



*IN VINO?*



# ULTRAFAST LASER-INDUCED STRUCTURAL CHANGES IN SEMICONDUCTORS

*Eric Mazur*

*Eli Glezer*

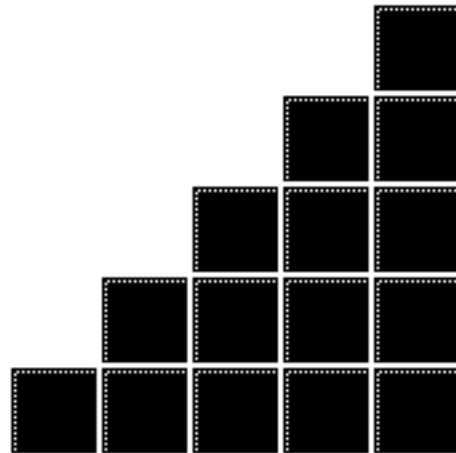
*Li Huang*

*Paul Callan*

*Journées "Maxwell"*

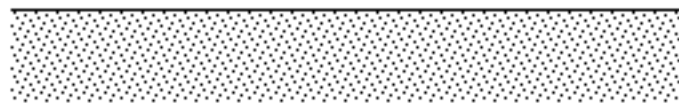
*Bordeaux, France*

*20 May 1997*



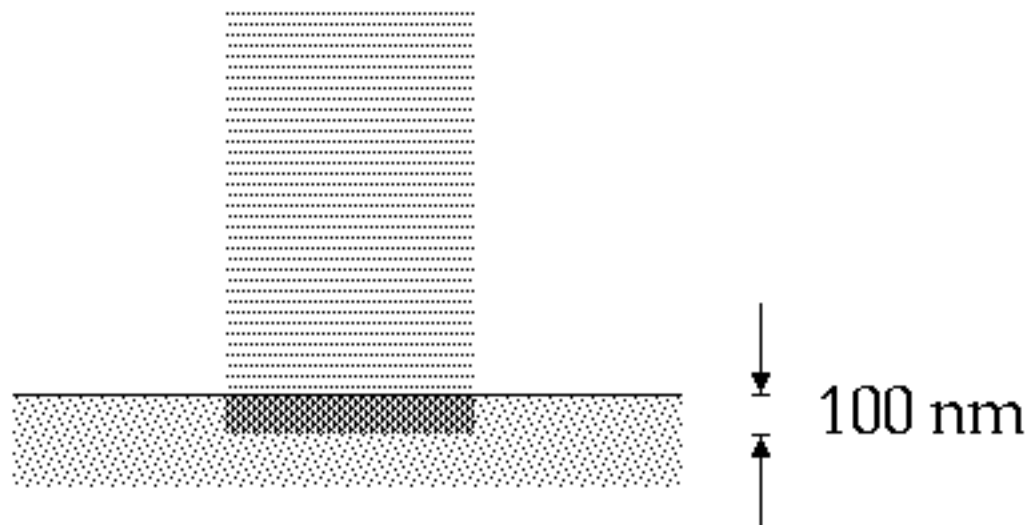
## ***INTRODUCTION***

- 1976: (nanosecond) laser annealing of crystals damaged by ion bombardment
- 1985: femtosecond laser 'melting' of Si



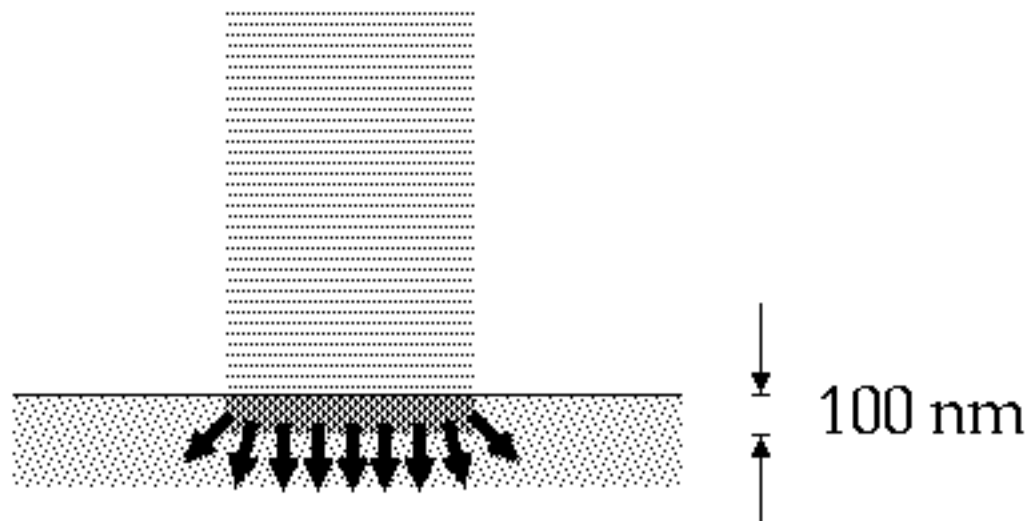
# INTRODUCTION

laser deposits energy near surface...



# INTRODUCTION

energy rapidly diffuses into bulk...

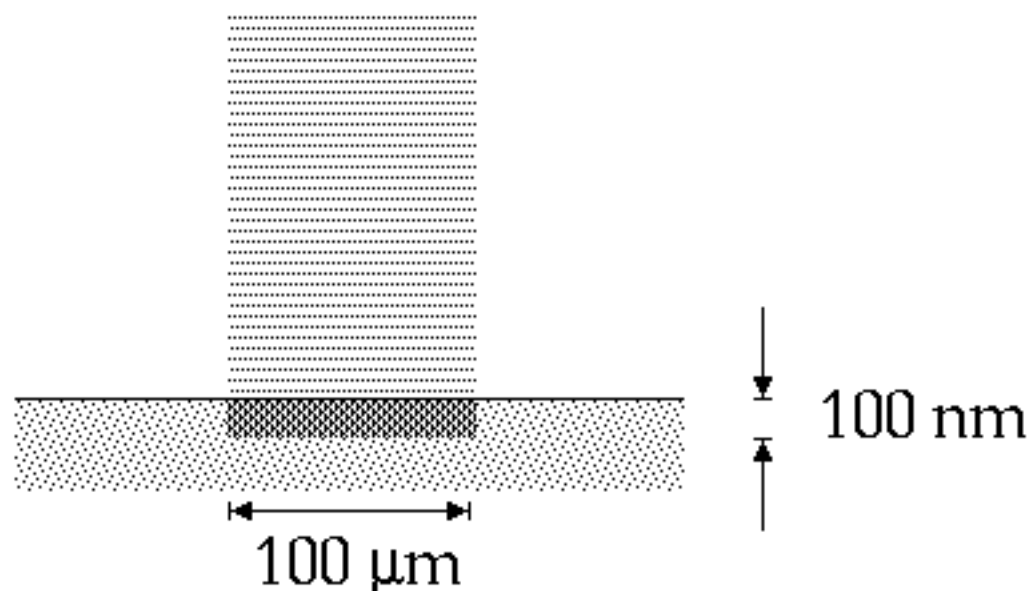


in 1 ns, diffusion length  $\approx 100$  nm



# INTRODUCTION

For  $t < 1$  ns can ignore diffusion



$10\ \mu\text{J}$  into a volume of about  $10^{-15}\ \text{m}^3$ !

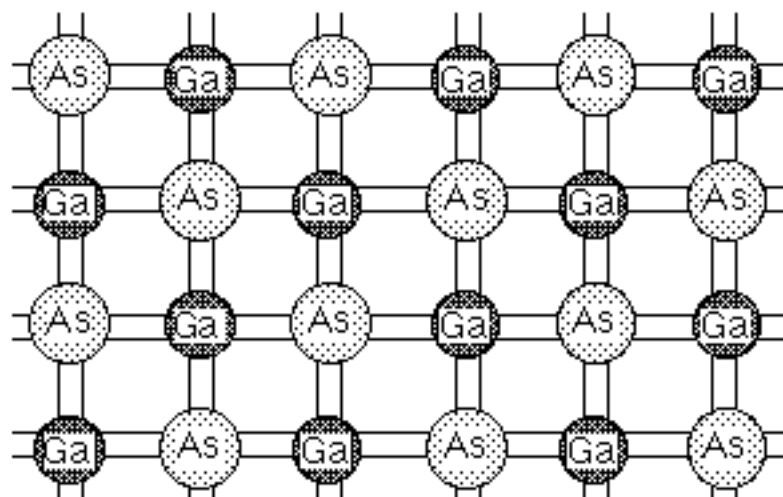


- ① Background
- ② Dielectric function
- ③ Nonlinear properties





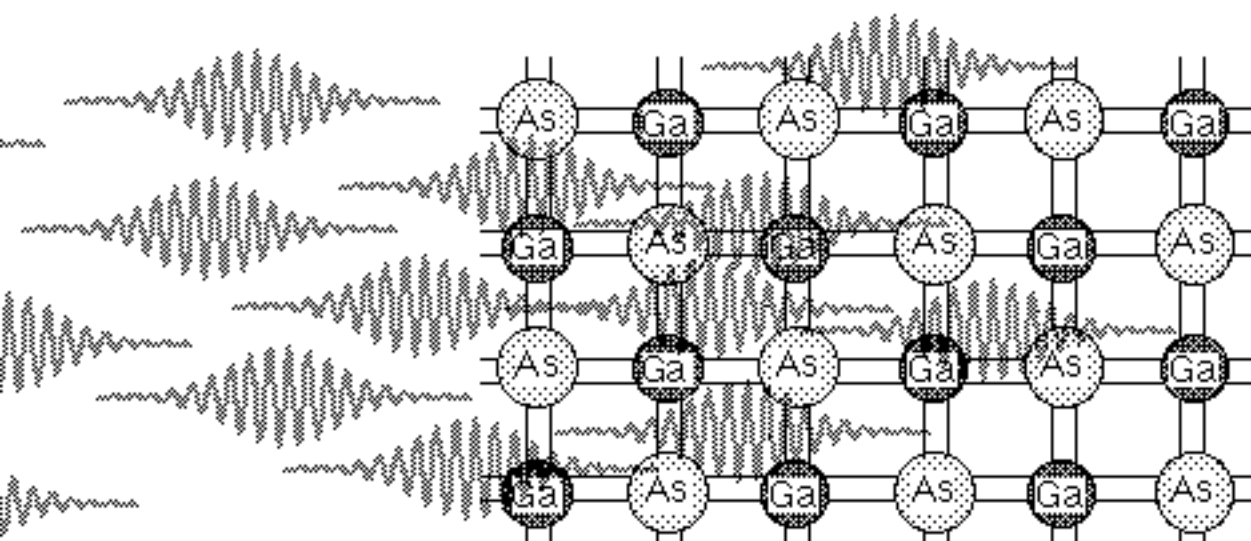
## *EARLY MODELS*



how does light melt a solid?



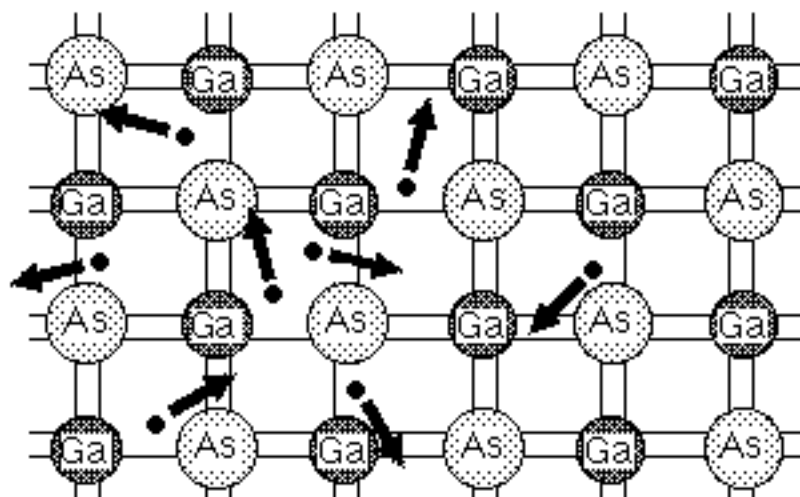
# ***THERMAL MODEL***



photons excite valence electrons...



