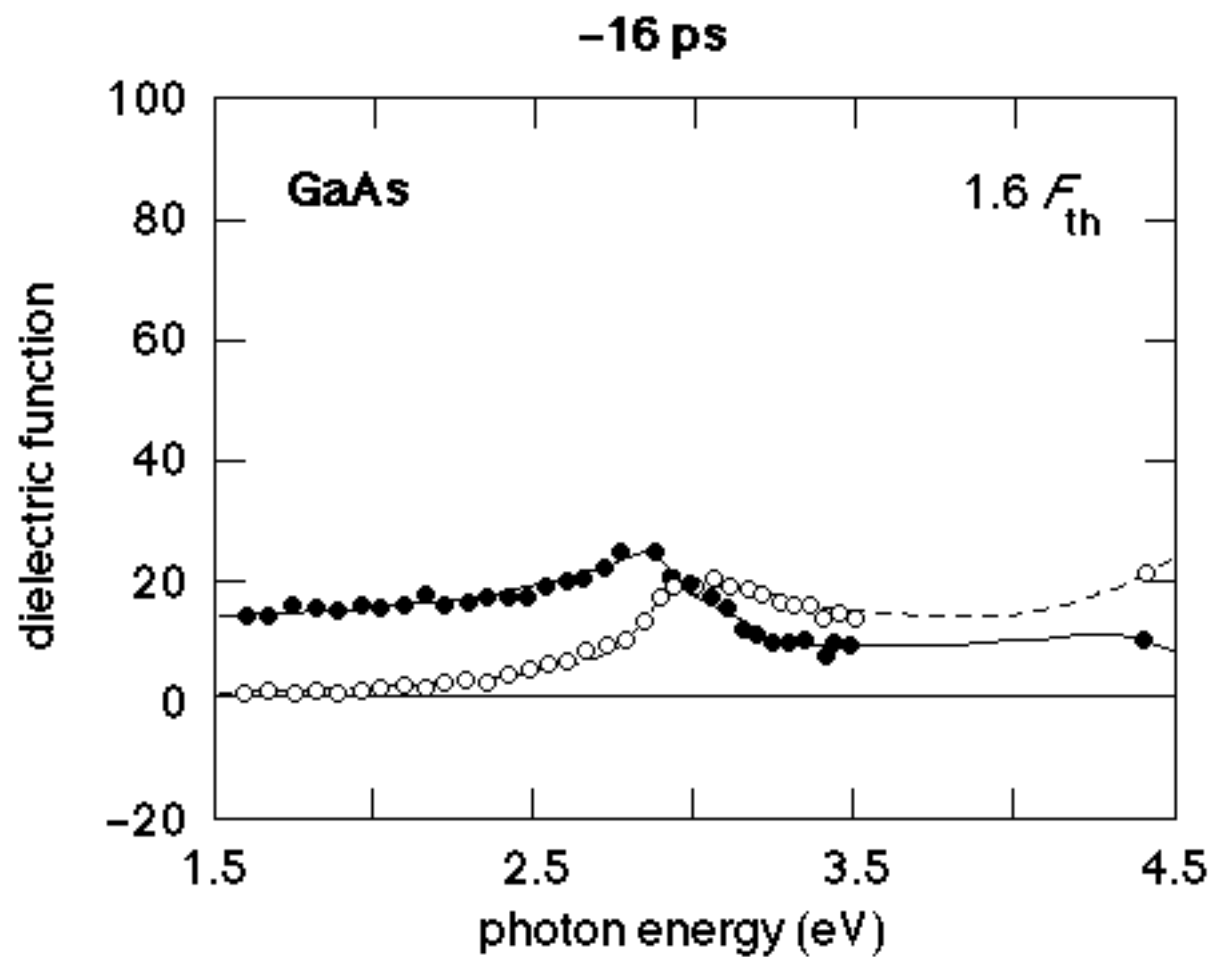
MEDIUM FLUENCE DATA



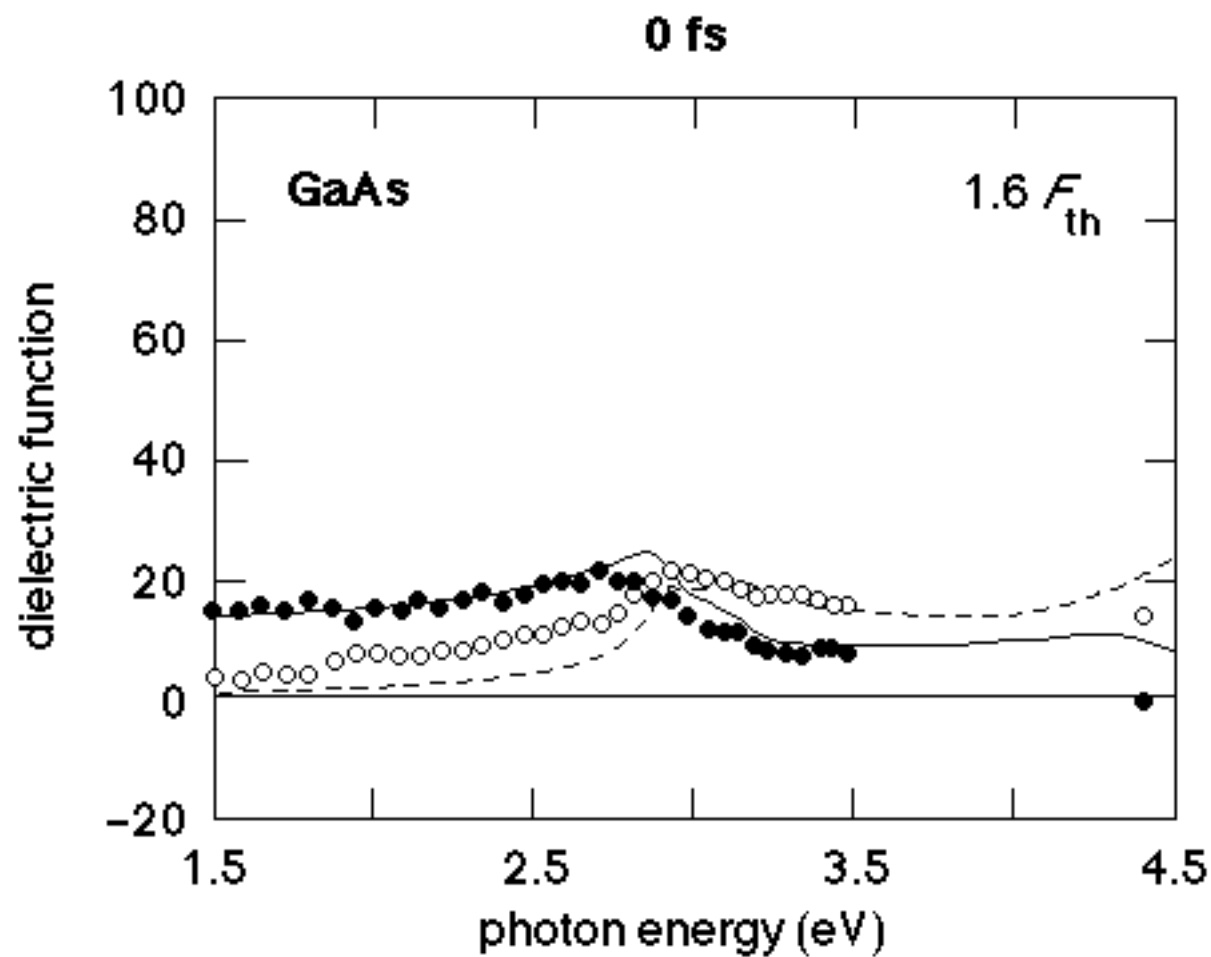
MEDIUM FLUENCE DATA



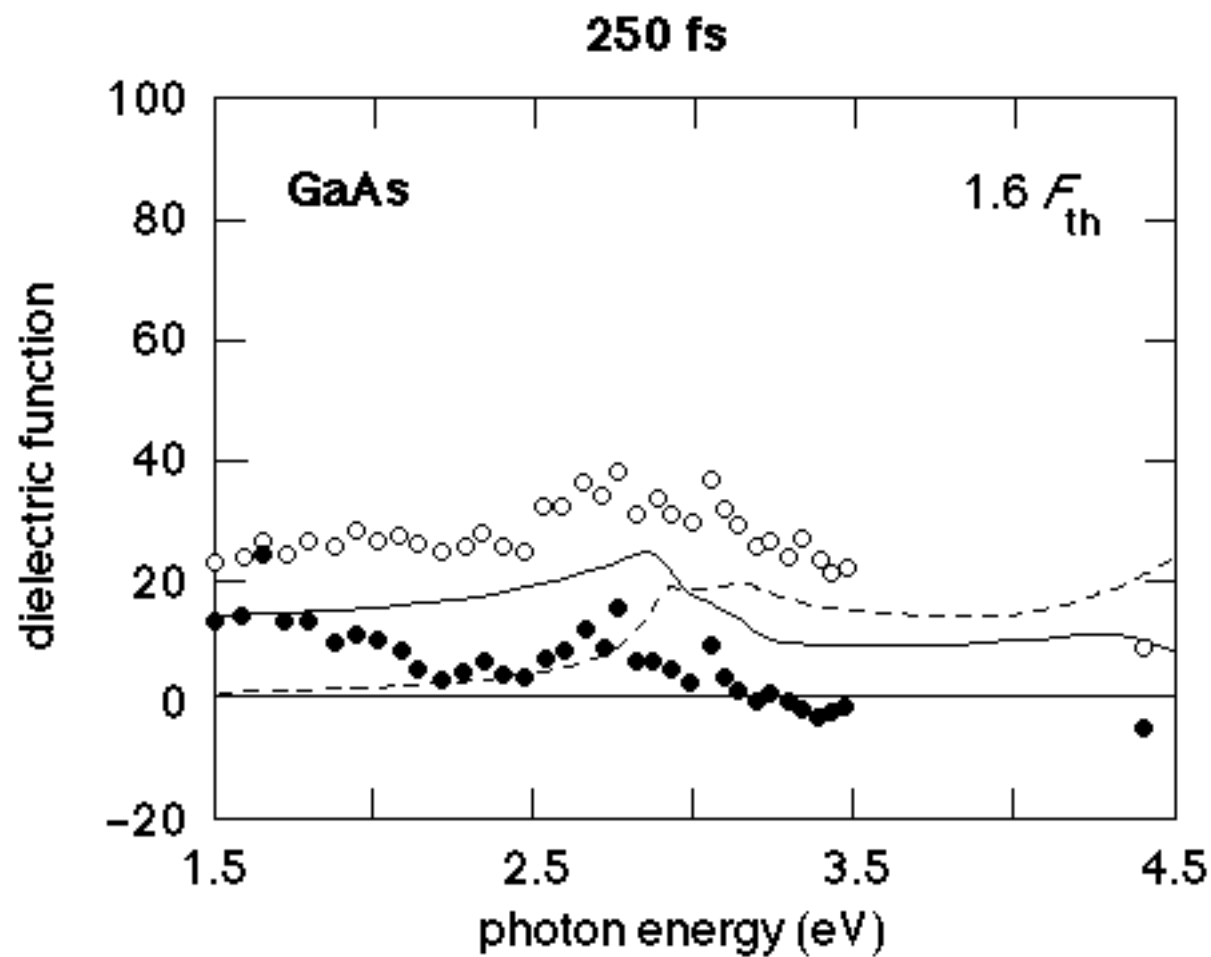
HIGH FLUENCE DATA



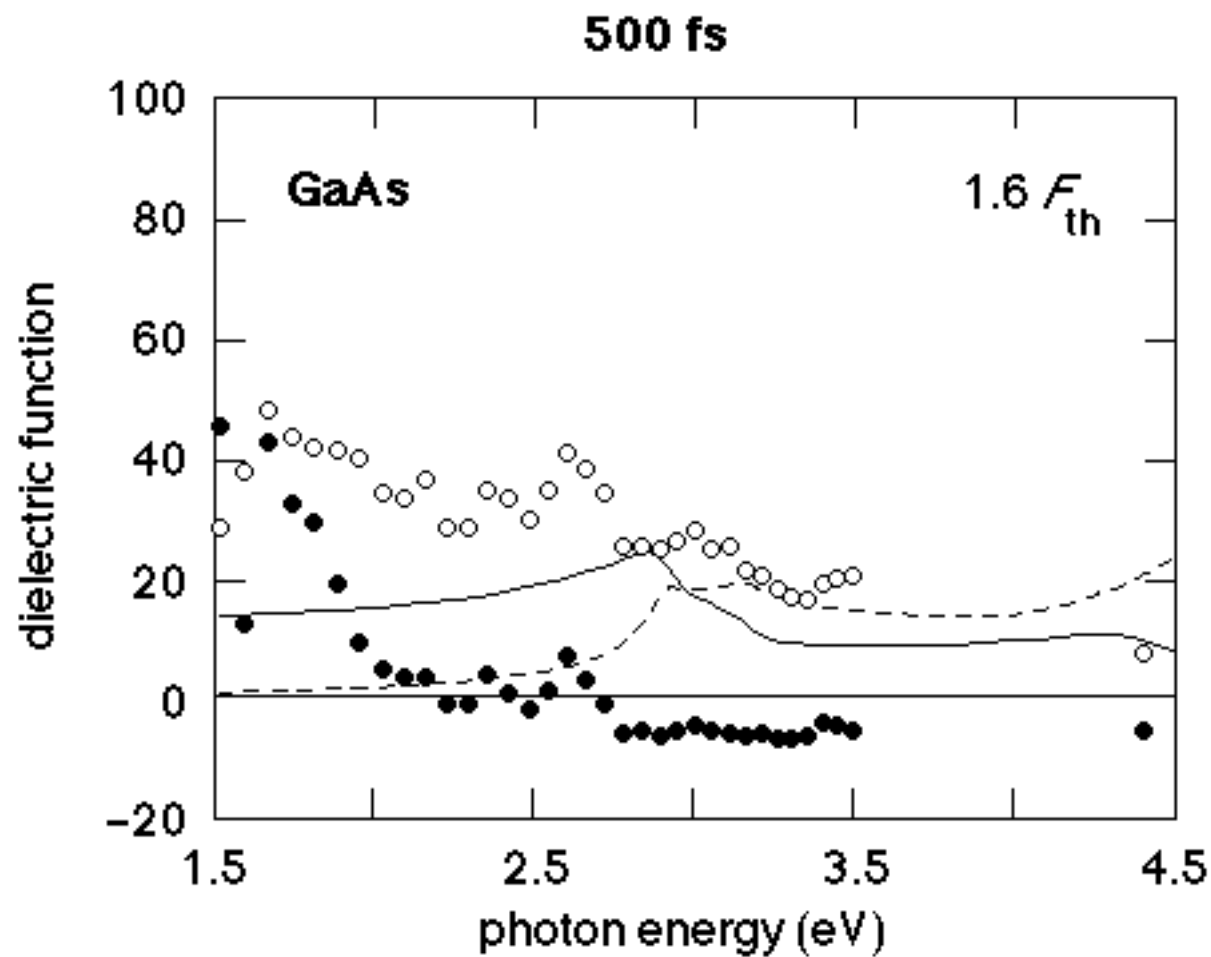
HIGH FLUENCE DATA



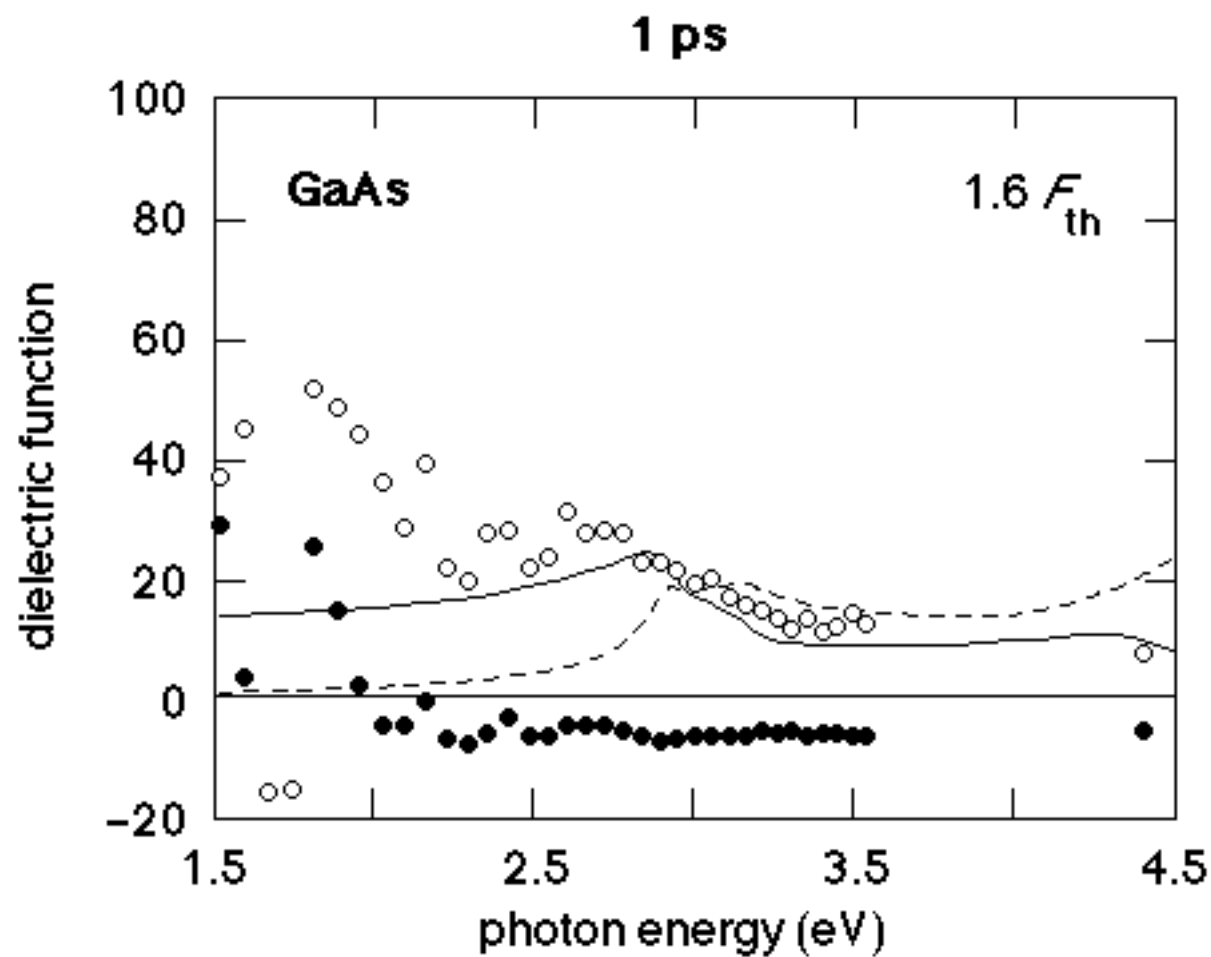
HIGH FLUENCE DATA