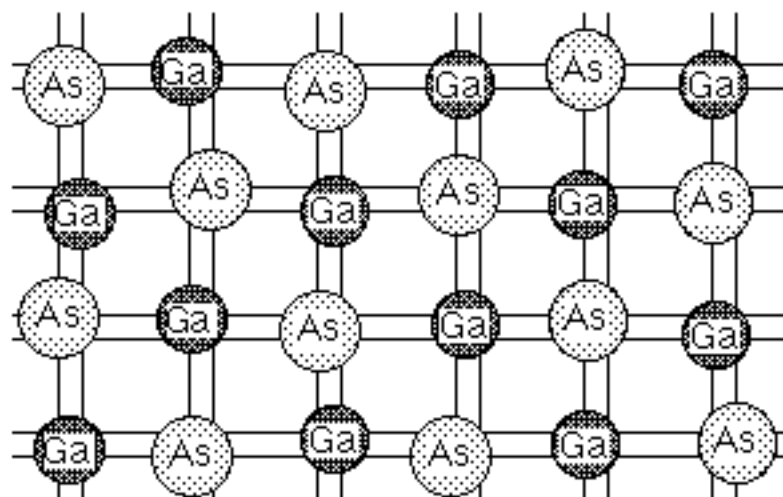
# *THERMAL MODEL*



...and create hot electrons...



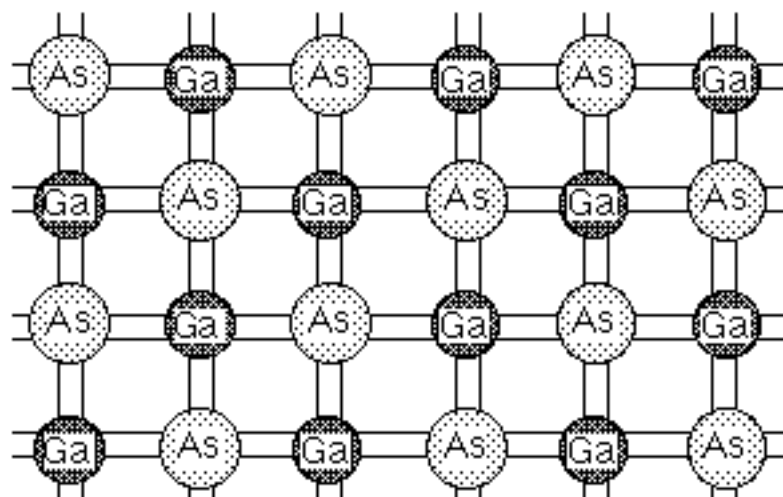
# *THERMAL MODEL*



...which heat lattice



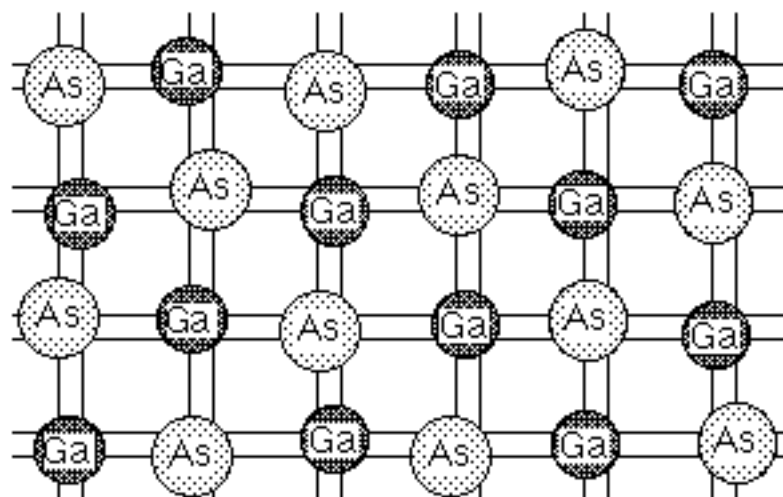
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...which heat lattice



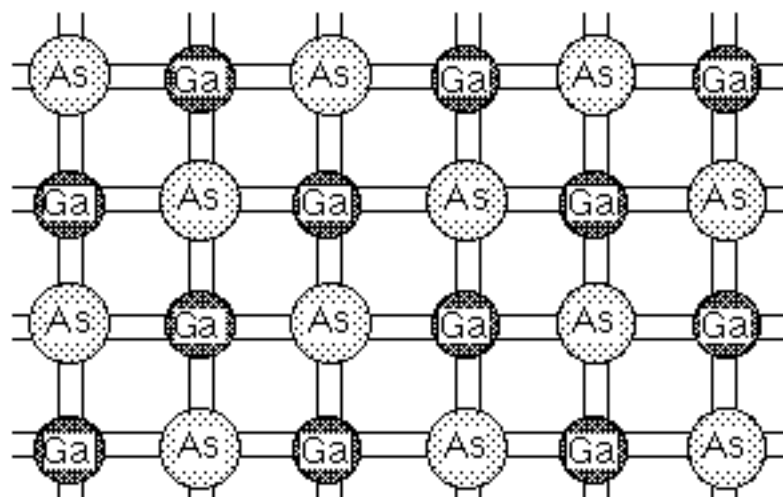
# ***THERMAL MODEL***



...which heat lattice



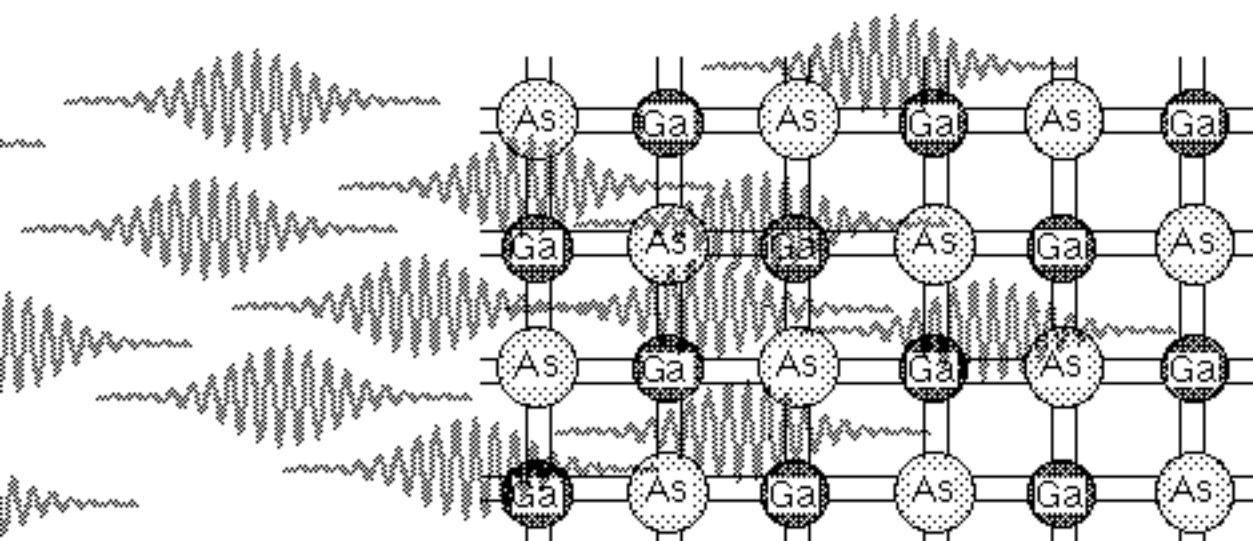
# EARLY MODELS



...or...



# *PLASMA ANNEALING*

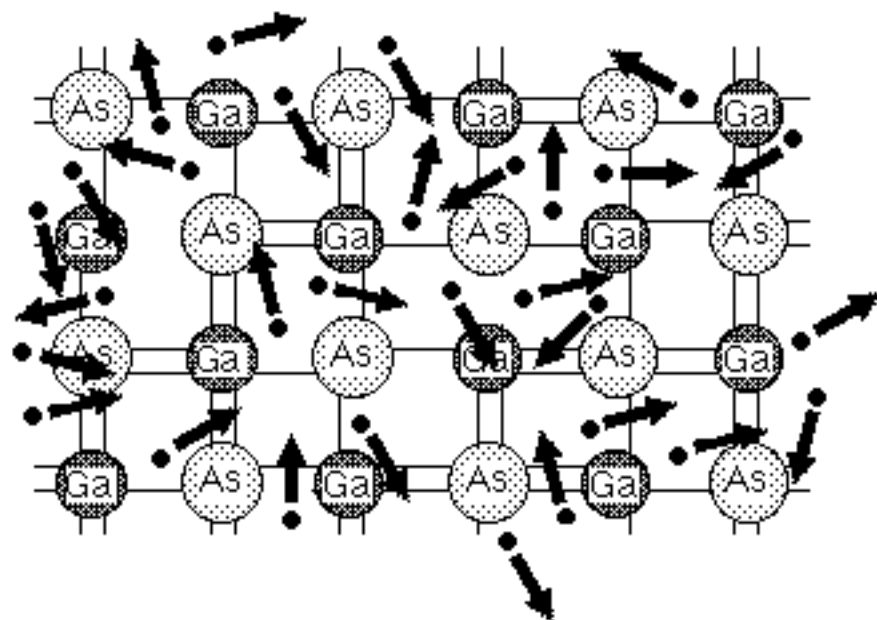


photons excite valence electrons...





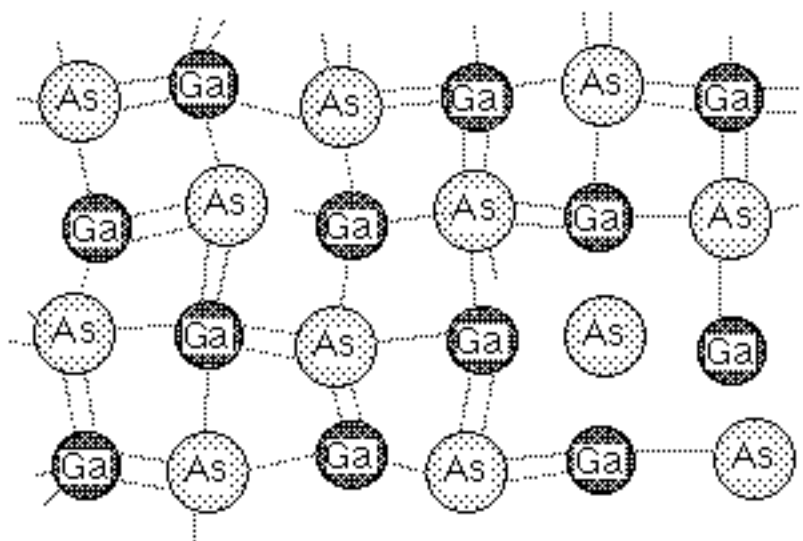
# *PLASMA ANNEALING*



...creating a dense plasma...



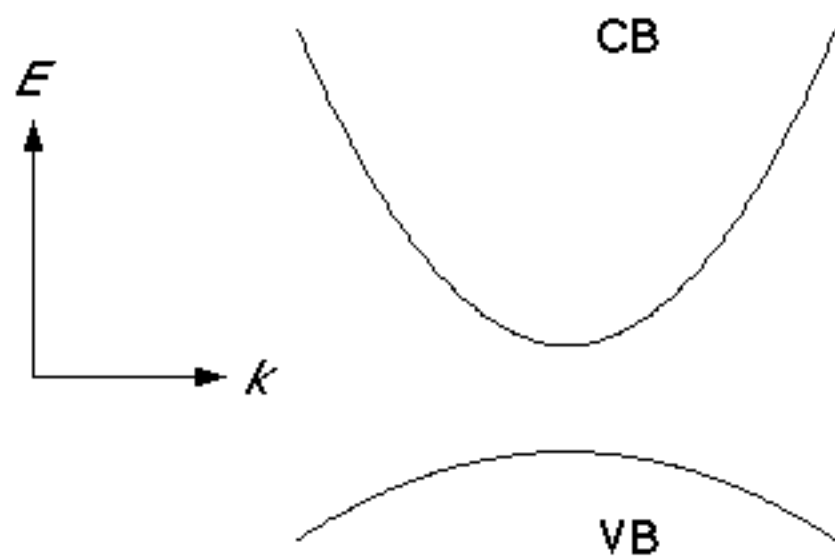
# *PLASMA ANNEALING*



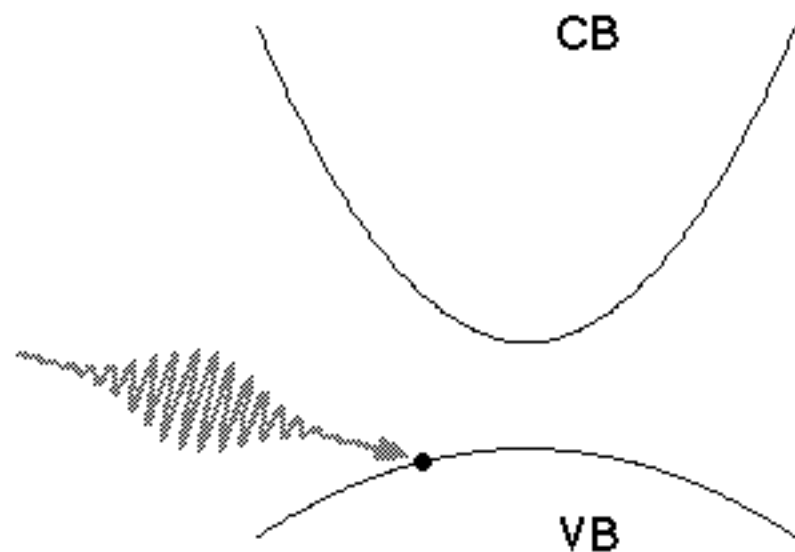
...and breaking bonds



# EXCITATION



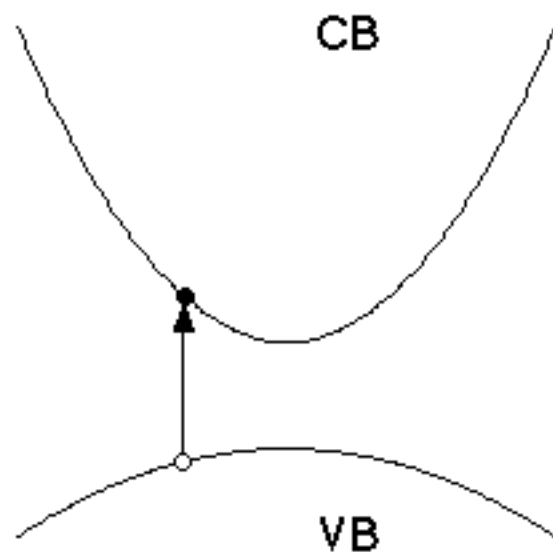
# *EXCITATION*



photon excites valence electron



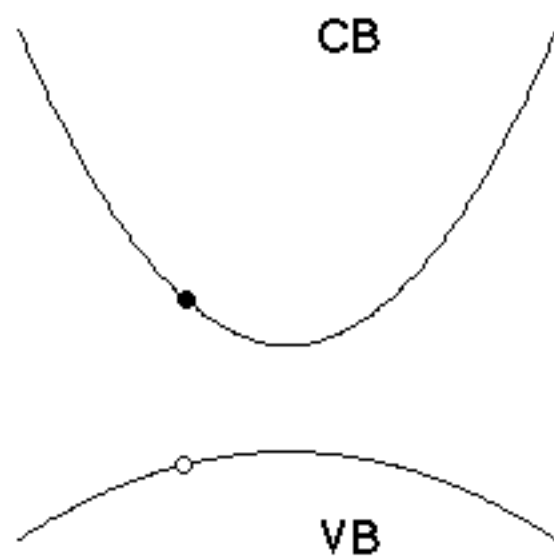
# *EXCITATION*



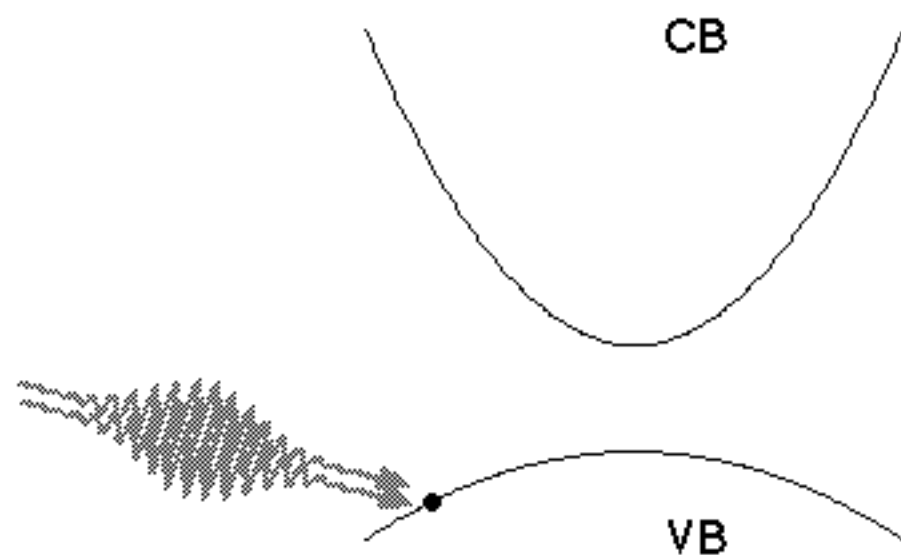
creating an electron-hole pair



# *EXCITATION*



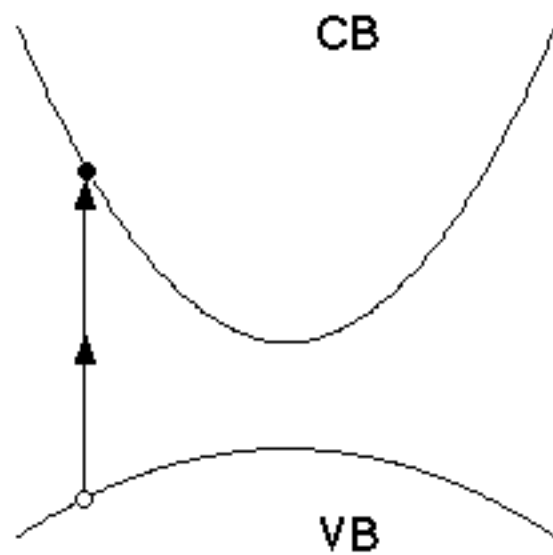
# *EXCITATION*



multiphoton excitation

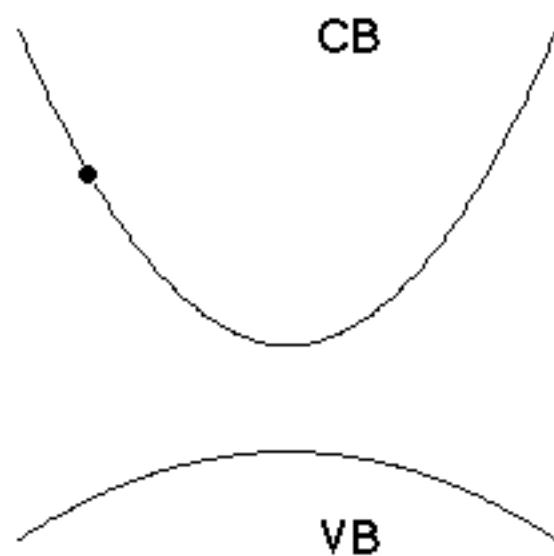


# EXCITATION

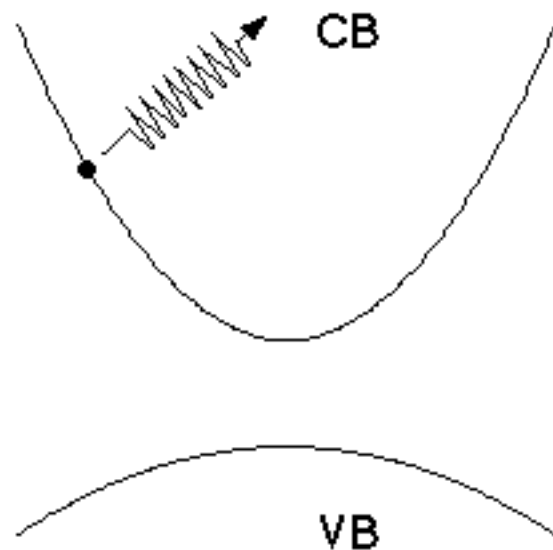




# *EXCITATION*



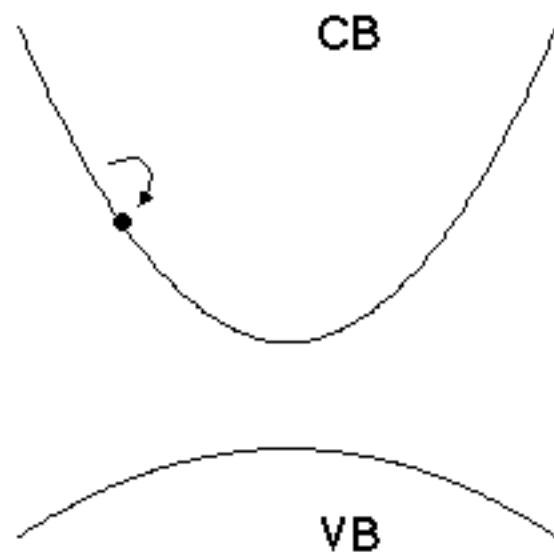
# RELAXATION



emission of LO phonons



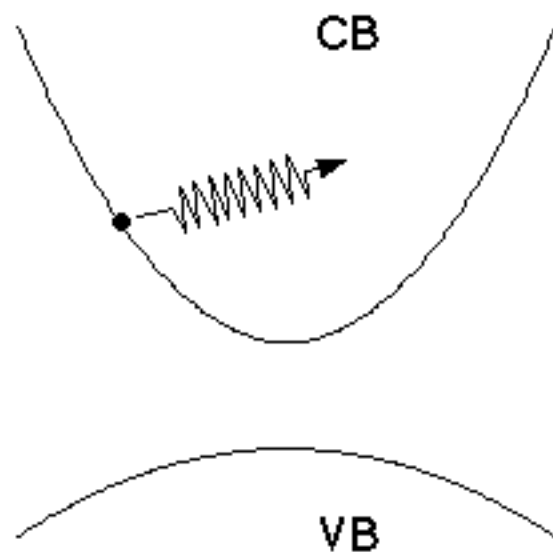
# RELAXATION



emission of LO phonons



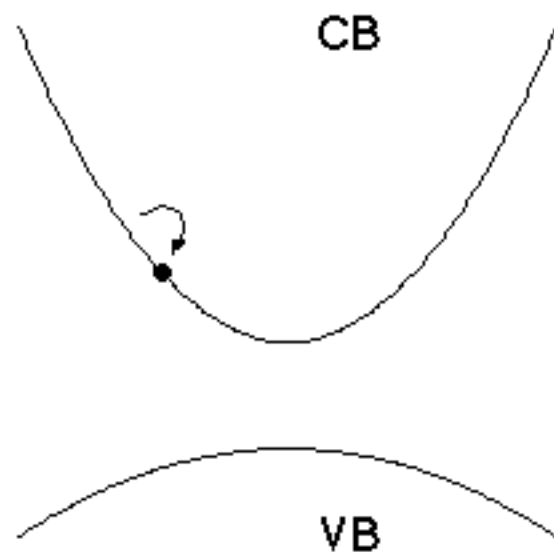
# RELAXATION



emission of LO phonons



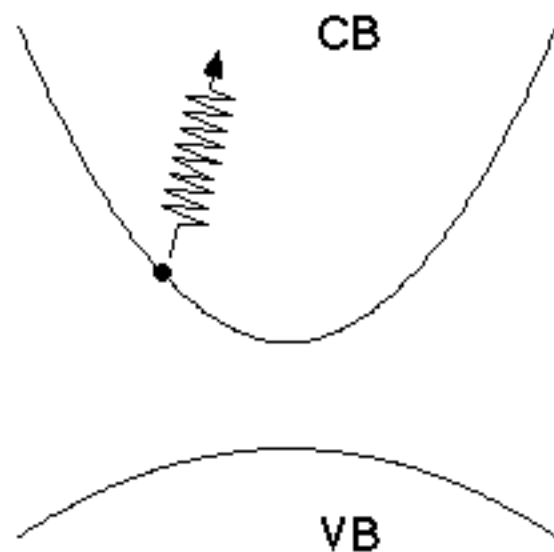
# RELAXATION



emission of LO phonons



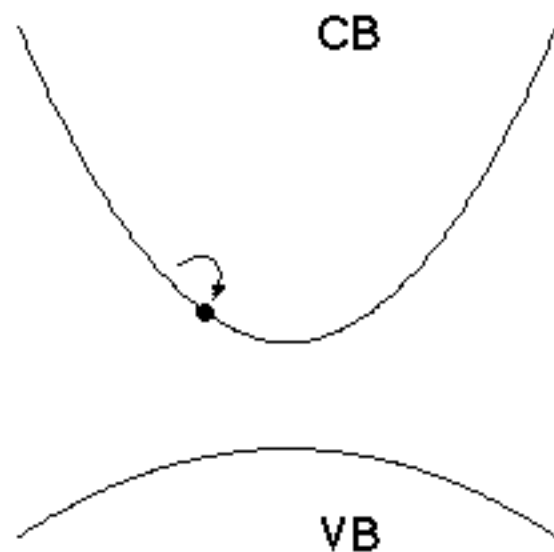
# RELAXATION



emission of LO phonons