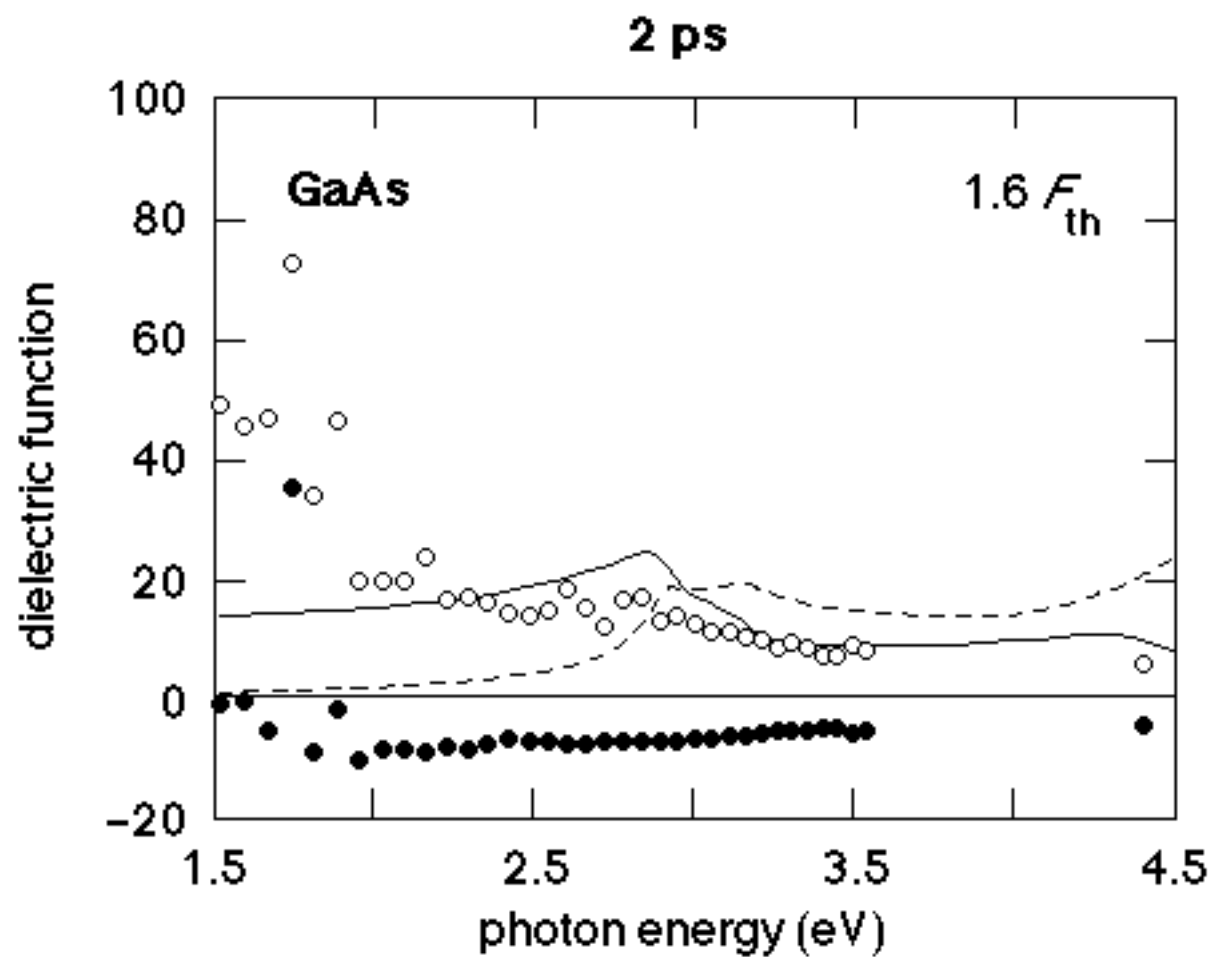
HIGH FLUENCE DATA



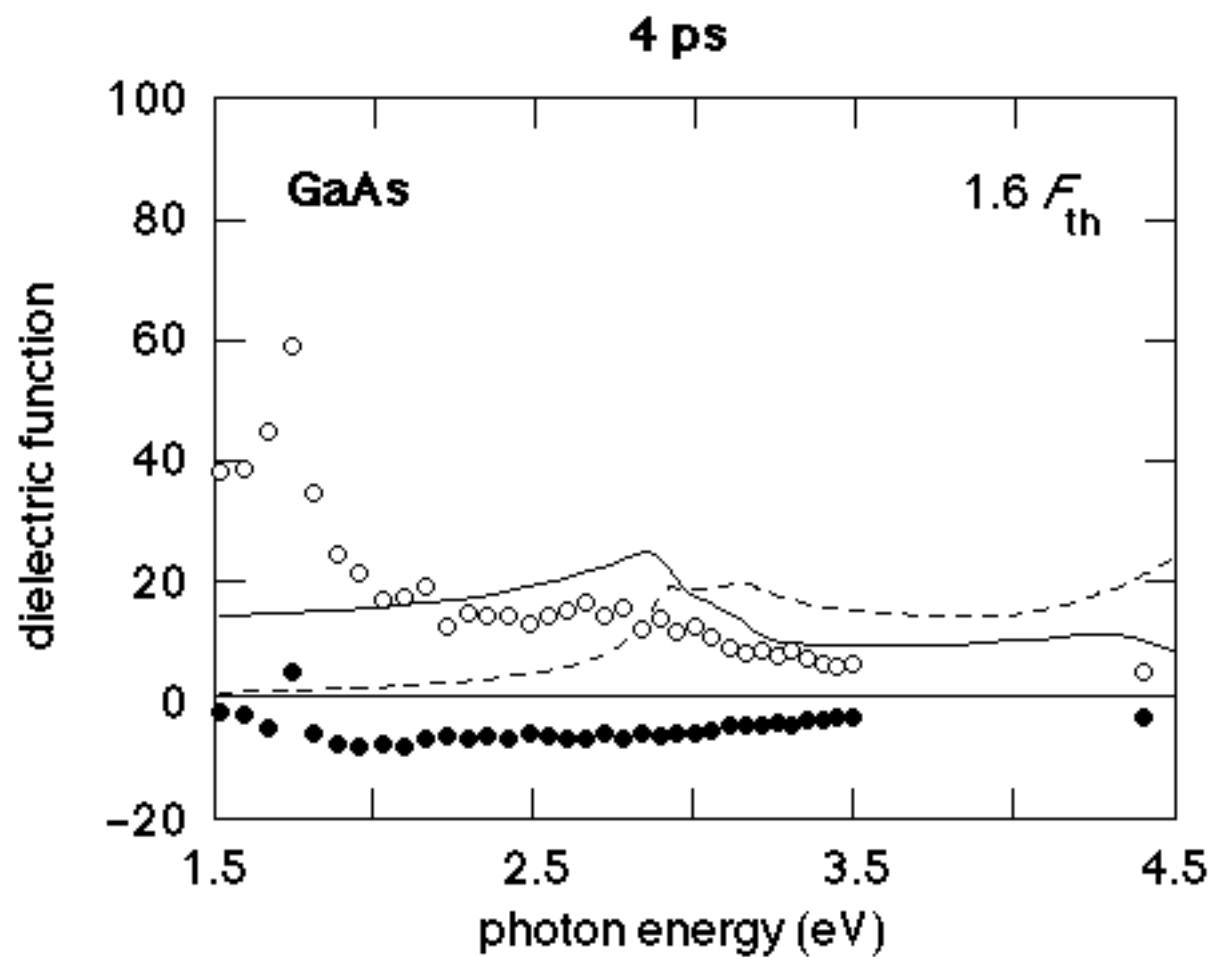
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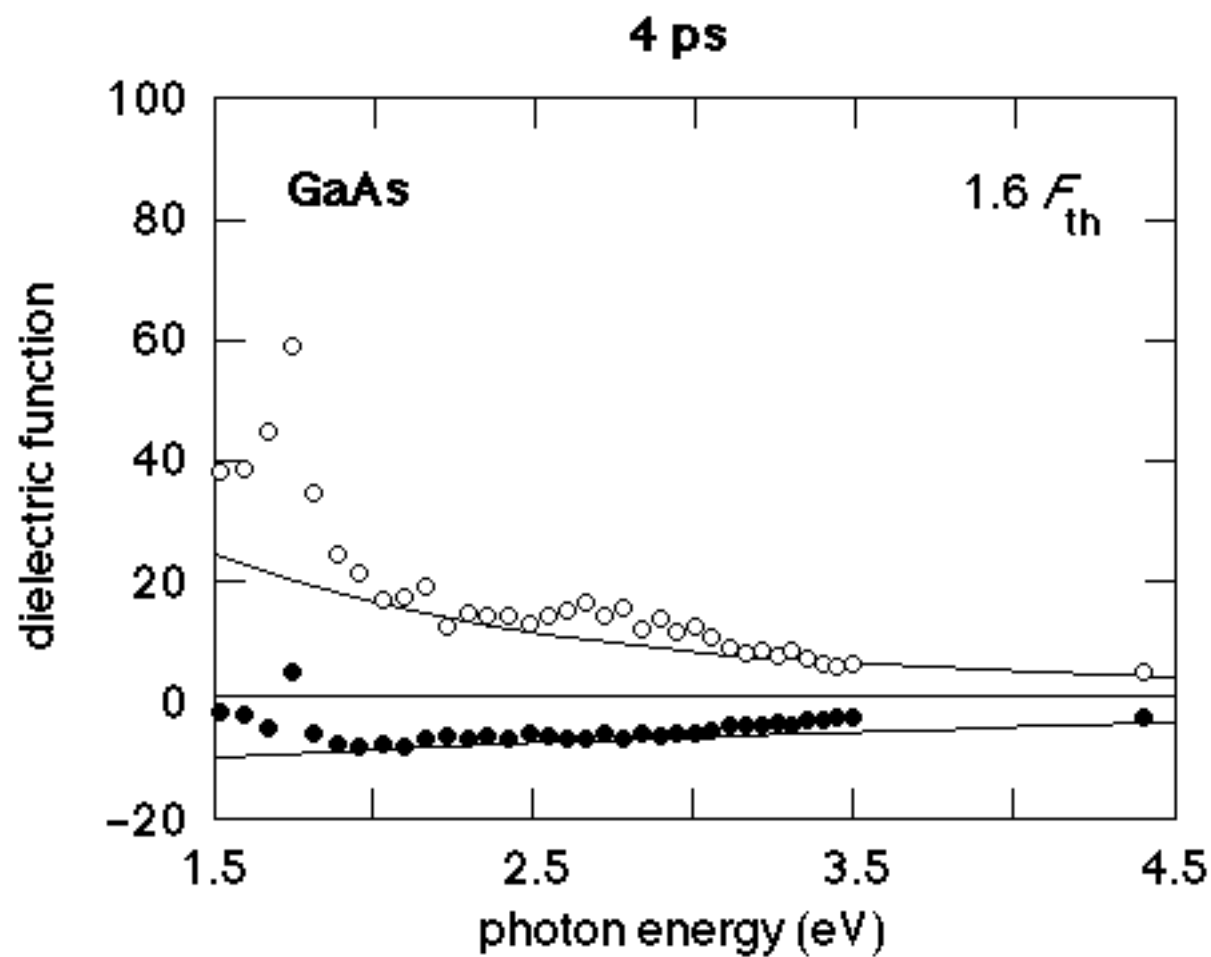
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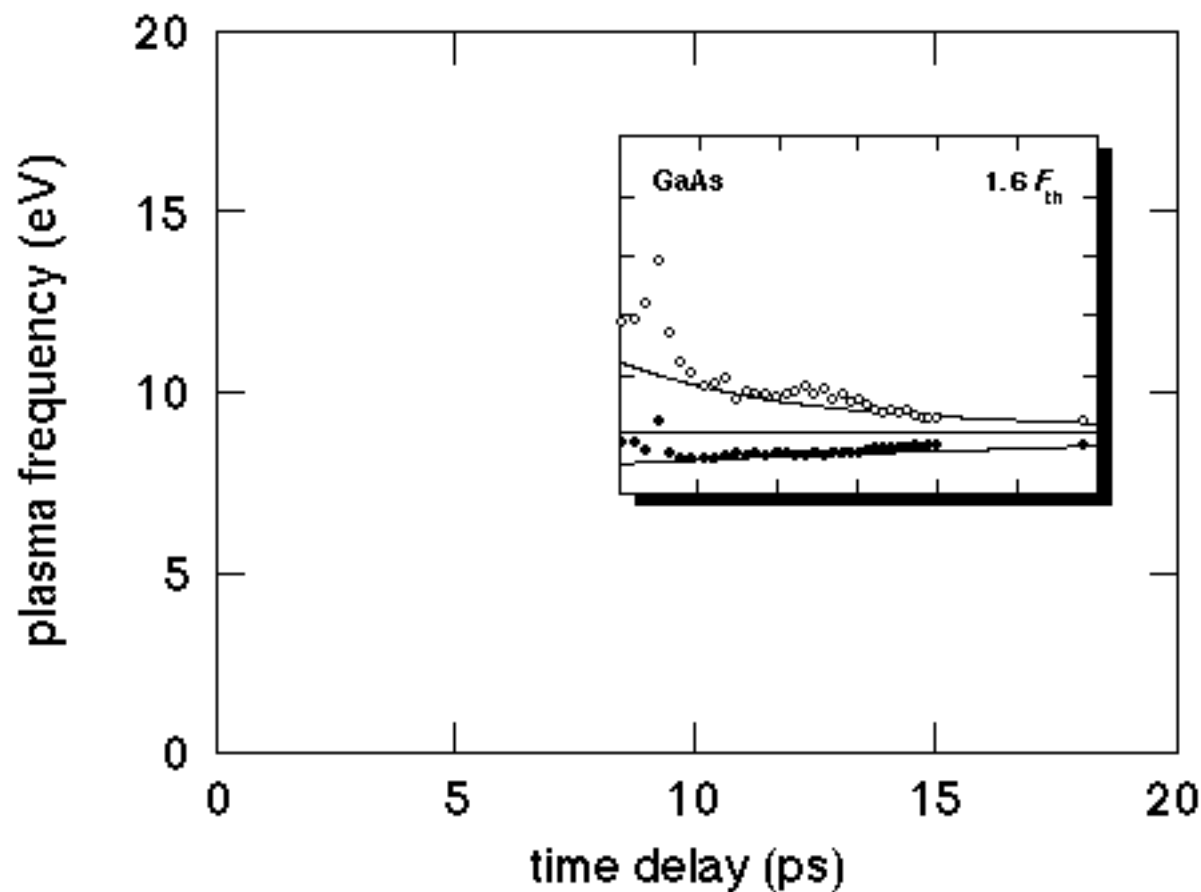
HIGH FLUENCE DATA