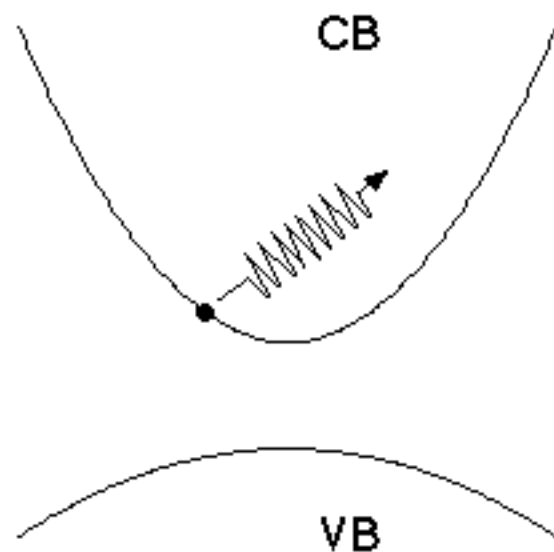
# RELAXATION



emission of LO phonons



# RELAXATION

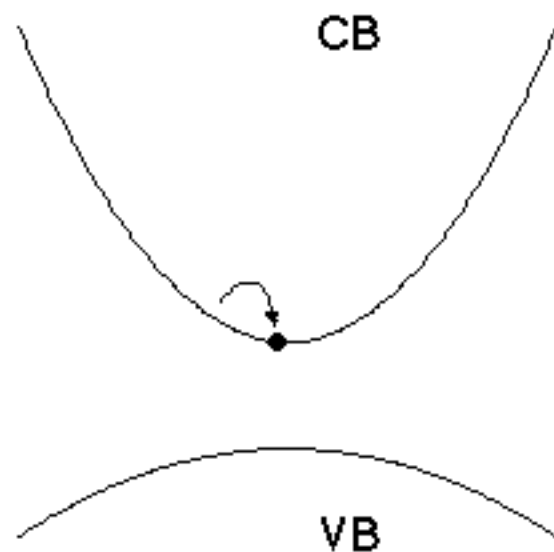


emission of LO phonons





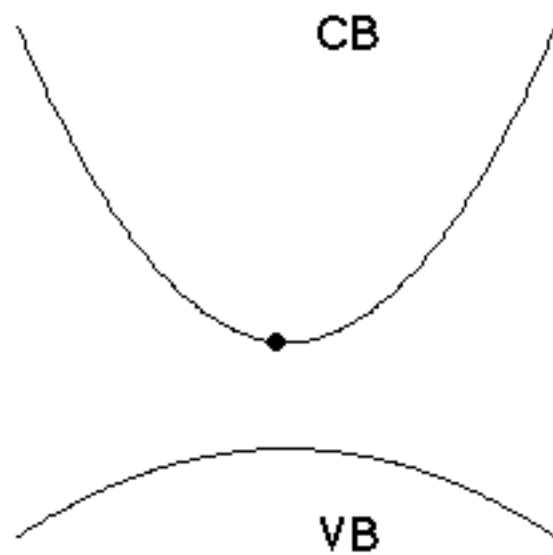
# *RELAXATION*



emission of LO phonons

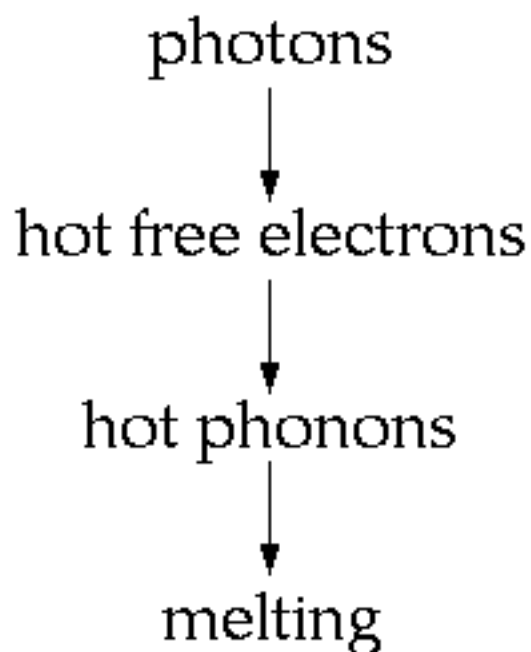


# RELAXATION



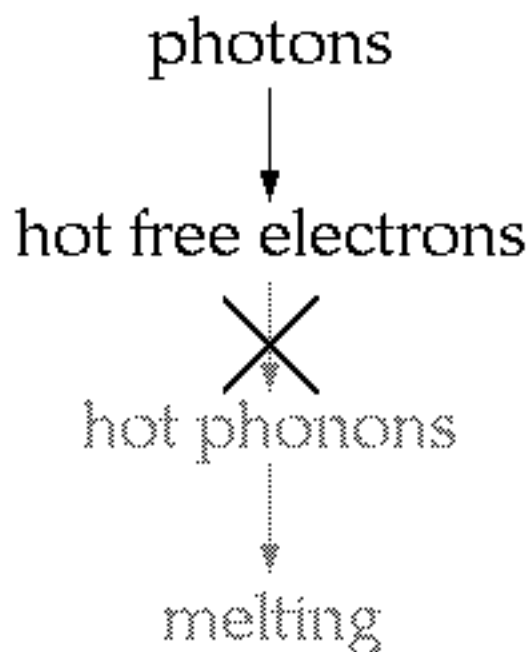
## ***TIME SCALE***

long pulses ( $\tau_{exc} \gg 1$  ps):      *thermal melting*



## ***TIME SCALE***

short pulses ( $\tau_{exc} \ll 1$  ps):      *nonthermal disordering*

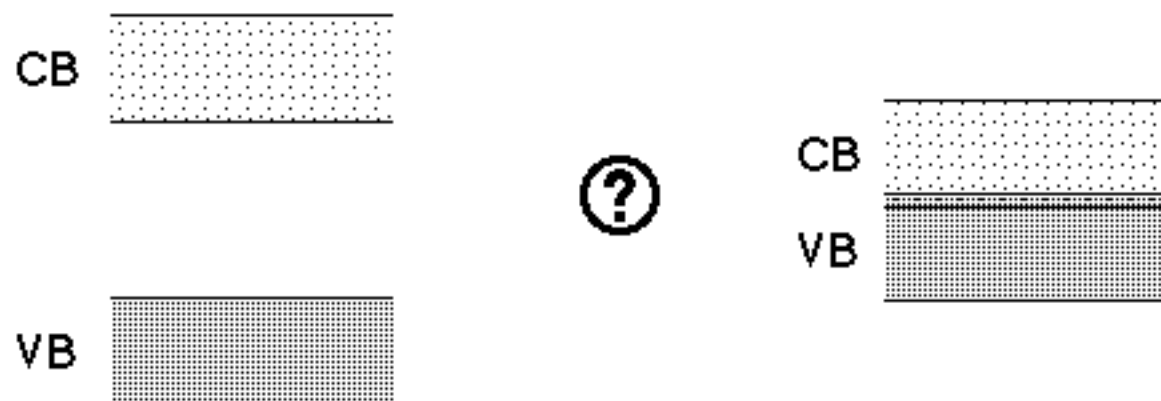


- ① Background
- ② Dielectric function
- ③ Nonlinear properties



# FEMTOSECOND WORK

intense pulses can 'melt' GaAs, Si, but how?



# FEMTOSECOND WORK

intrinsic properties

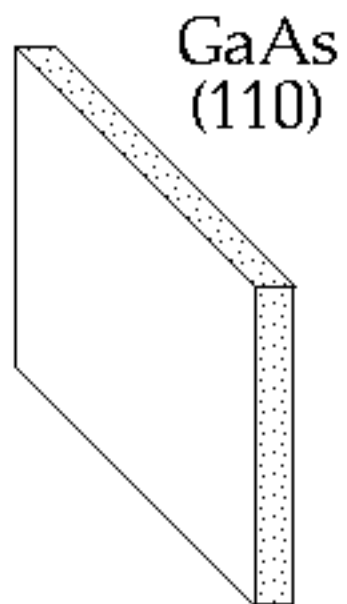
measured properties

$$\varepsilon', \varepsilon''(\omega) \longrightarrow R$$

$$\begin{array}{l} \varepsilon', \varepsilon''(\omega) \\ \varepsilon', \varepsilon''(2\omega) \\ \chi_{(2)} \end{array} \longrightarrow \text{SHG}$$

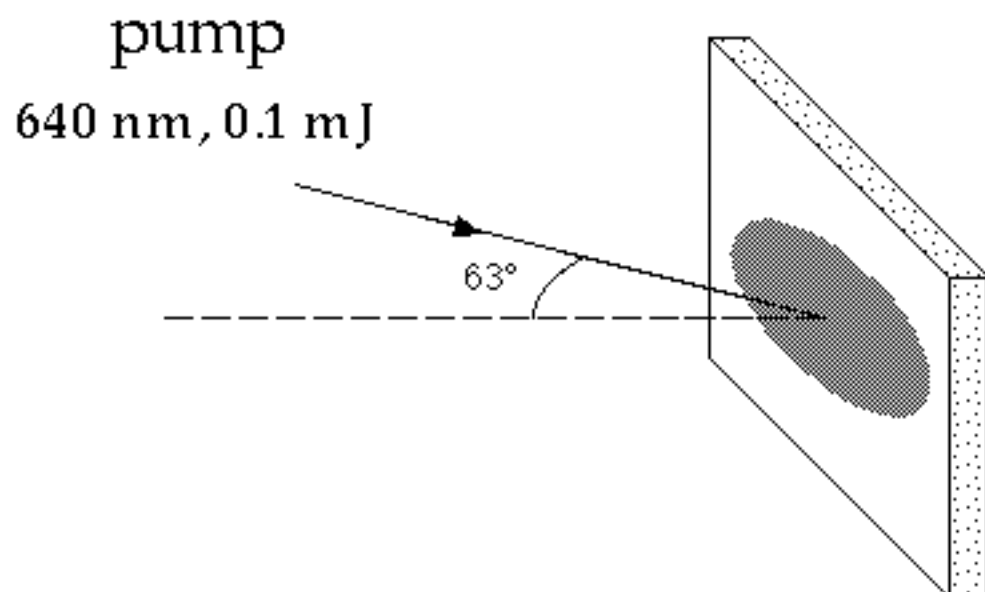


## *2.2 eV DIELECTRIC CONSTANT*

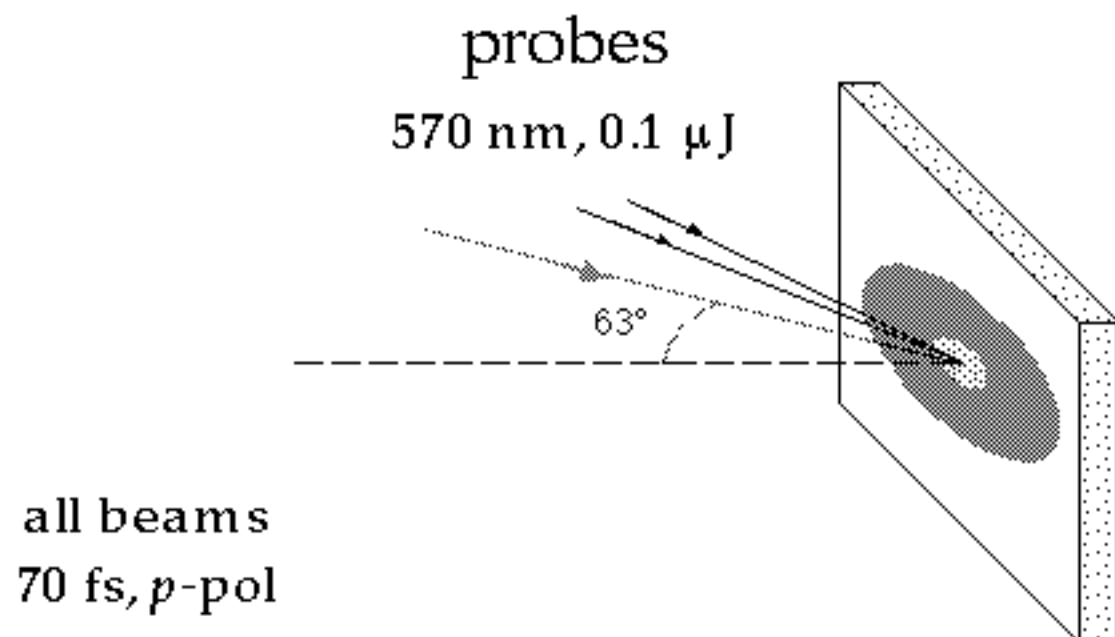




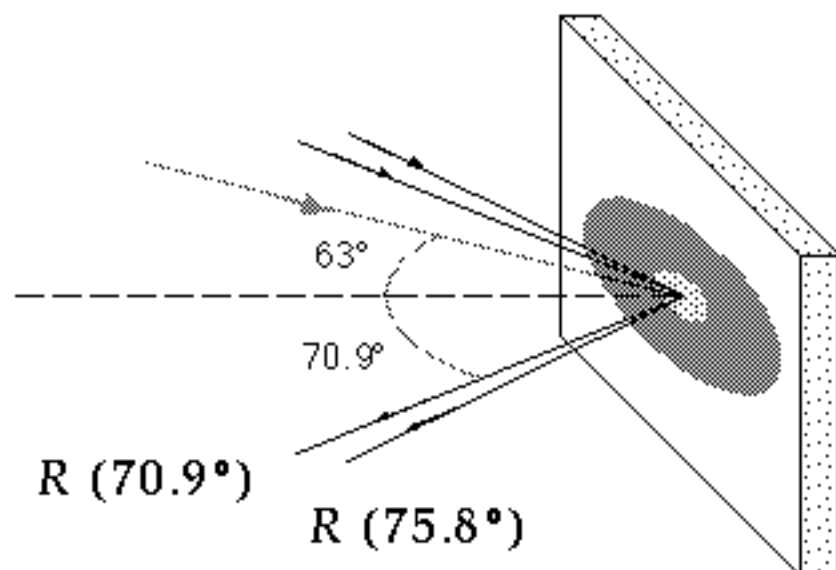
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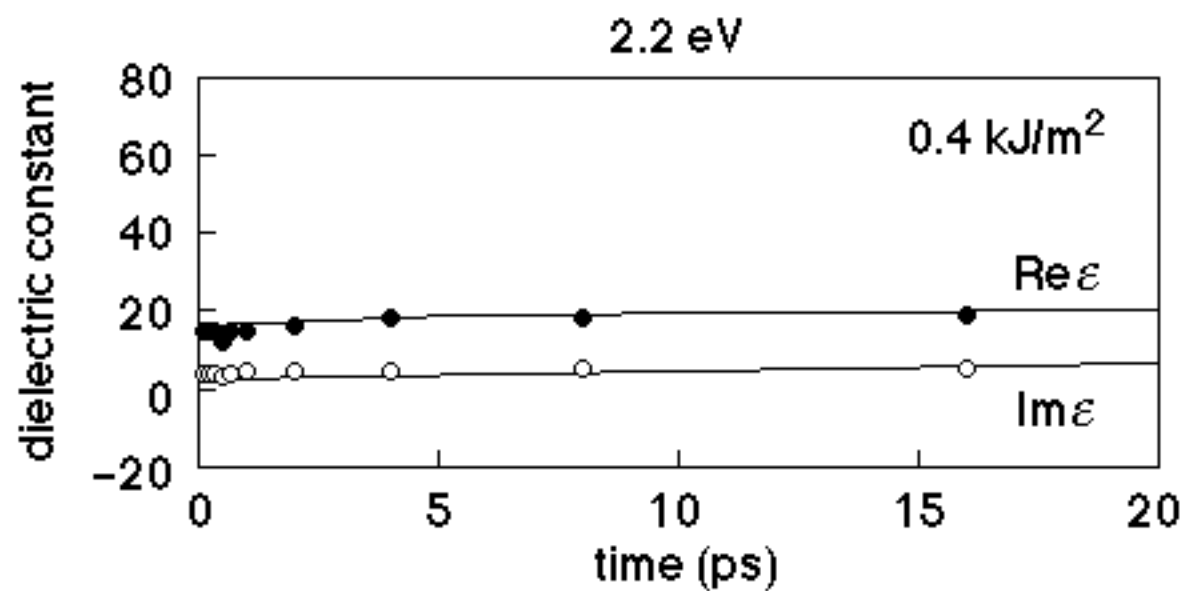
## 2.2 eV DIELECTRIC CONSTANT



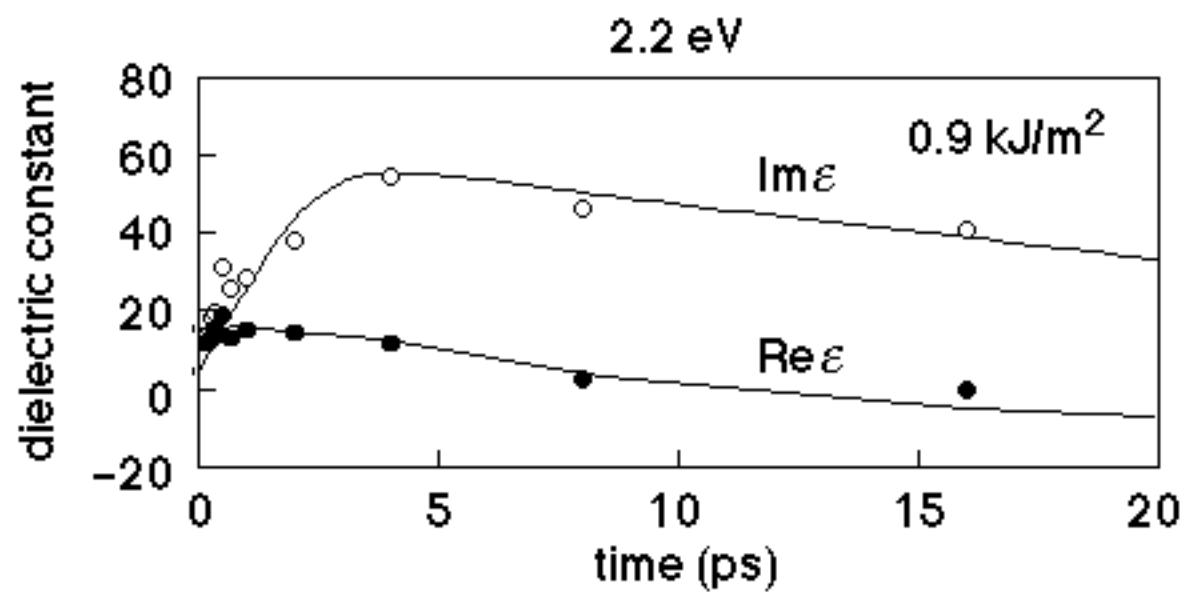
## *2.2 eV DIELECTRIC CONSTANT*



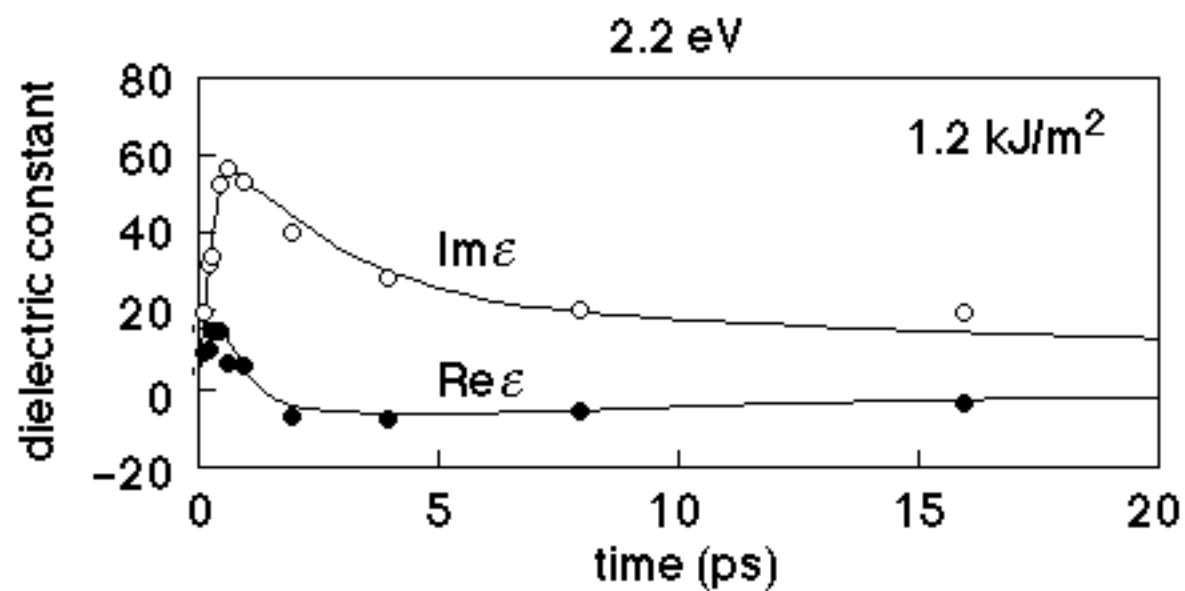
# TIME DEPENDENCE



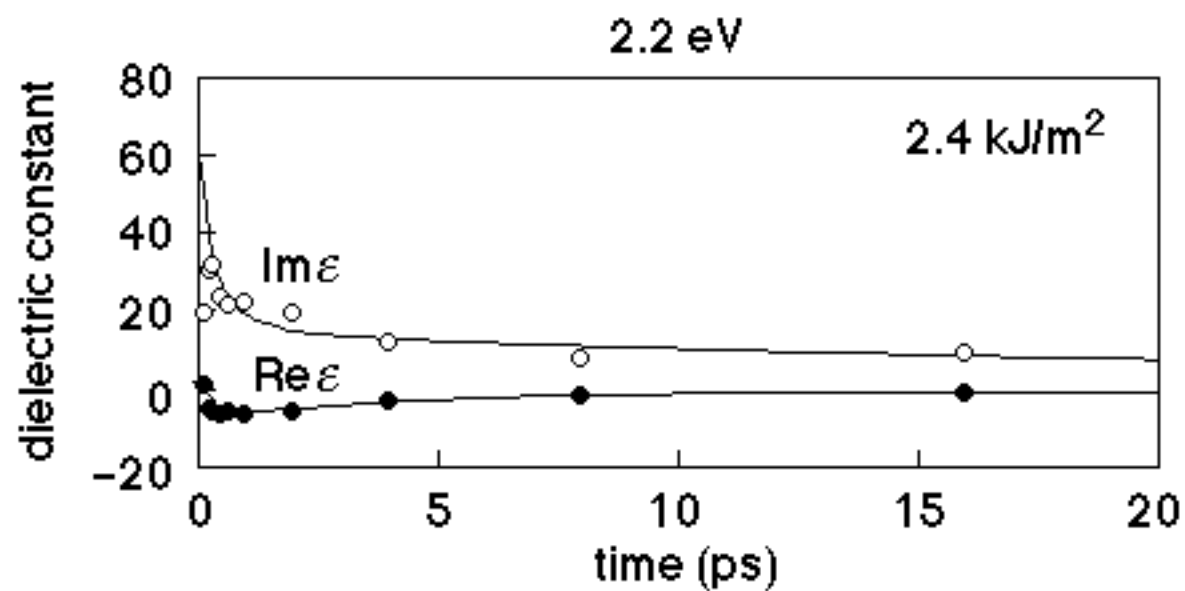
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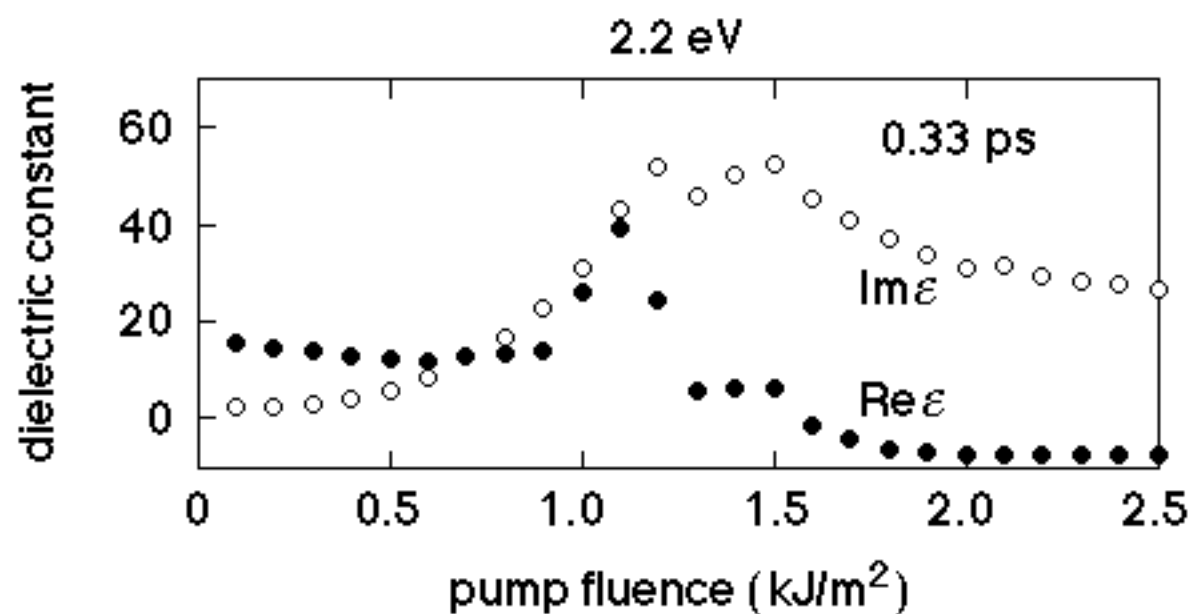
# TIME DEPENDENCE