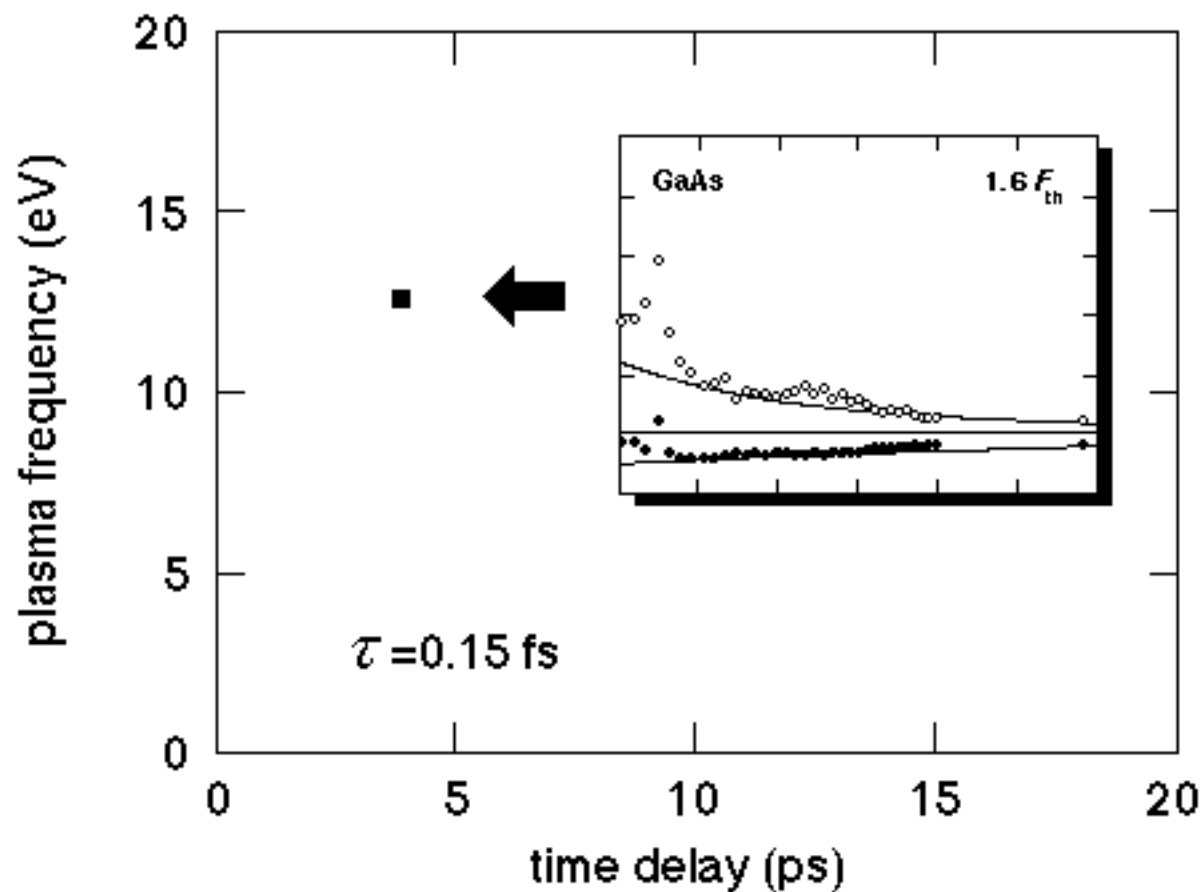
HIGH FLUENCE DATA



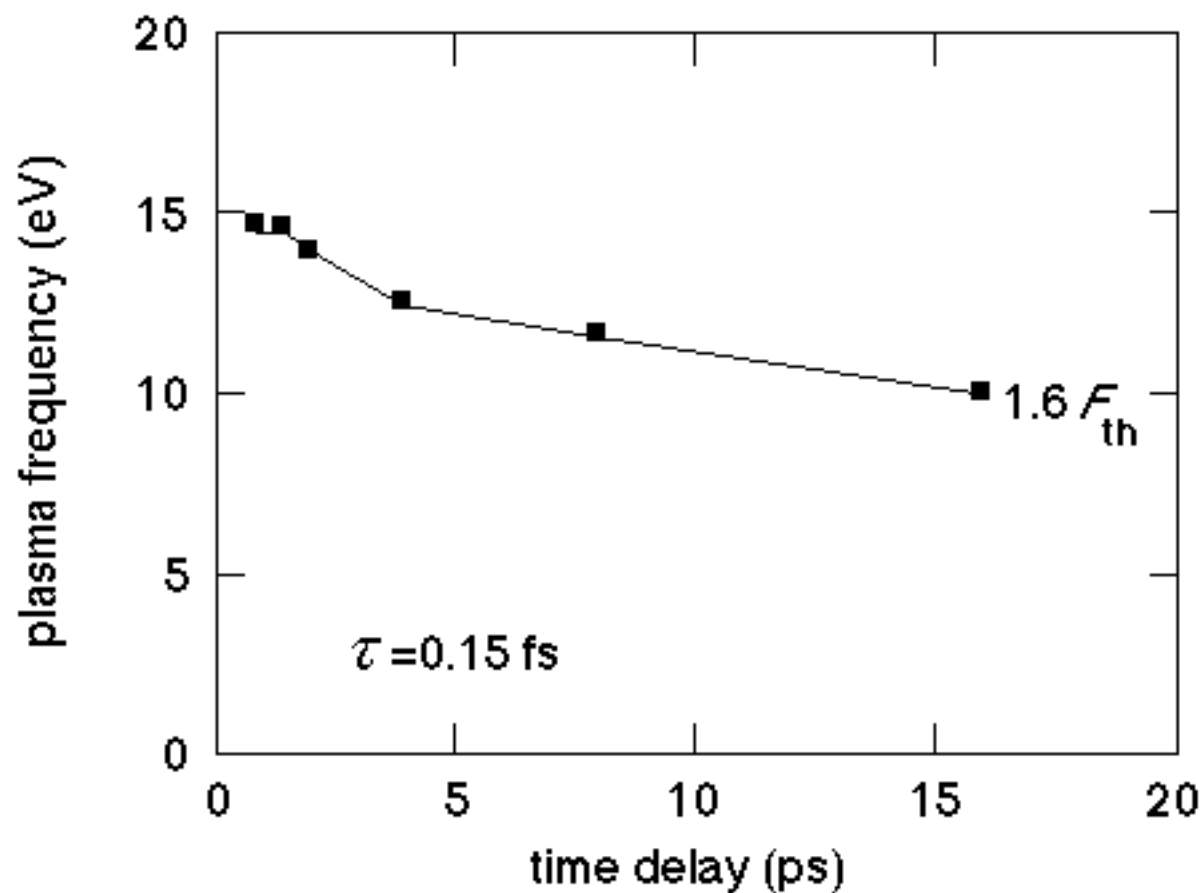
CARRIER DENSITY



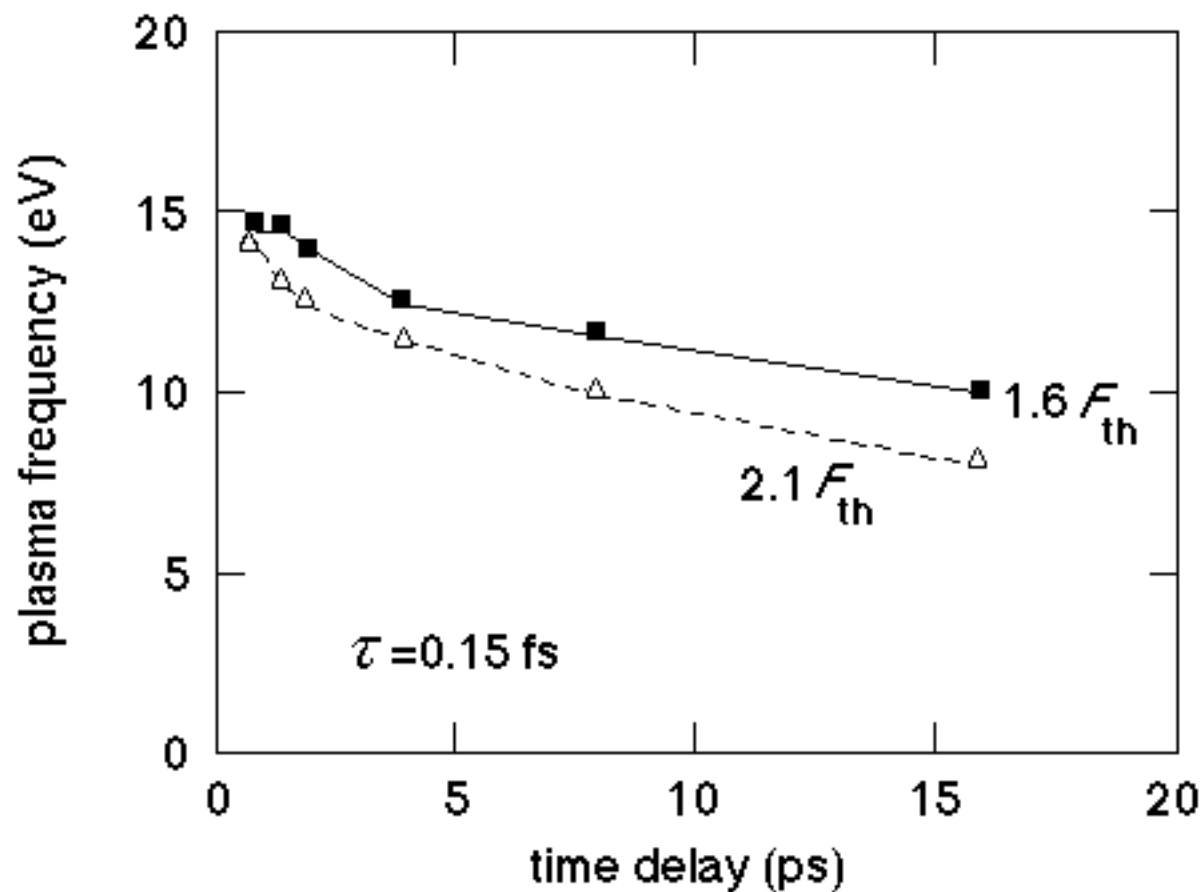
CARRIER DENSITY



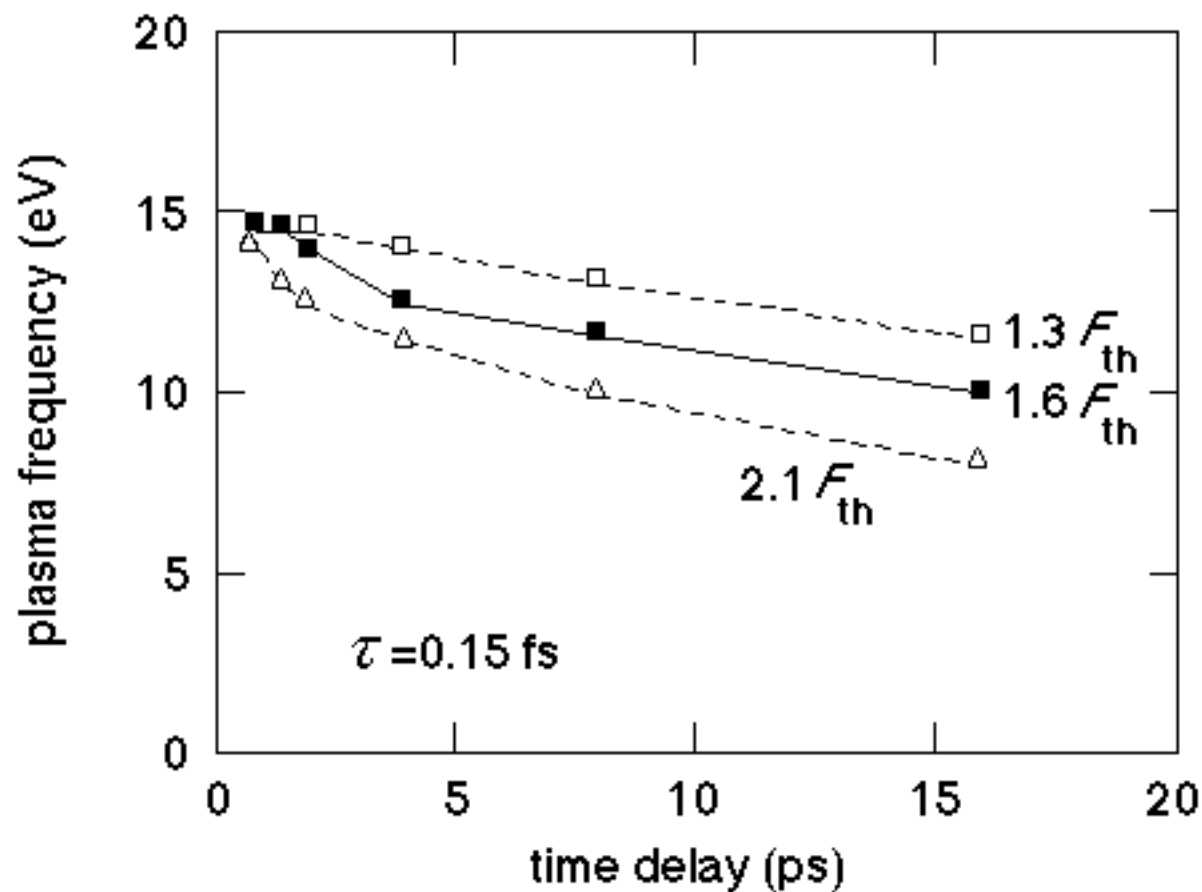
CARRIER DENSITY



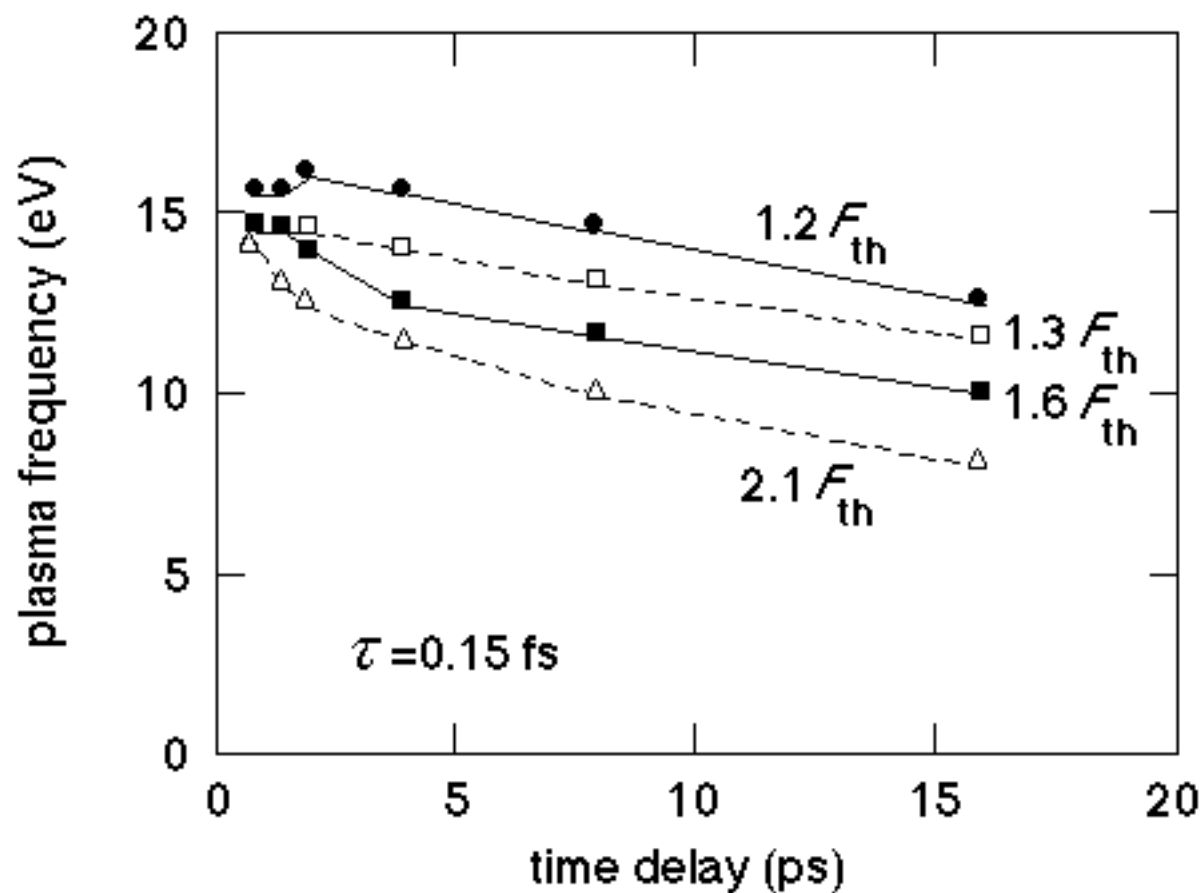
CARRIER DENSITY



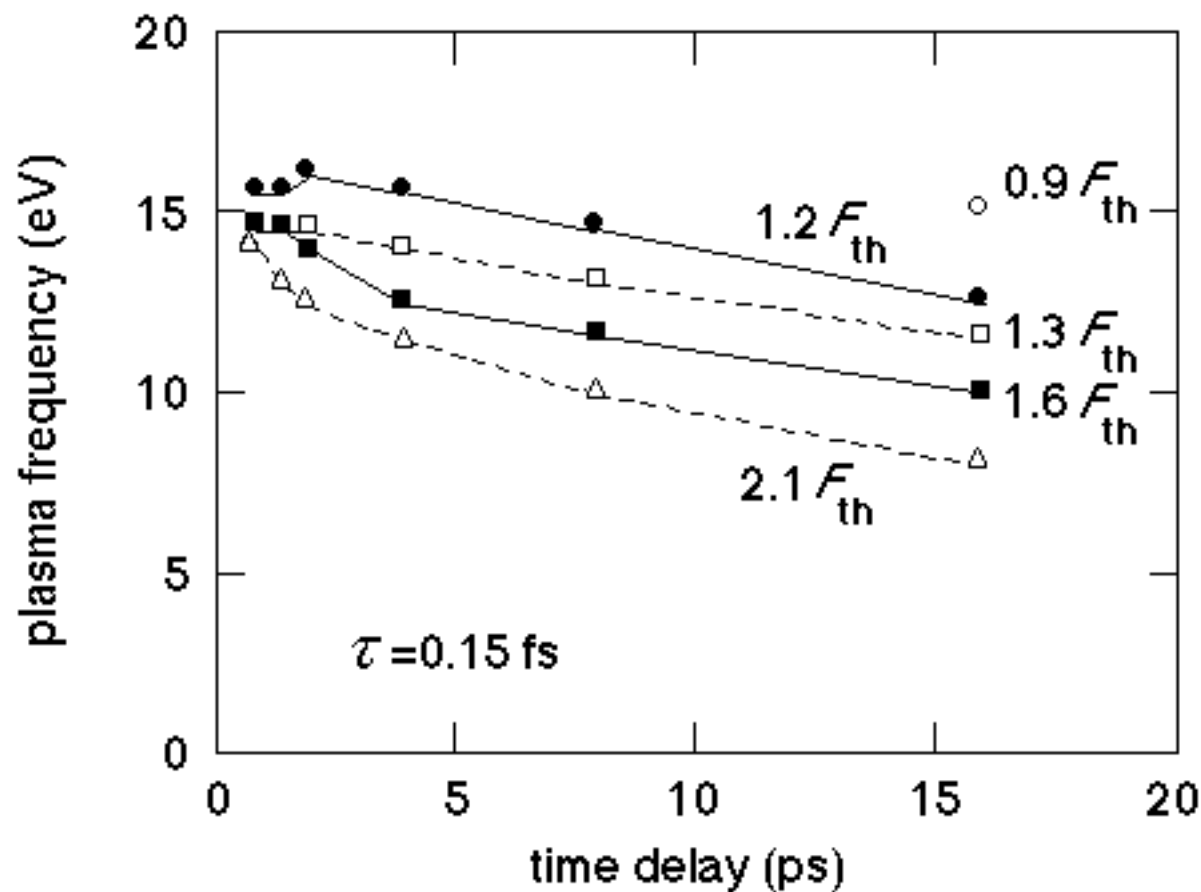
CARRIER DENSITY



CARRIER DENSITY



CARRIER DENSITY

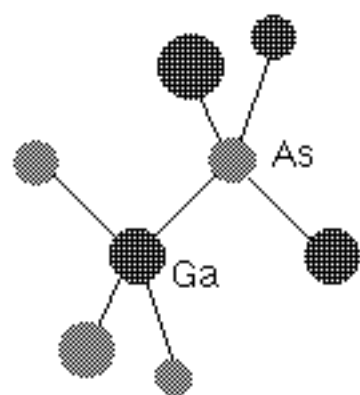


- ① Method
- ② Results
- ③ Discussion

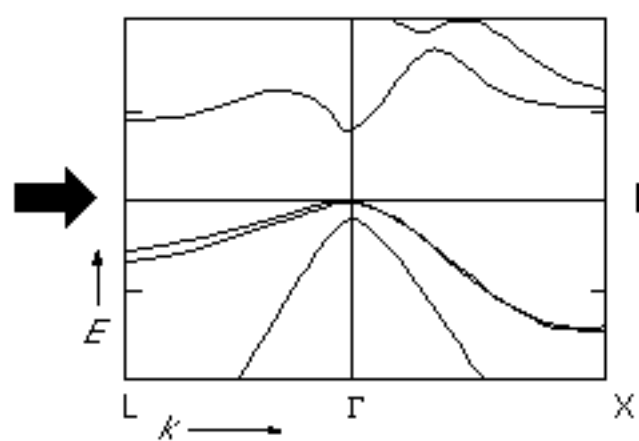


DISCUSSION

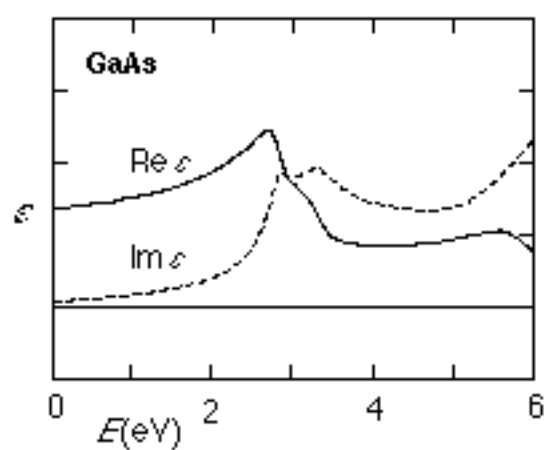
structure



bandstructure

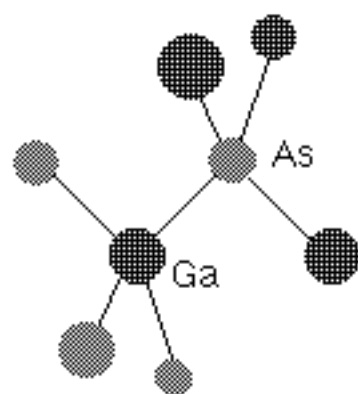


dielectric function

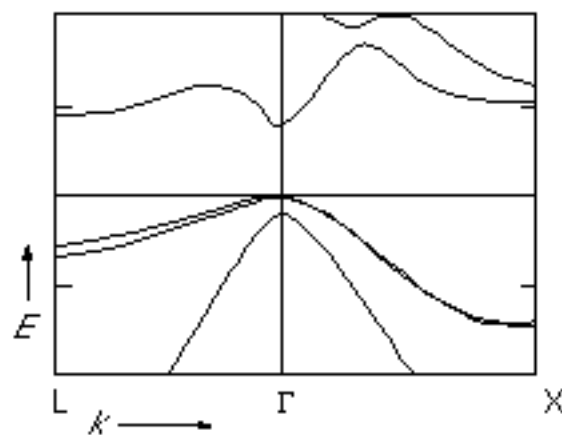


SHORT TIME SCALE (≈ 0.5 ps)

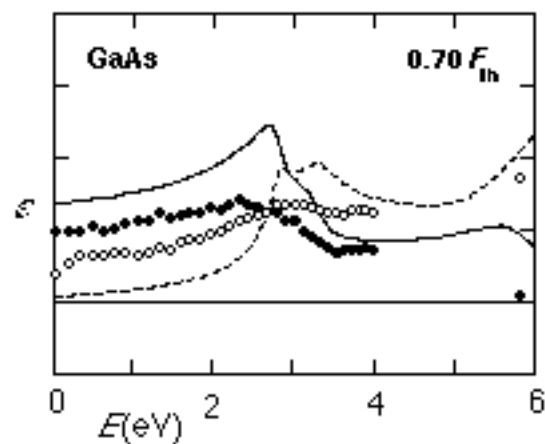
structure



bandstructure

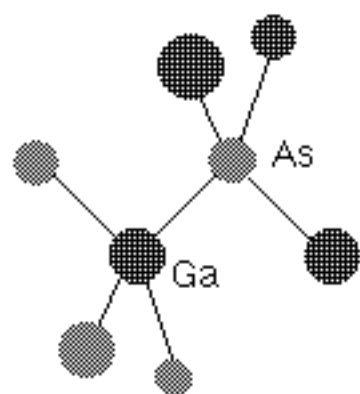


dielectric function

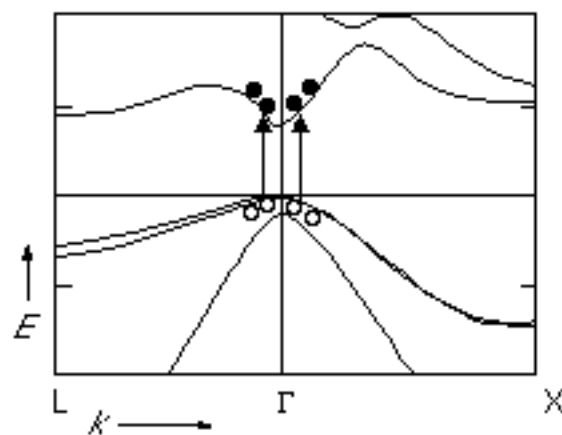


SHORT TIME SCALE (≈ 0.5 ps)