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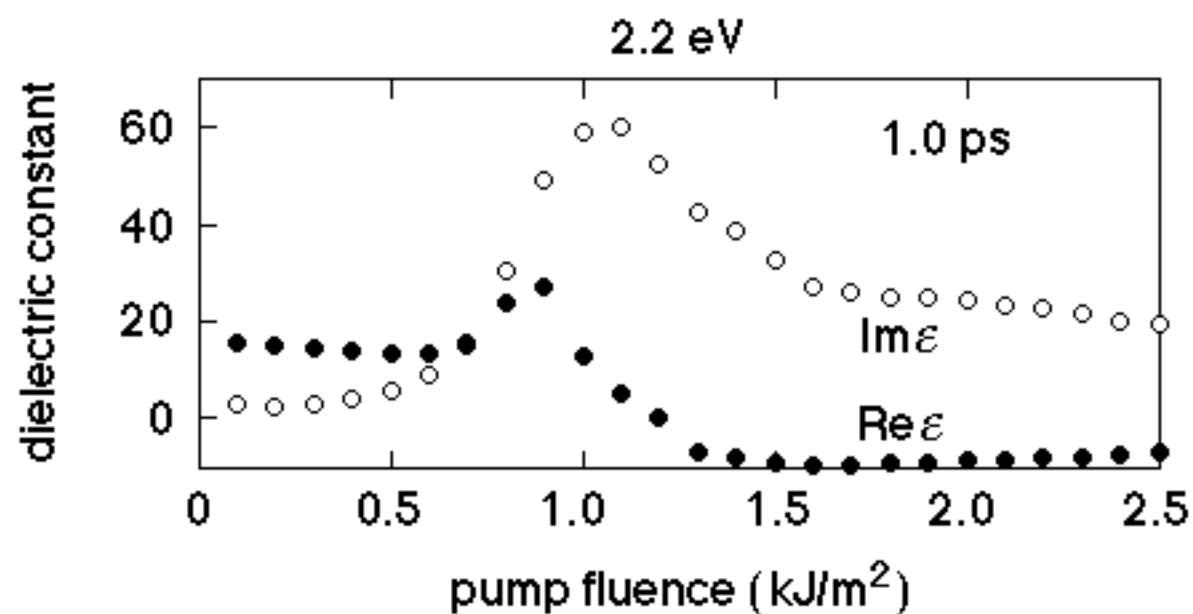


# FLUENCE DEPENDENCE

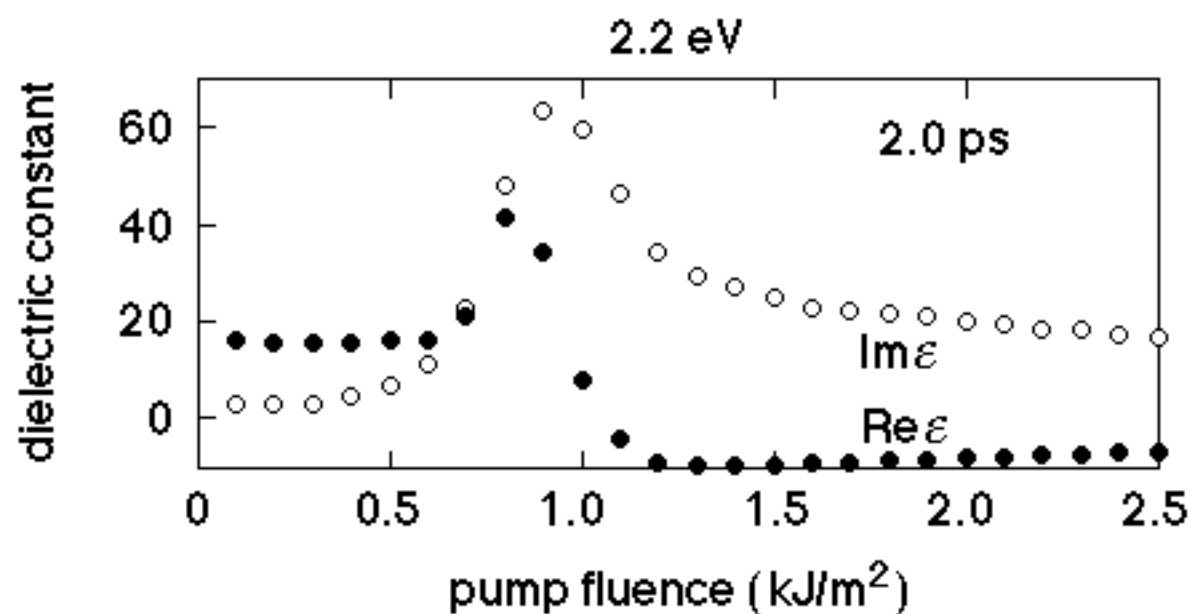




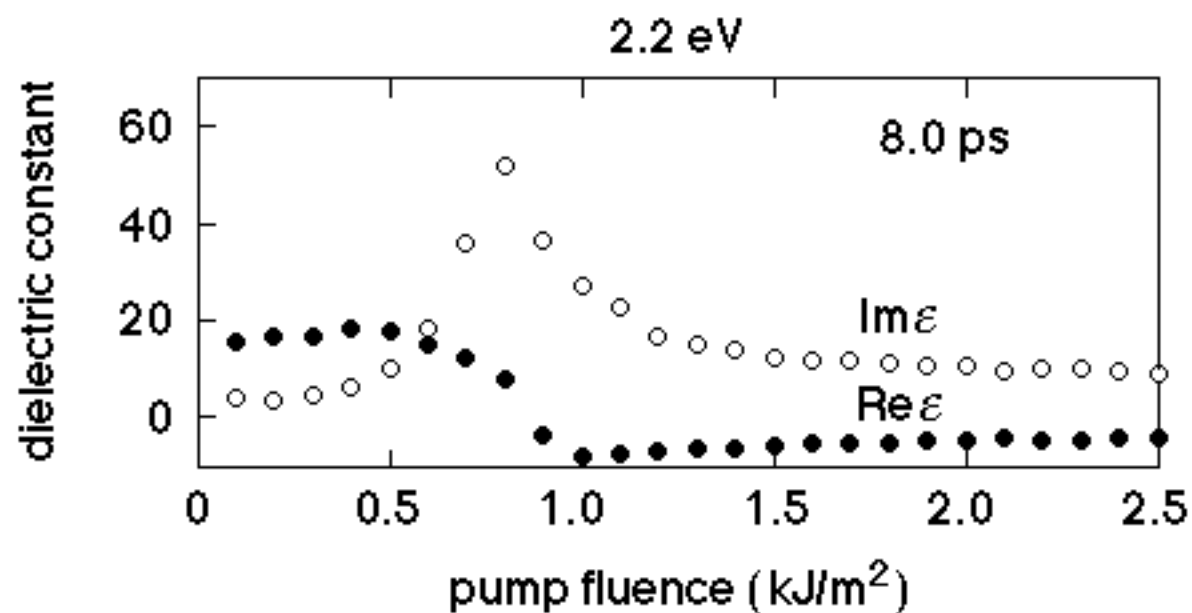
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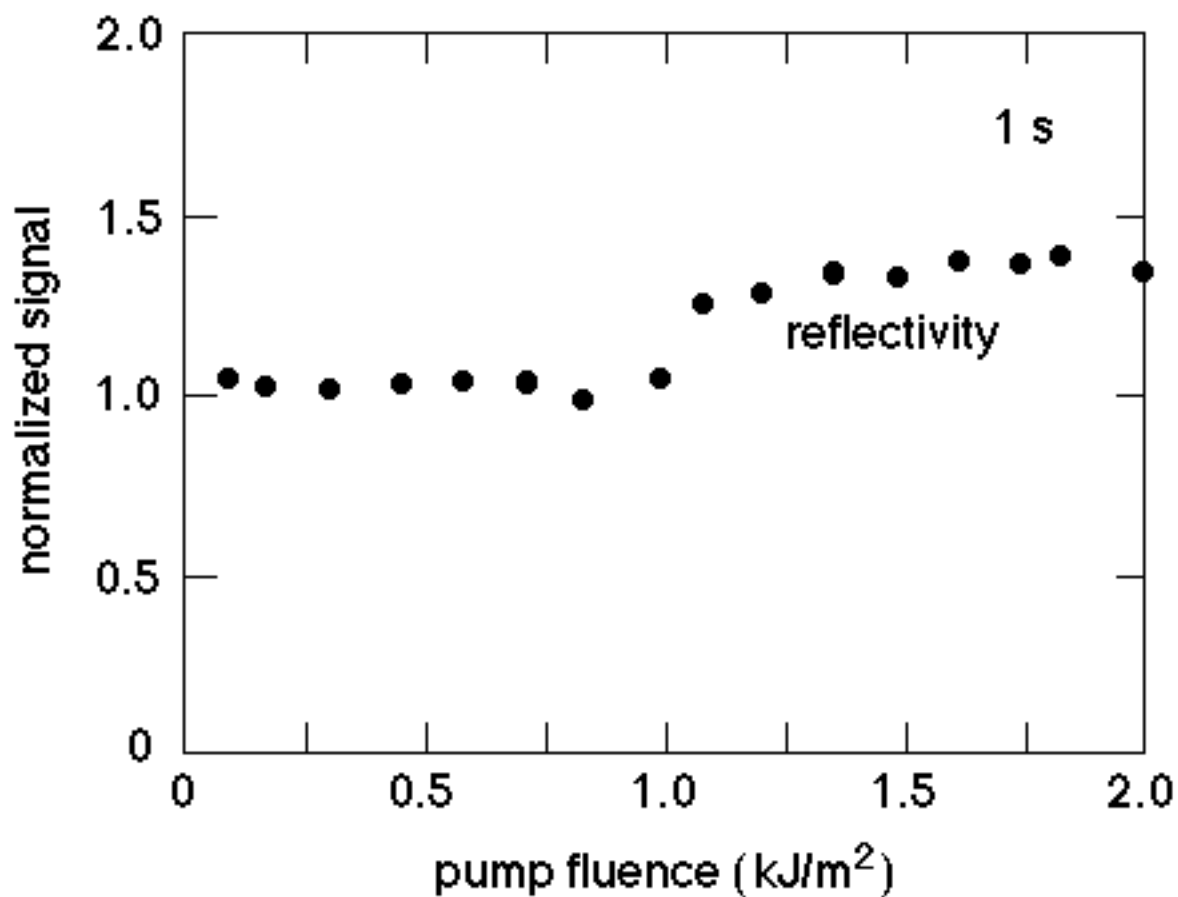
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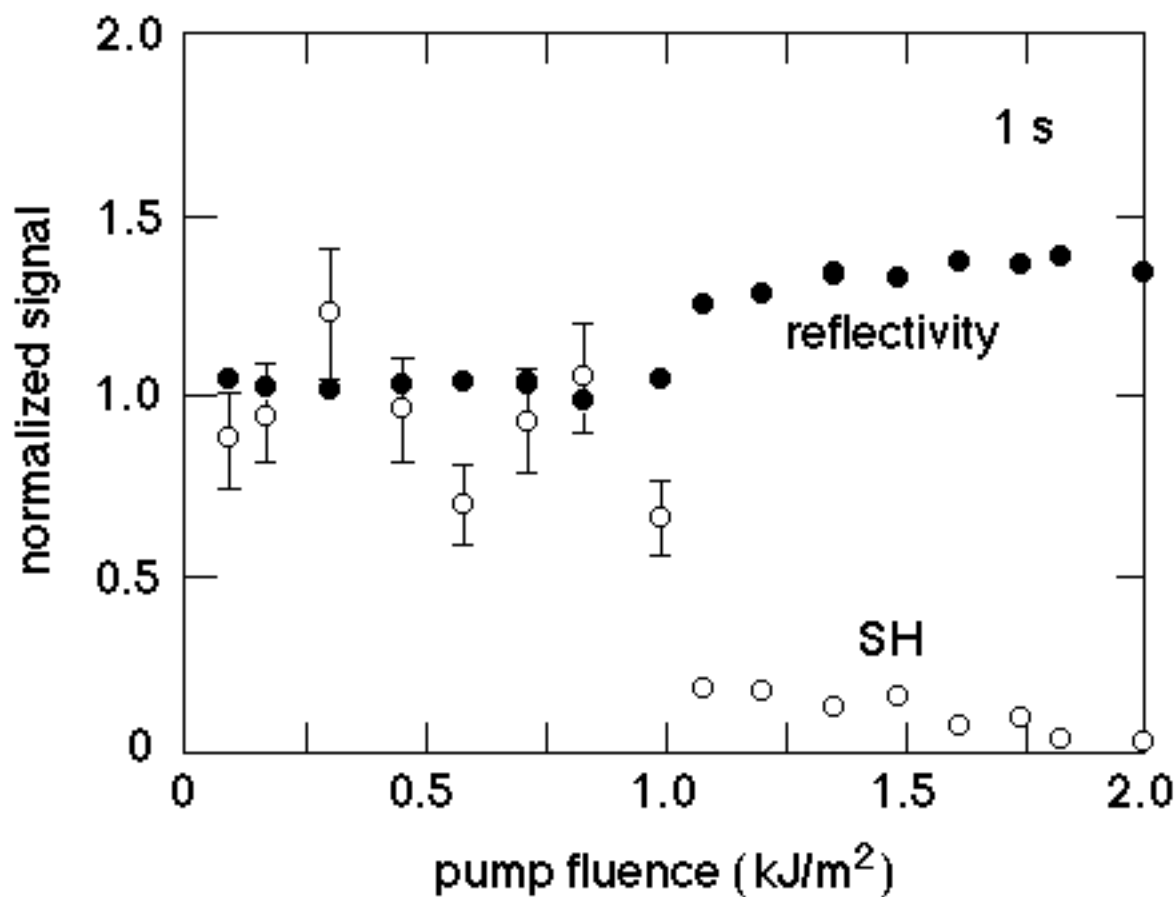
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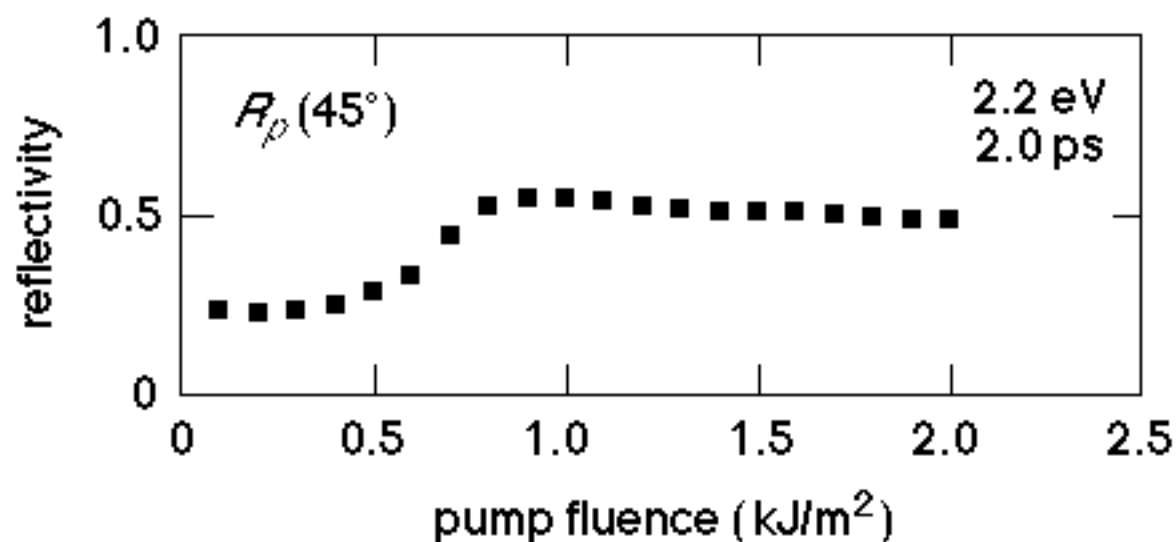
# REVERSIBLE vs. IRREVERSIBLE