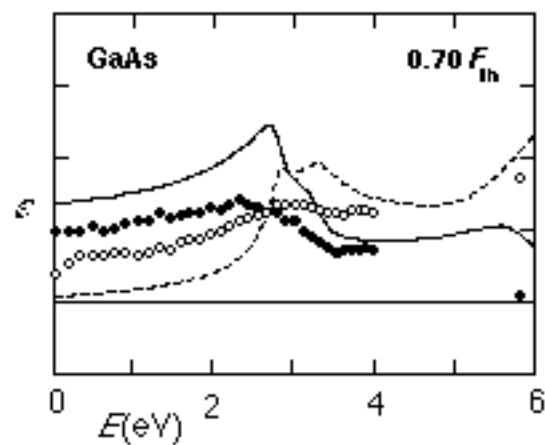
structure



bandstructure

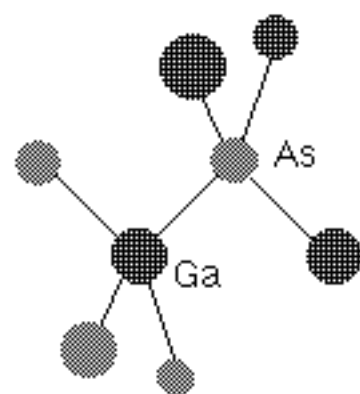


dielectric function

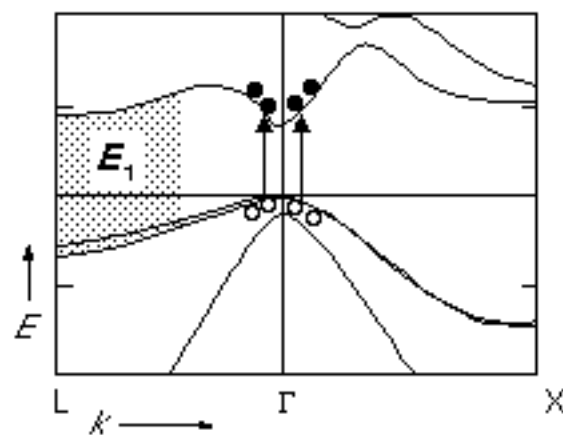


SHORT TIME SCALE (≈ 0.5 ps)

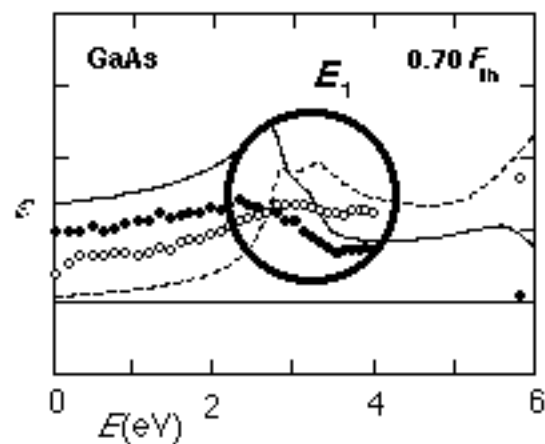
structure



bandstructure

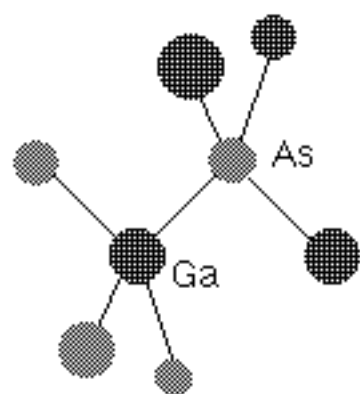


dielectric function

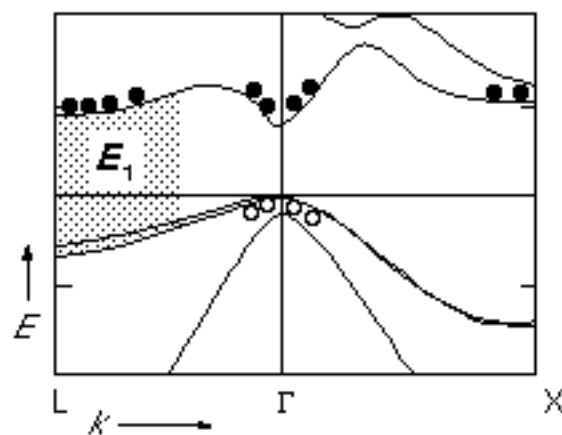


SHORT TIME SCALE (≈ 0.5 ps)

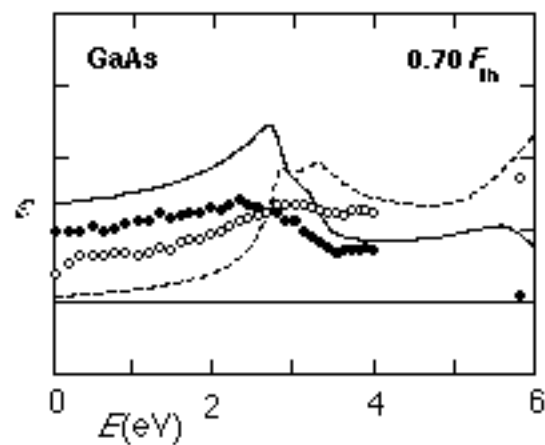
structure



bandstructure

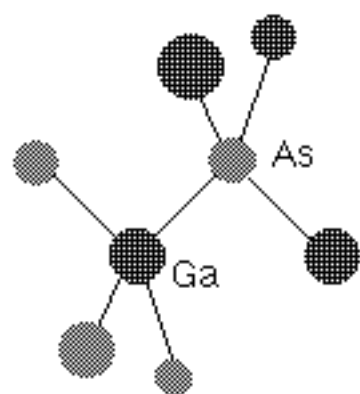


dielectric function

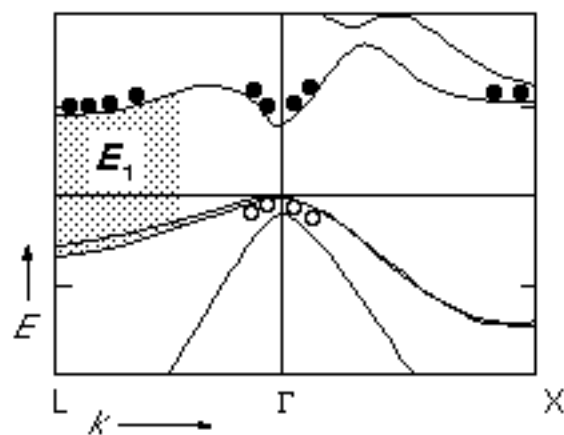


SHORT TIME SCALE (≈ 0.5 ps)

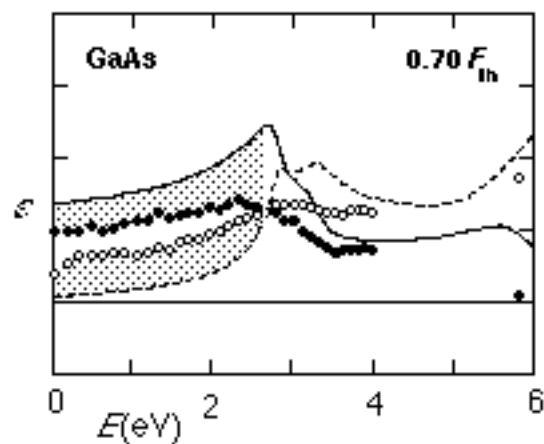
structure



bandstructure

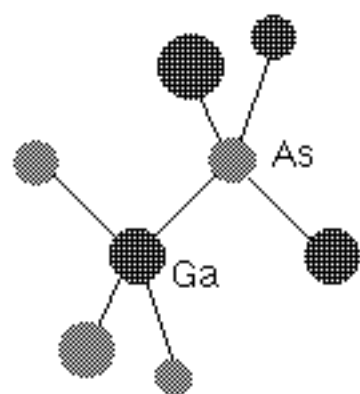


dielectric function

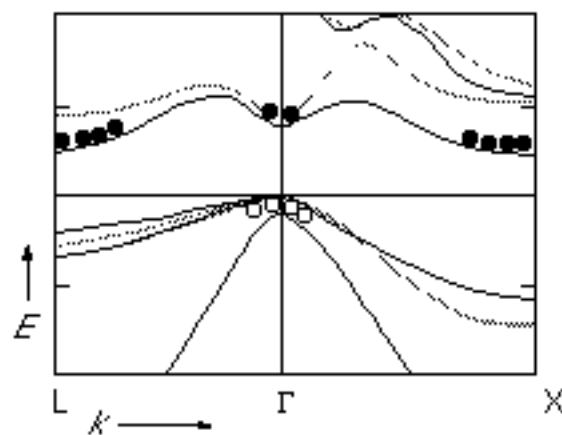


SHORT TIME SCALE (≈ 0.5 ps)

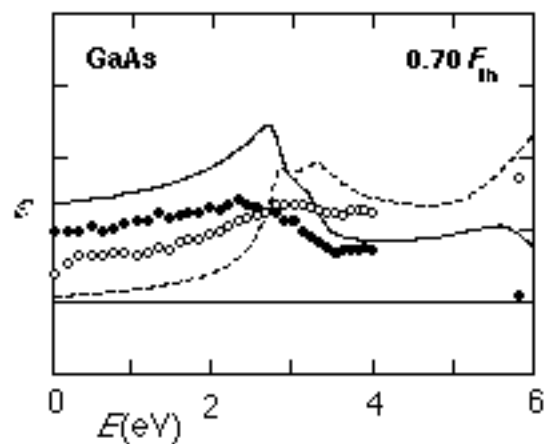
structure



bandstructure

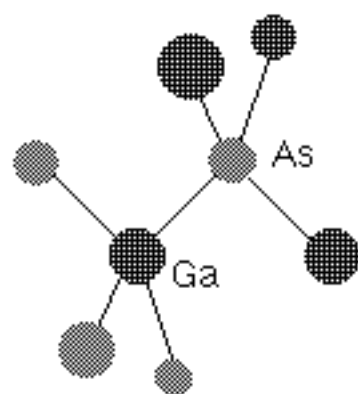


dielectric function

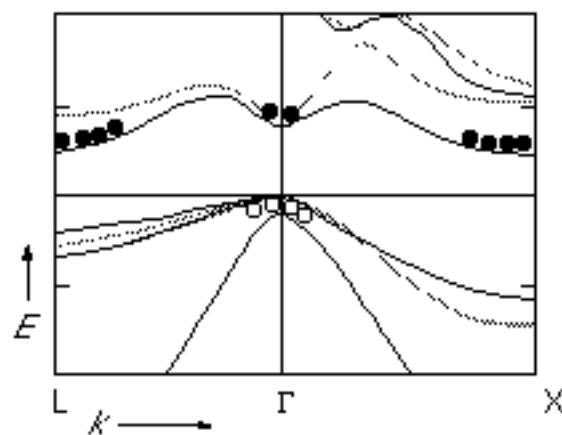


SHORT TIME SCALE (≈ 0.5 ps)

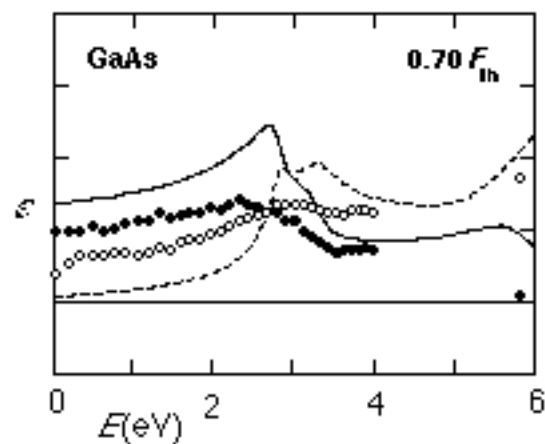
structure



bandstructure



dielectric function

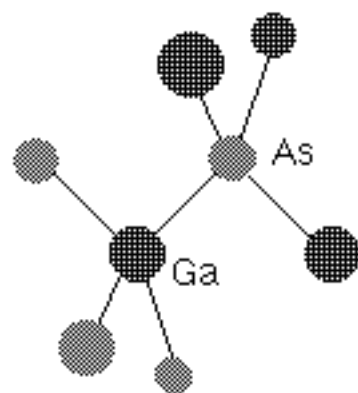


► electronic effects dominate at short time scales...

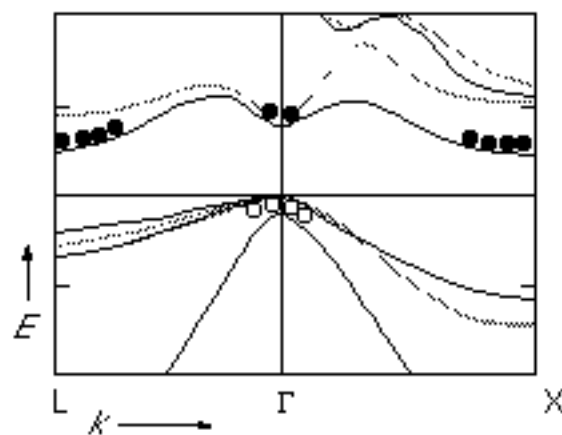


SHORT TIME SCALE (≈ 0.5 ps)

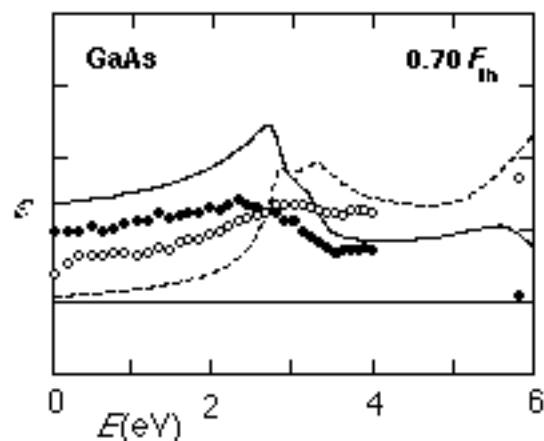
structure



bandstructure



dielectric function

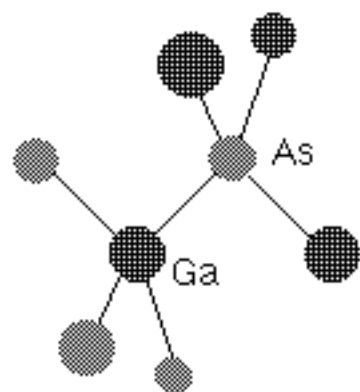


► ...but they are not as simple as we used to think

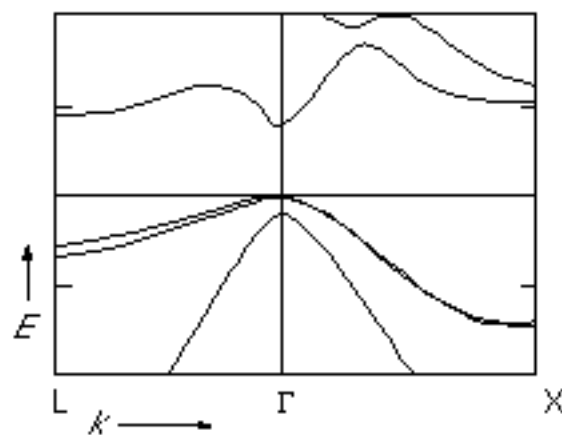


LOW FLUENCE, LONG TIME SCALE

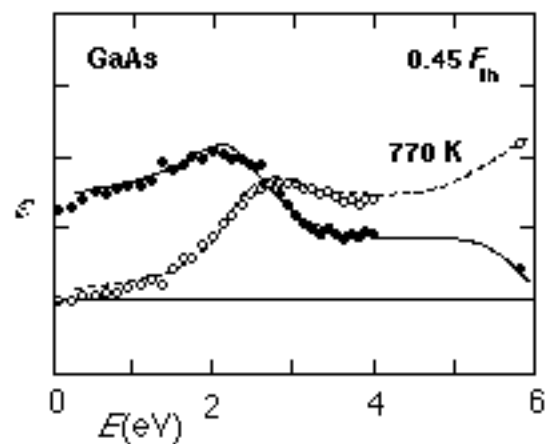
structure



bandstructure

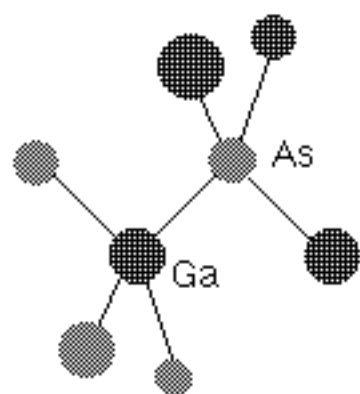


dielectric function

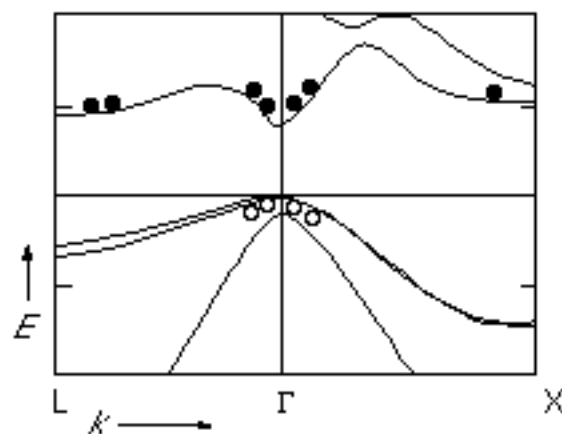


LOW FLUENCE, LONG TIME SCALE

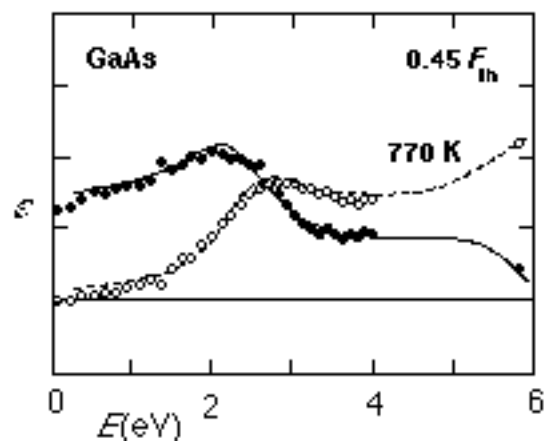
structure



bandstructure



dielectric function

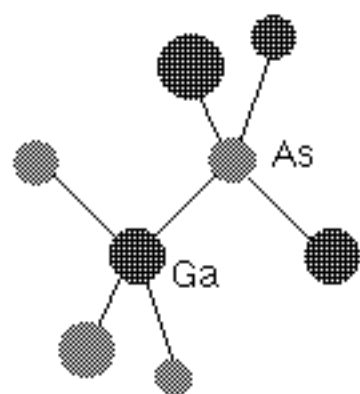


carrier density down, electronic effects subsided...

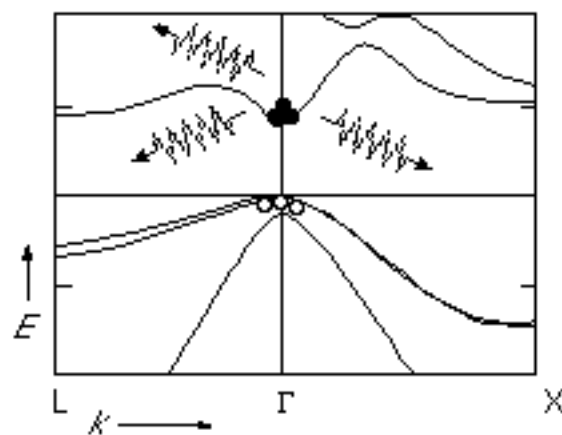


LOW FLUENCE, LONG TIME SCALE

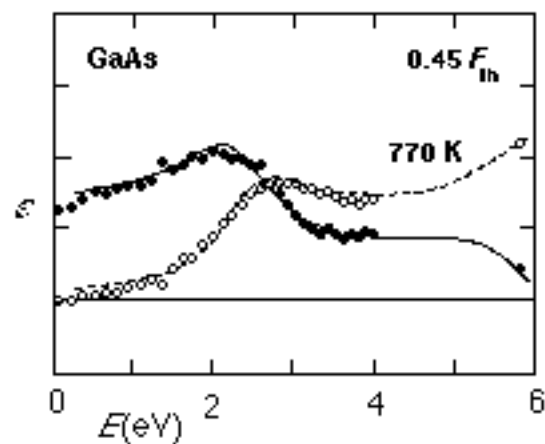
structure



bandstructure



dielectric function

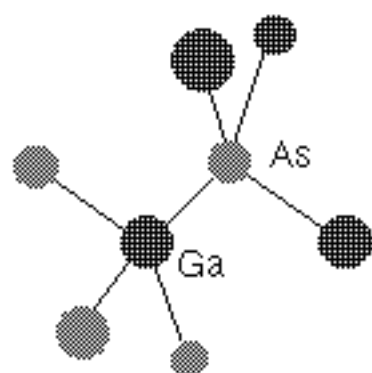


...and lattice heats due to carrier relaxation

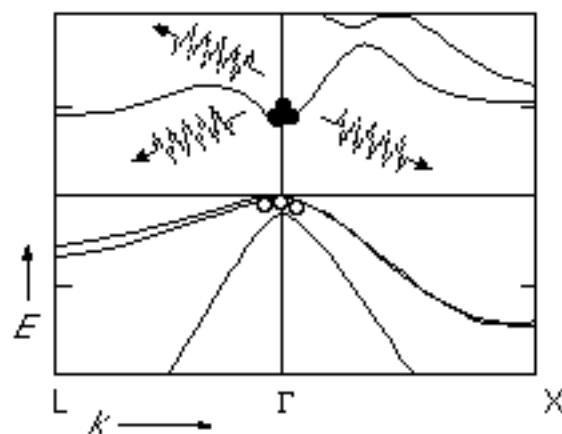


LOW FLUENCE, LONG TIME SCALE

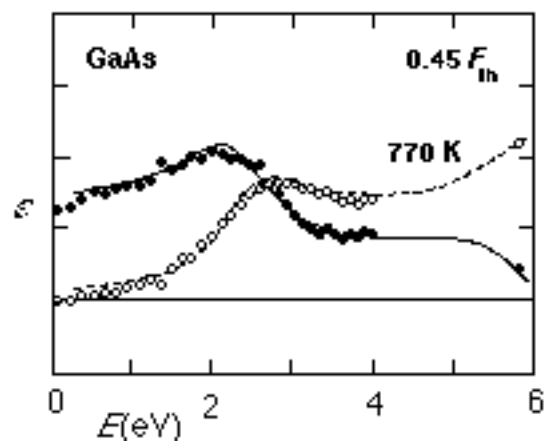
structure



bandstructure



dielectric function

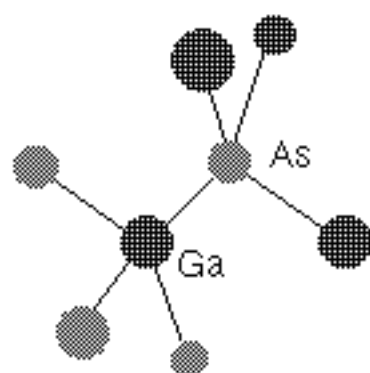


...and lattice heats due to carrier relaxation

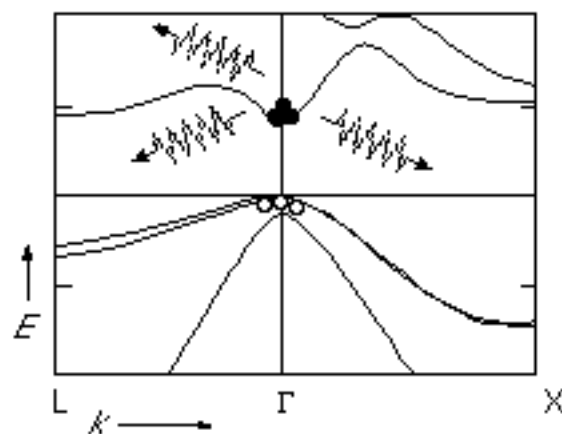


LOW FLUENCE, LONG TIME SCALE

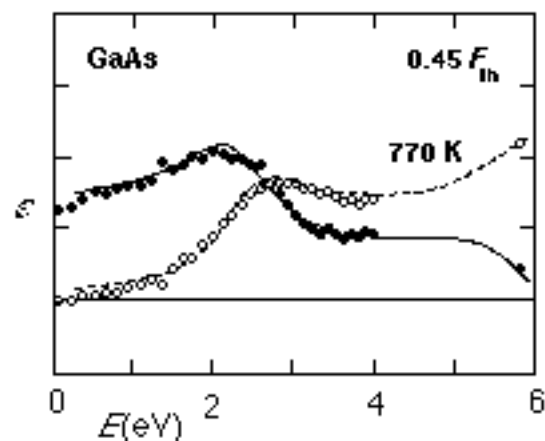
structure



bandstructure



dielectric function

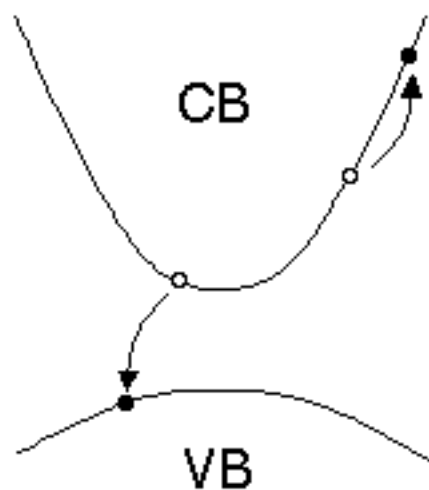


But... why do electronic effects disappear after 2 ps while lattice heats in 7 ps?



LOW FLUENCE, LONG TIME SCALE

Auger recombination

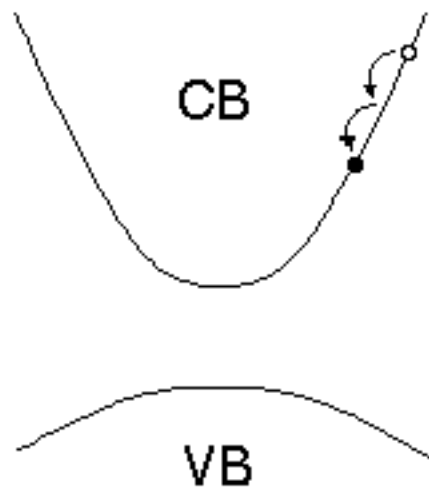


Auger lowers N without changing E in 2–4 ps...