# REVERSIBLE vs. IRREVERSIBLE



## SIMPLE MODEL



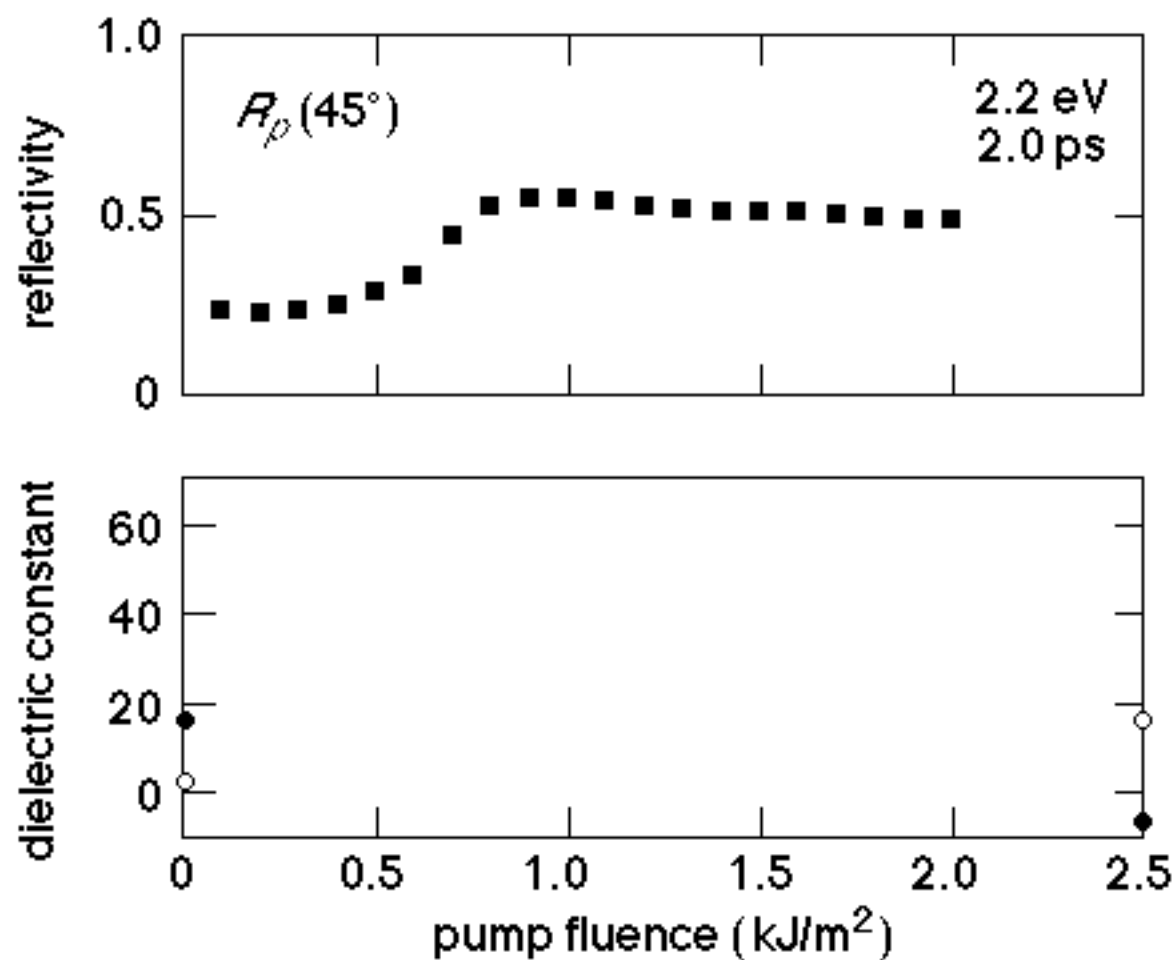
Drude model:

$$\text{Re}(\epsilon) = 1 - \frac{Ne^2}{m\epsilon_0} \frac{1}{\omega^2 + \gamma^2}$$

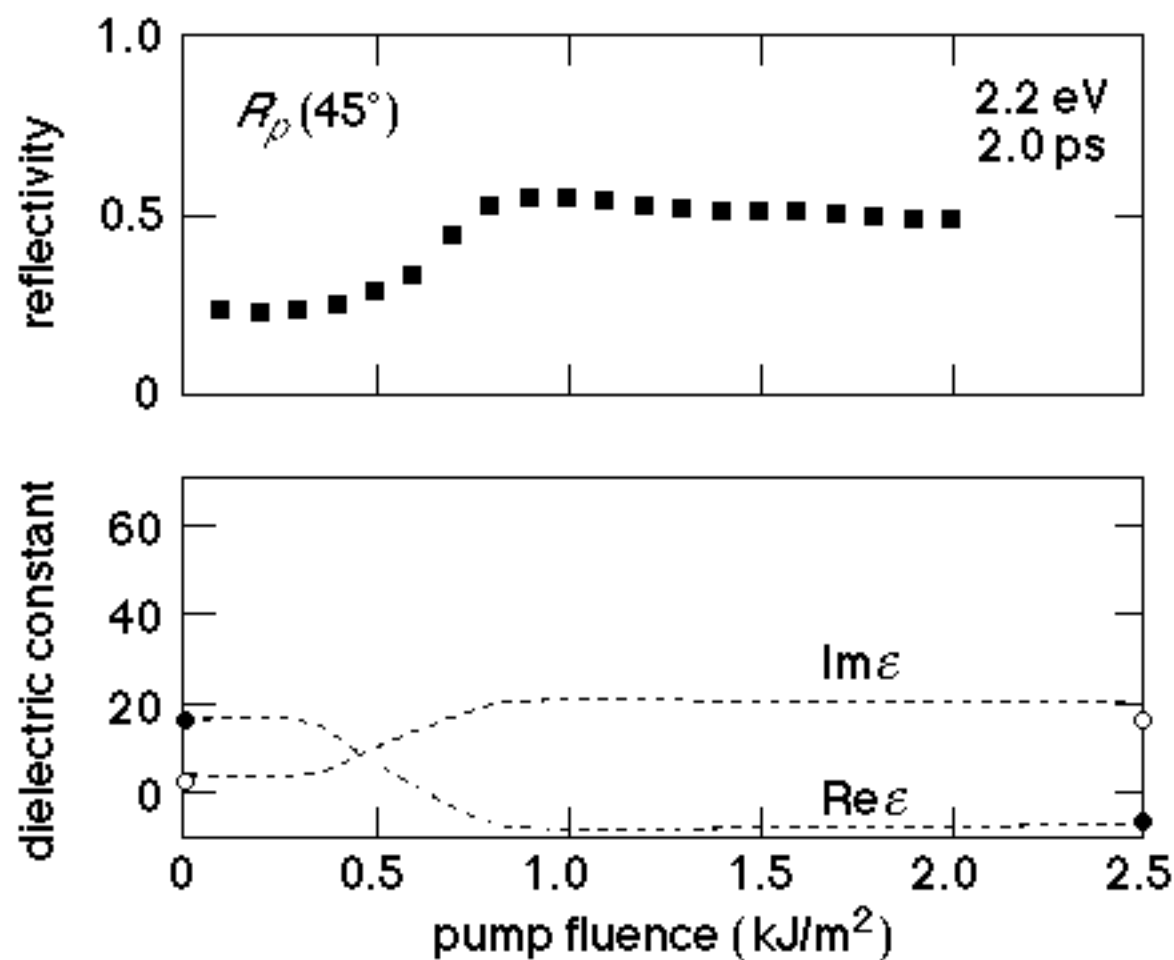
$$\text{Im}(\epsilon) = \frac{Ne^2}{m\epsilon_0} \frac{\gamma\omega}{\omega^4 + \gamma^2\omega^2}$$