LOW FLUENCE, LONG TIME SCALE

carrier-phonon scattering

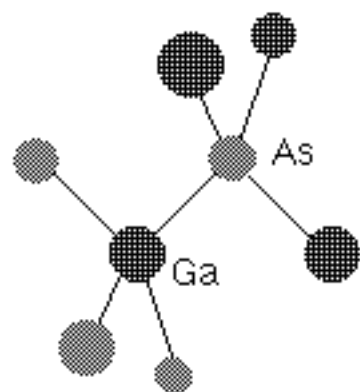


... and highly excited electrons cool to lattice in 7 ps

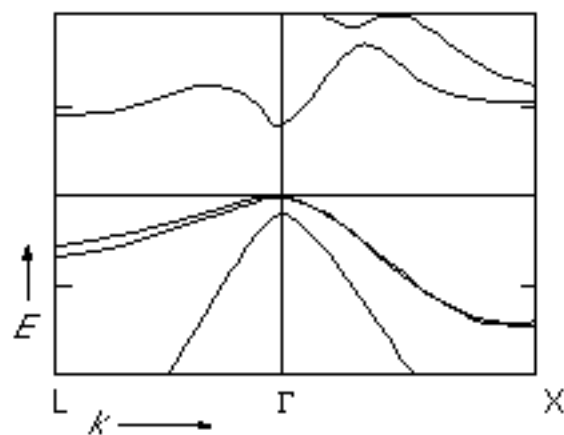


HIGH FLUENCE, LONG TIME SCALE

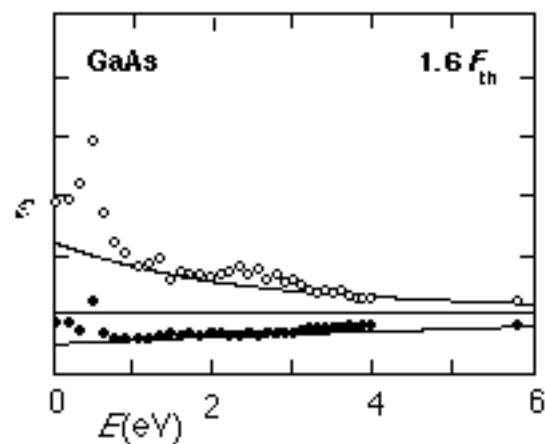
structure



bandstructure

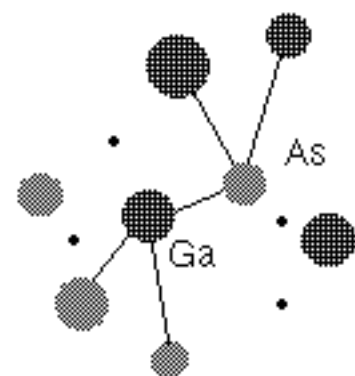


dielectric function

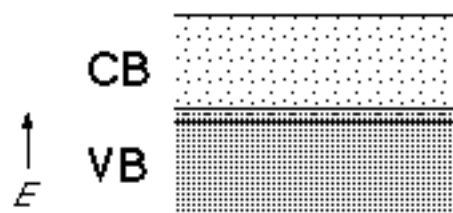


HIGH FLUENCE, LONG TIME SCALE

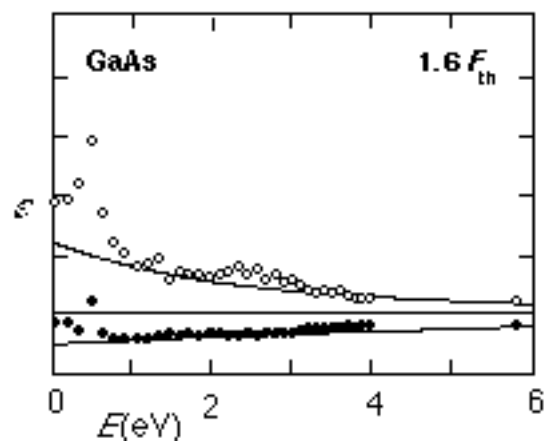
structure



bandstructure



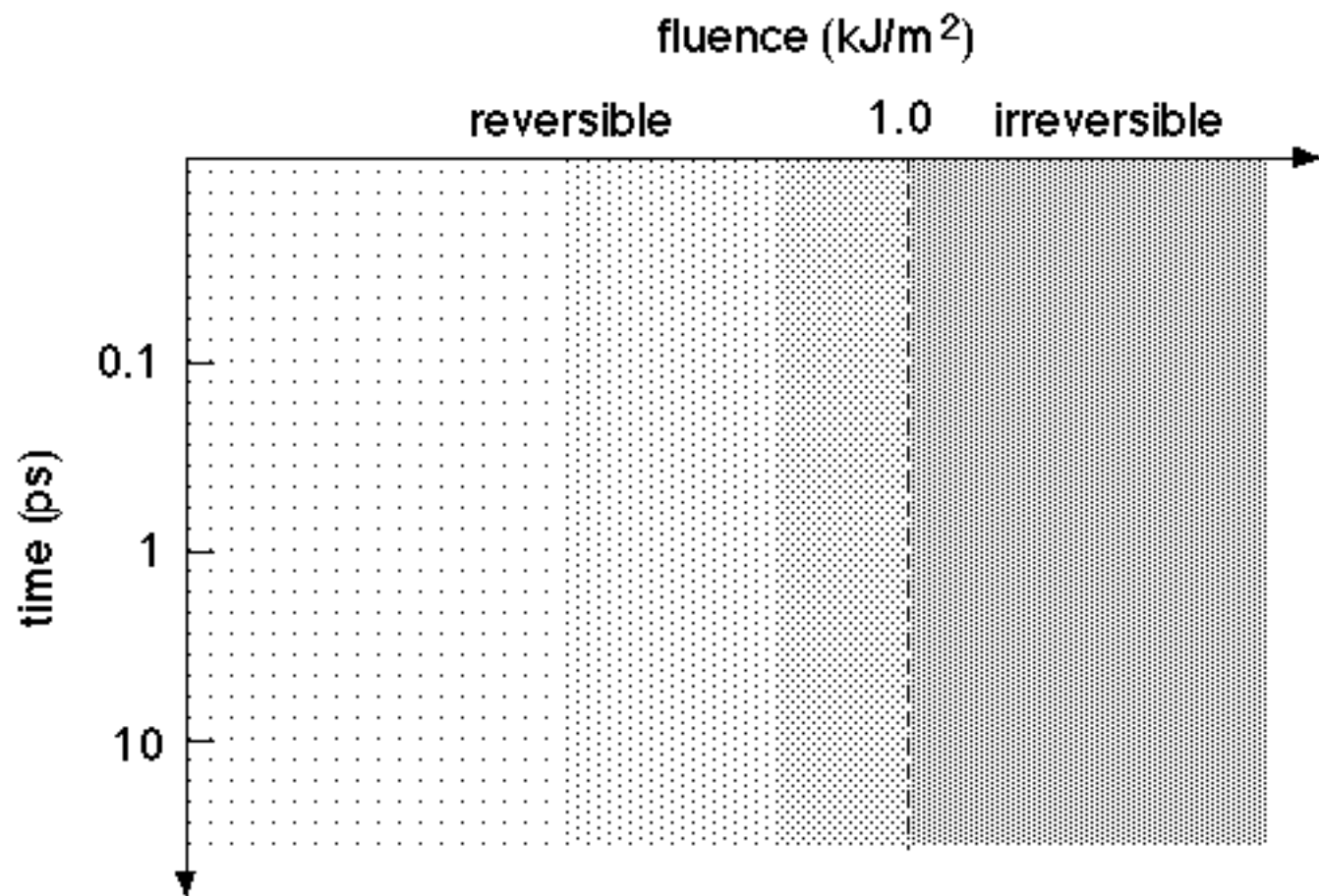
dielectric function



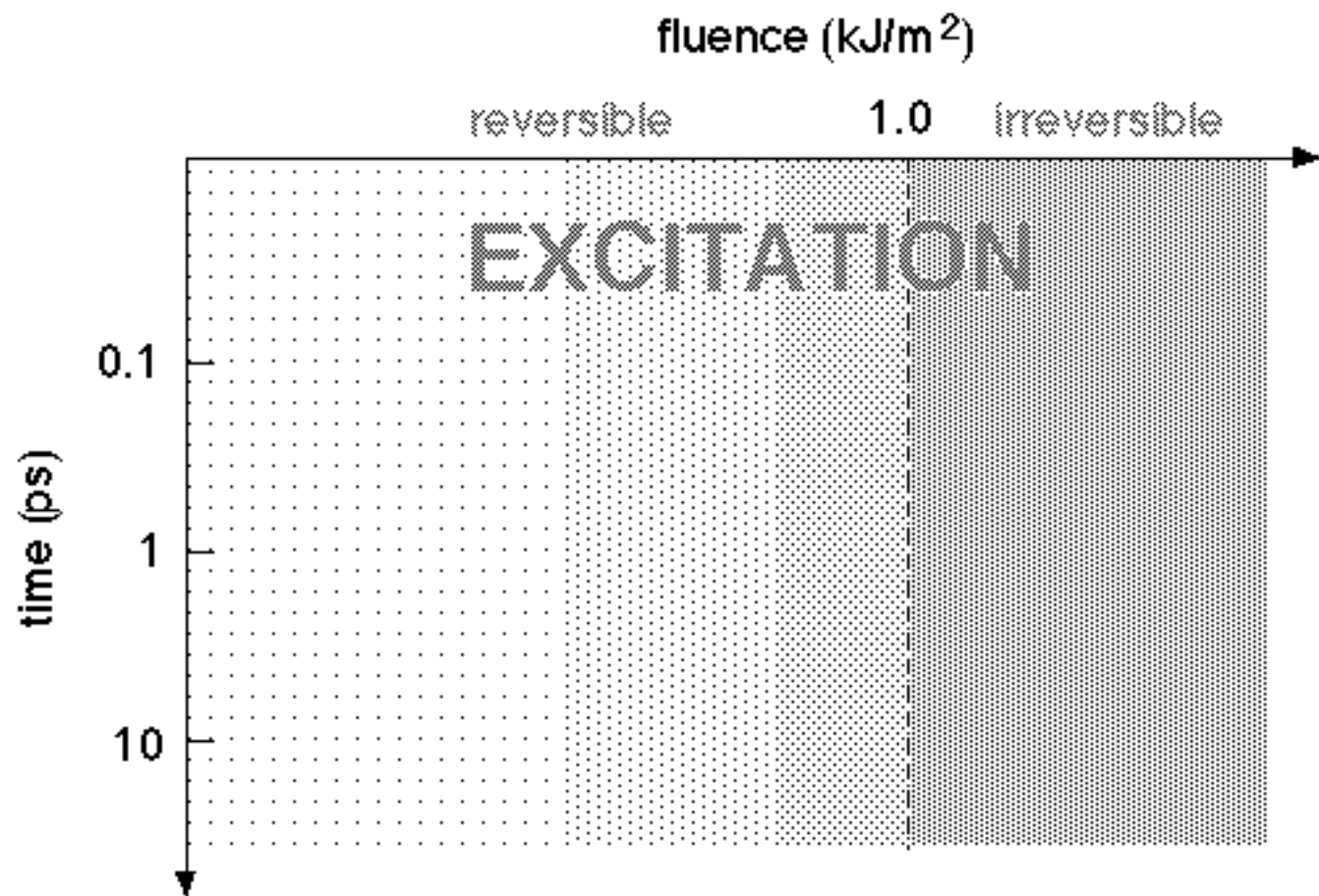
gradual drop in gap \rightarrow not electronic effect



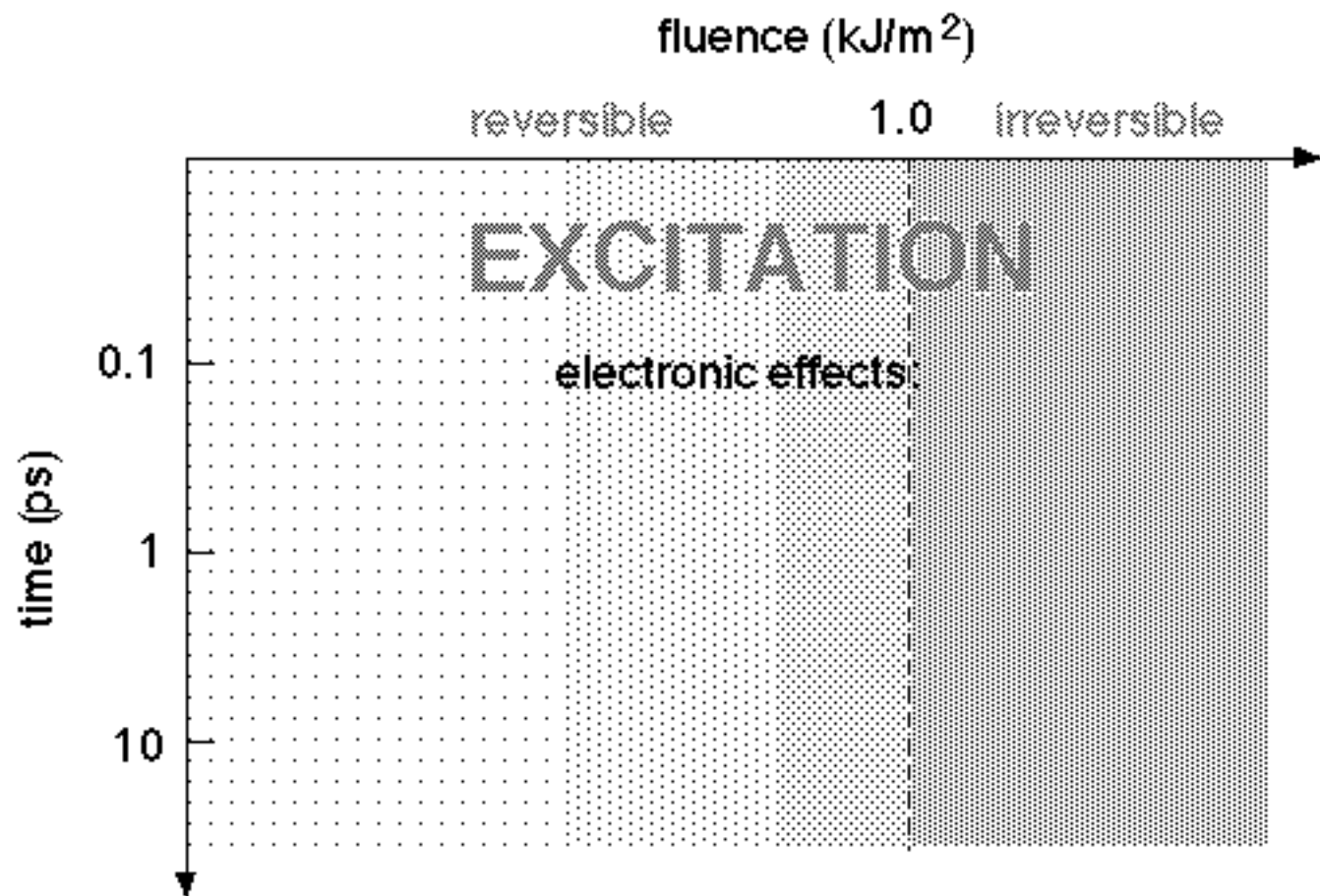
OVERVIEW



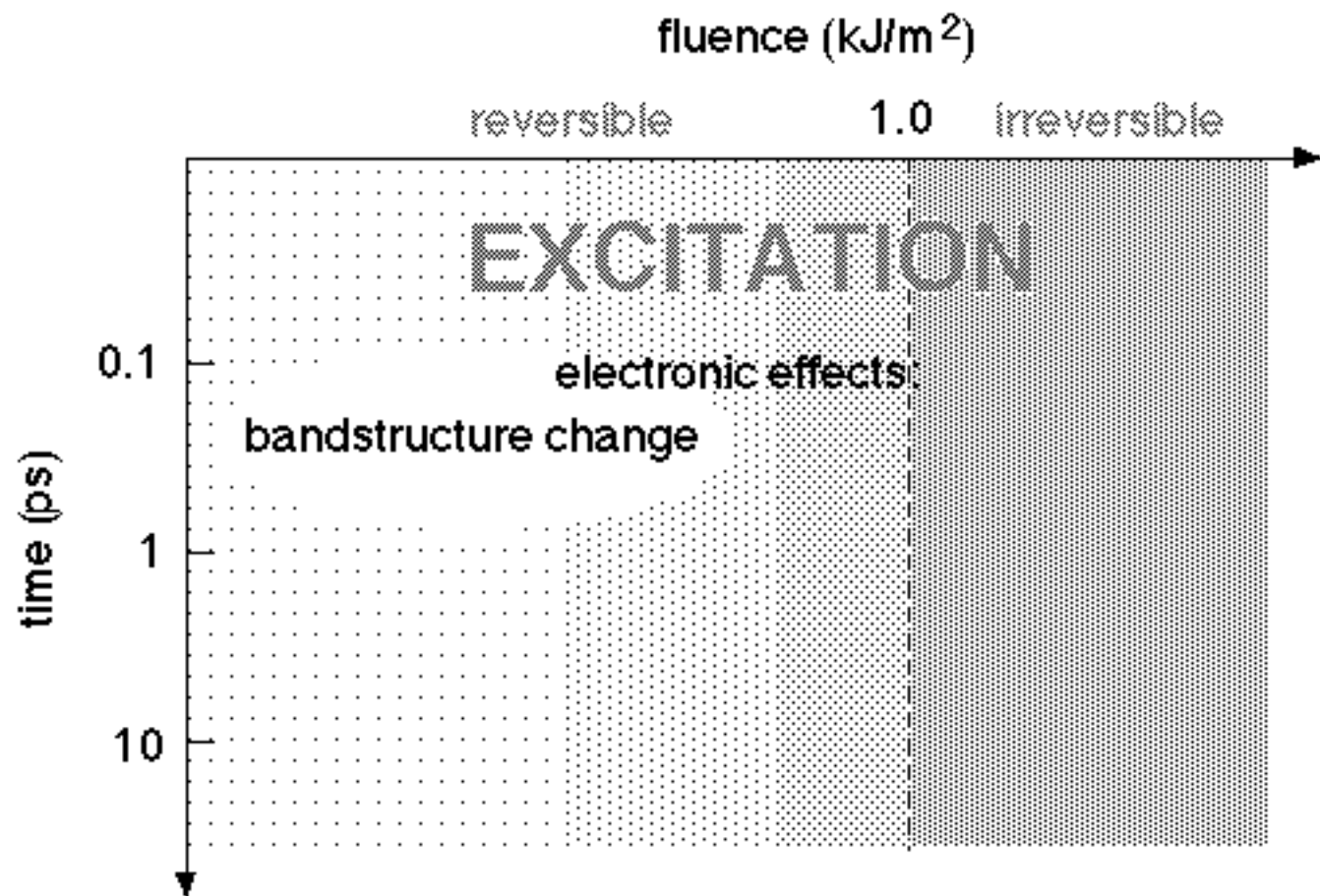
OVERVIEW



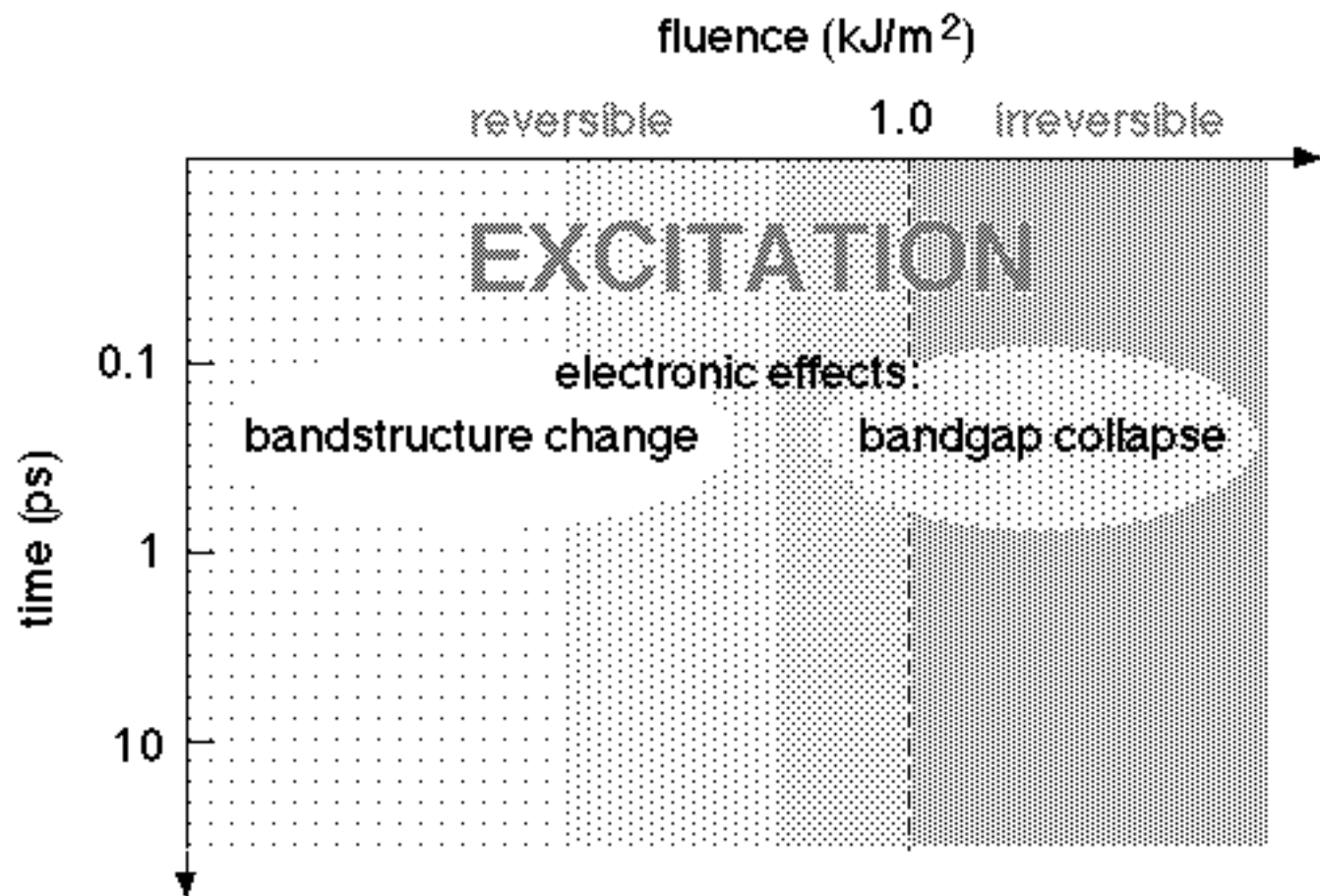
OVERVIEW



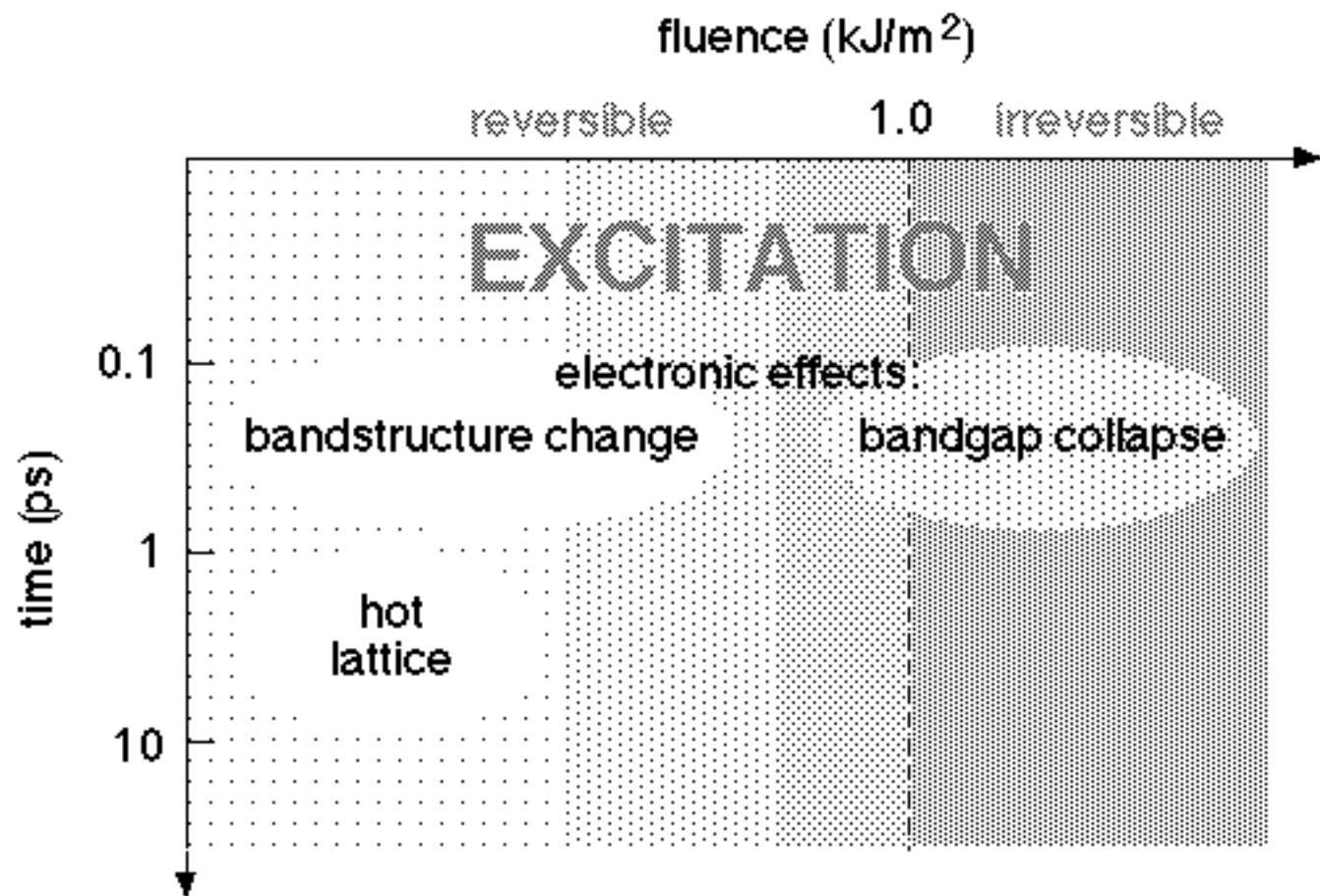
OVERVIEW



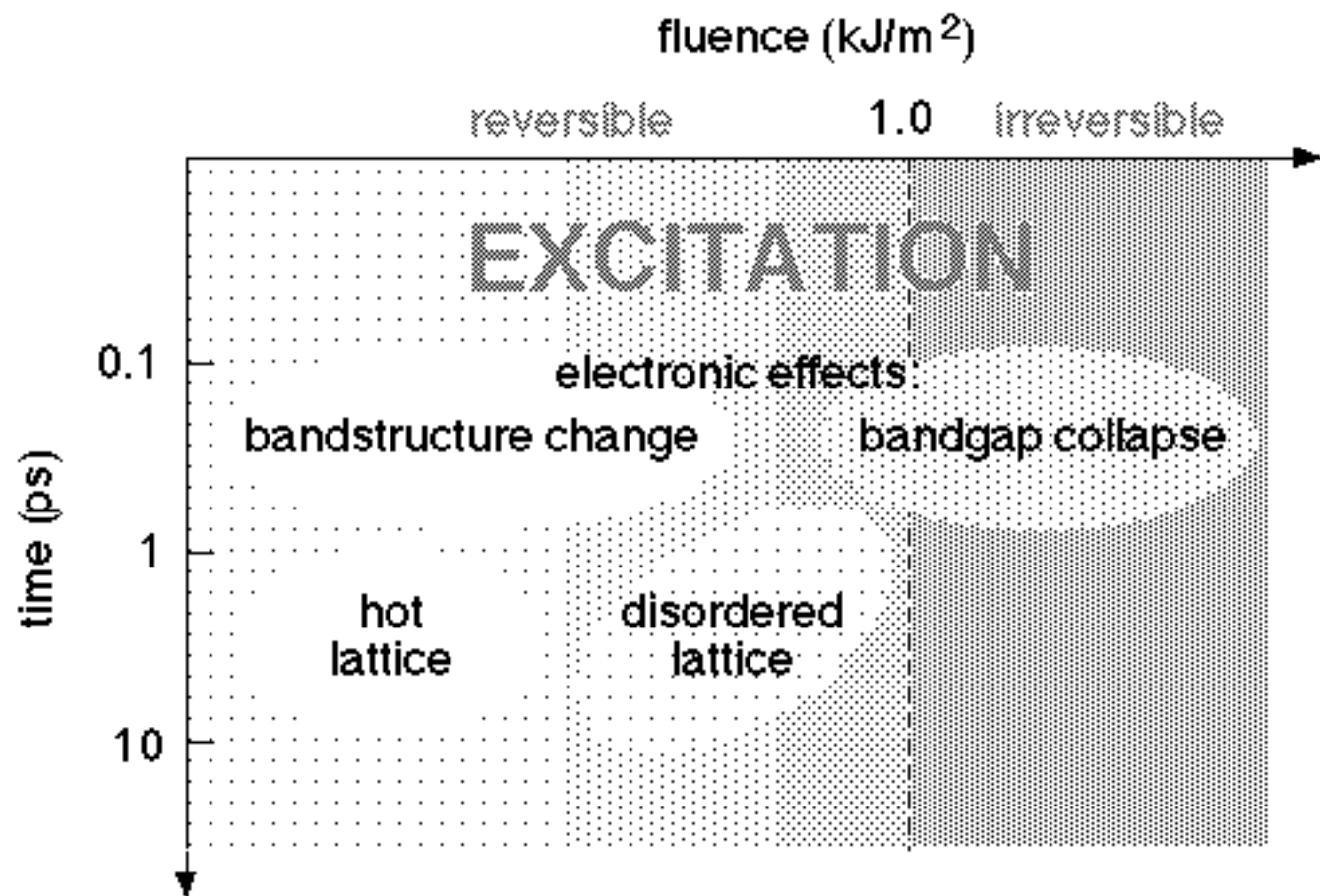
OVERVIEW



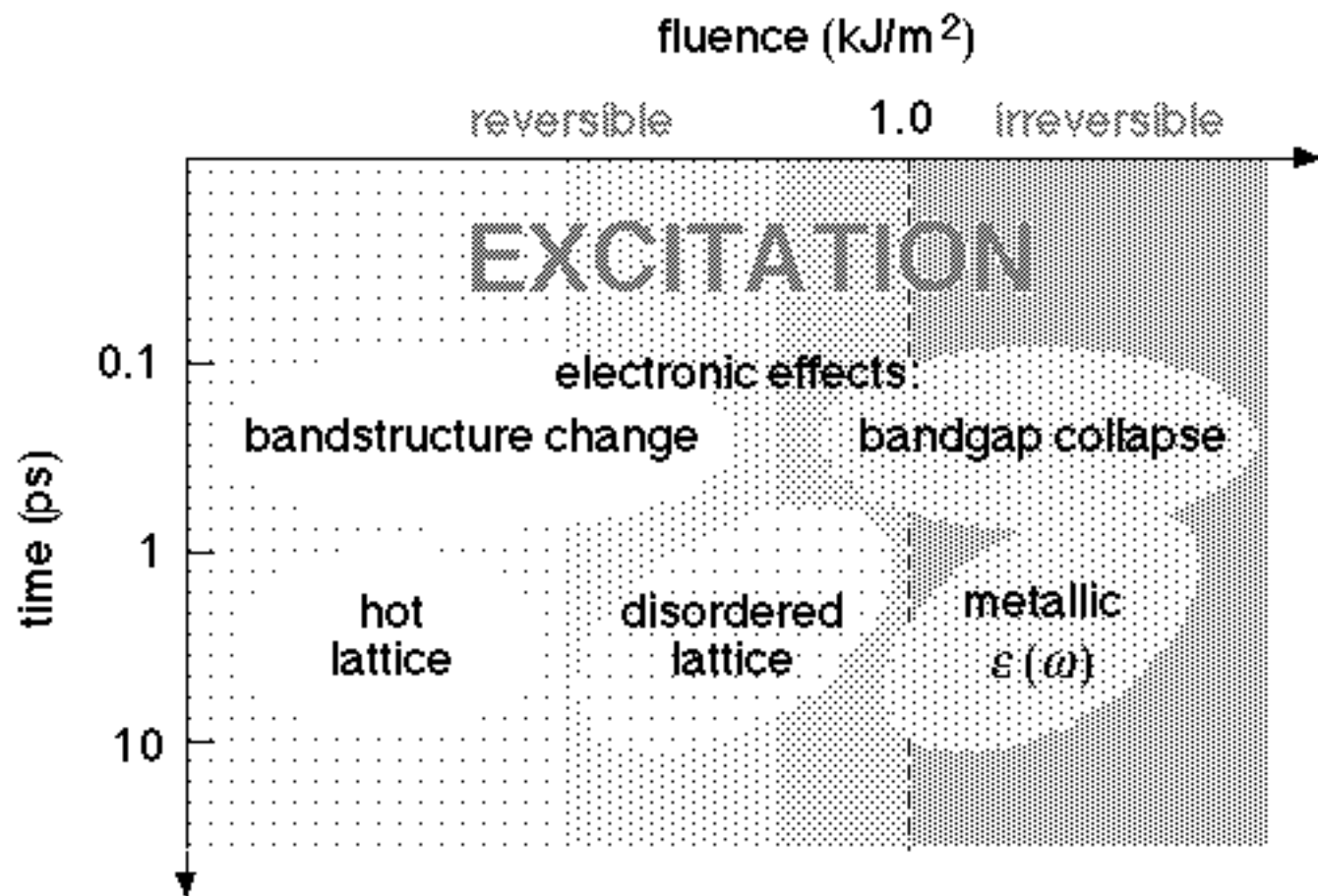
OVERVIEW



OVERVIEW



OVERVIEW



- ▶ measurement of $\epsilon(\omega)$ identifies ultrafast phase changes
- ▶ initial response is electronic, via band structure and electron occupation changes
- ▶ structural effects dominate after a few ps
- ▶ interesting reversible regime



- ▶ strong electronic excitation can drive a structural transition
- ▶ femtosecond lasers allow us to see the dynamics of the transition



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<http://mazor-www.harvard.edu>