# ***SIMPLE MODEL***

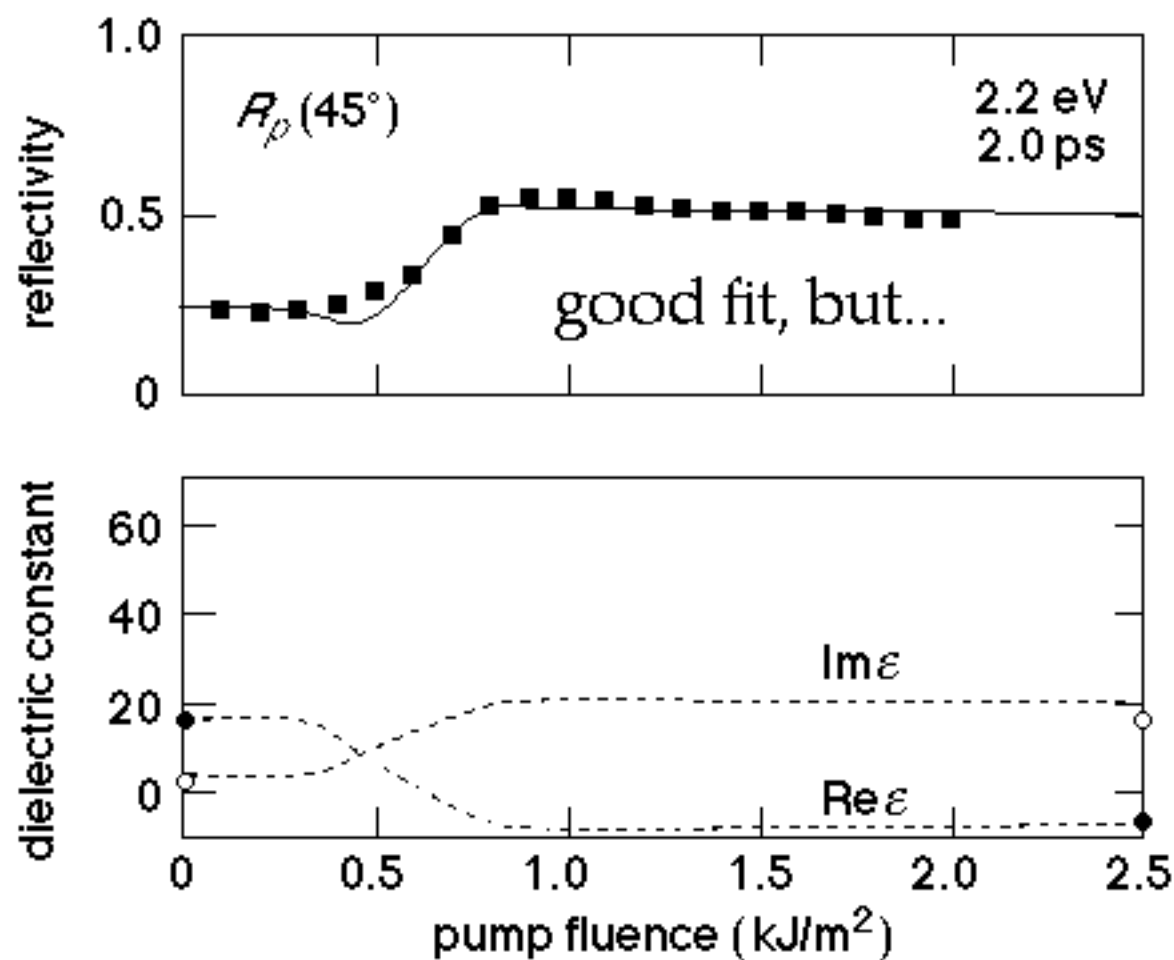


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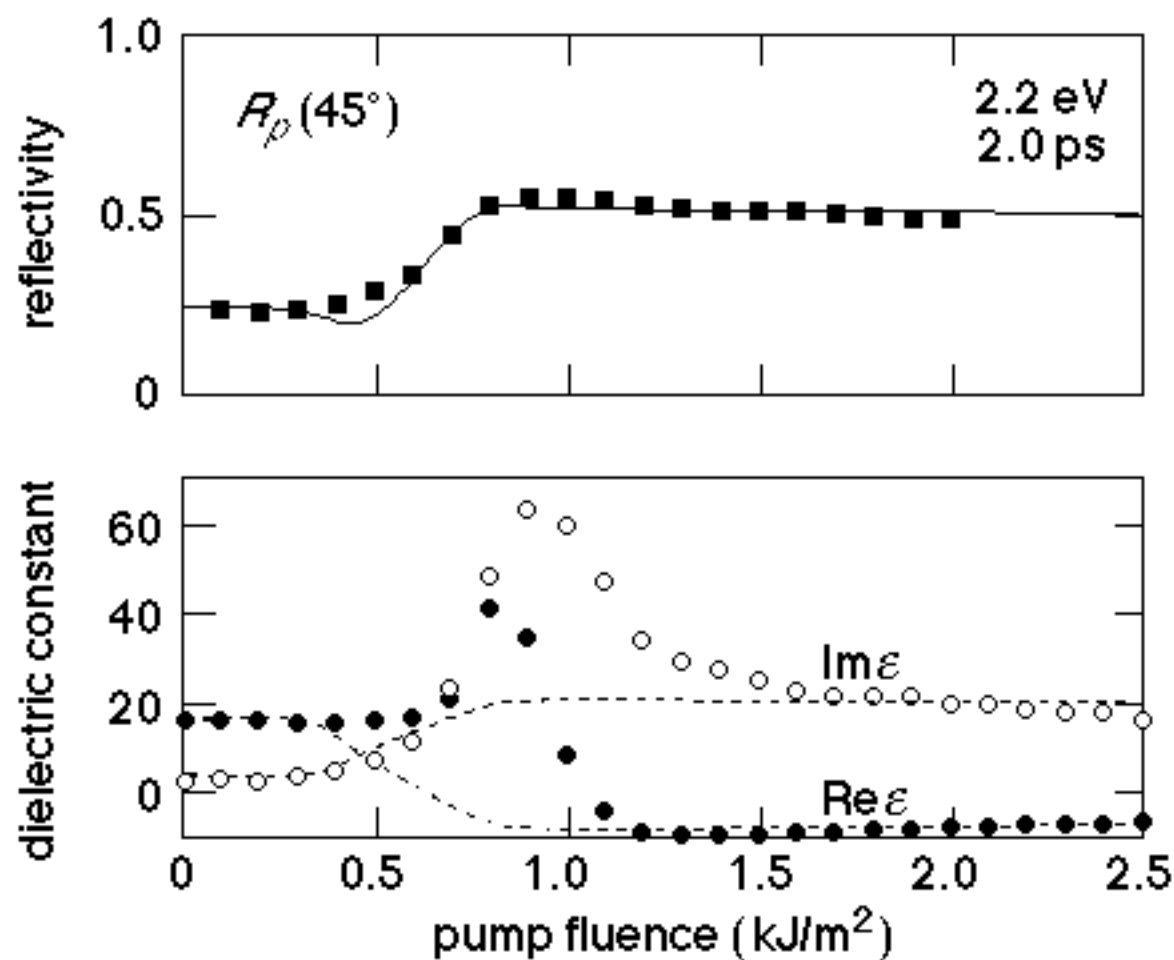




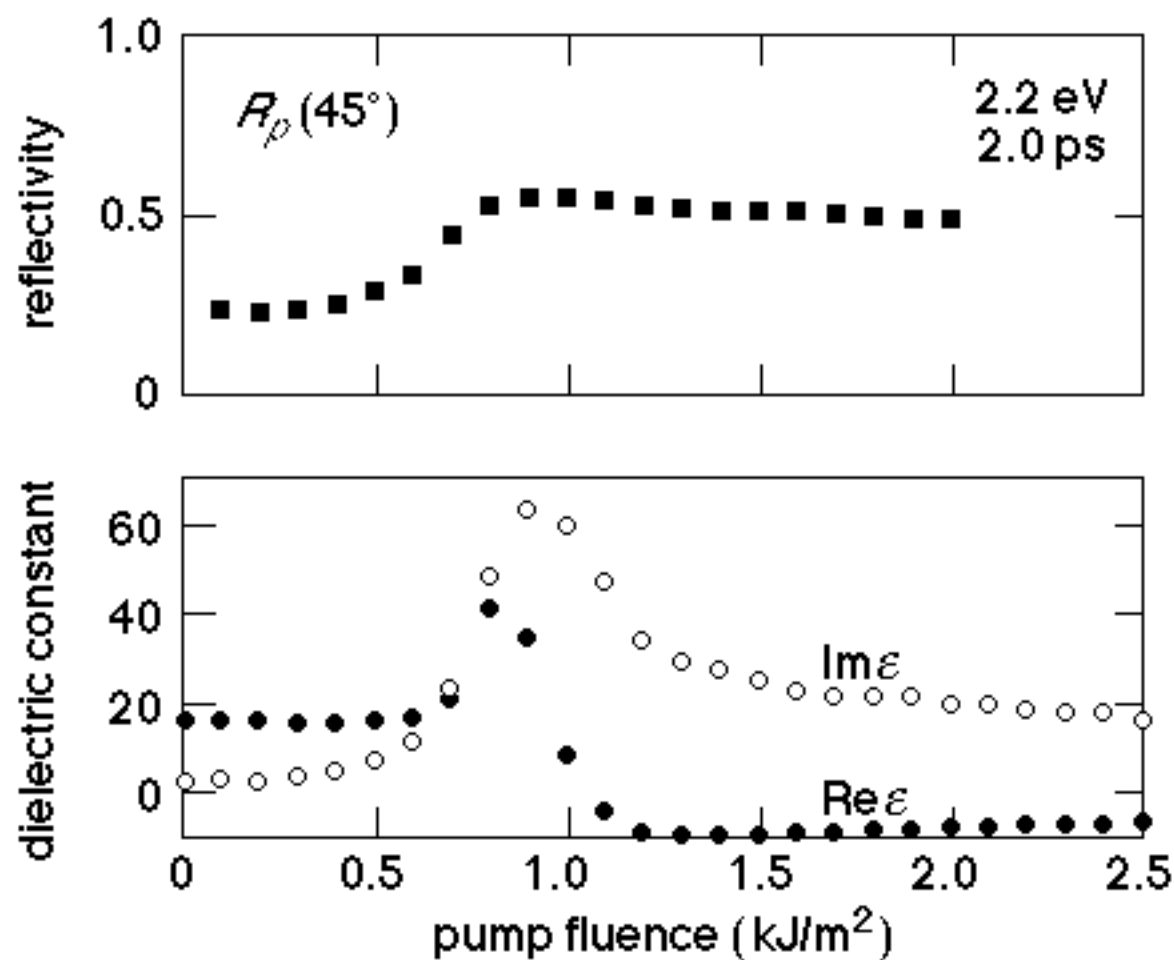
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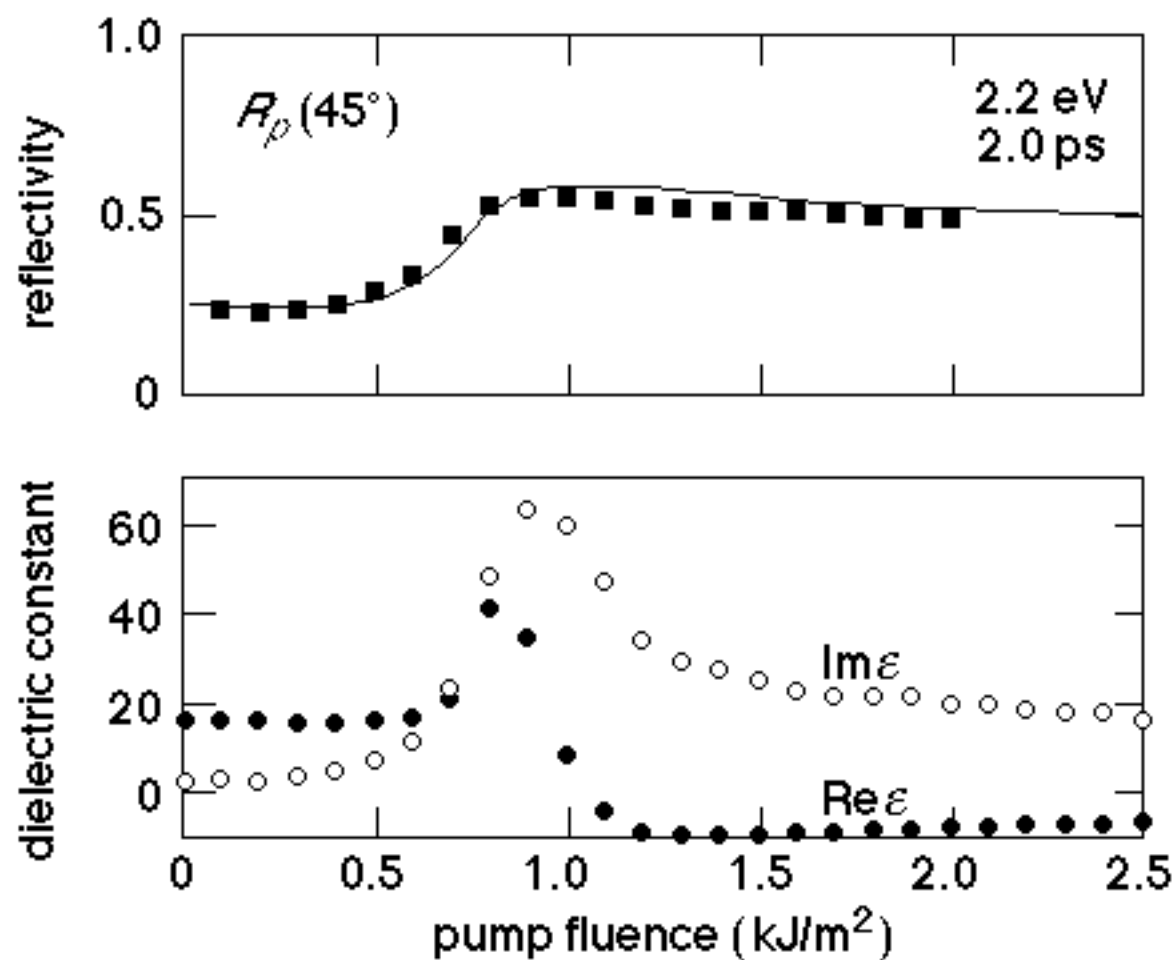
# ***SIMPLE MODEL***



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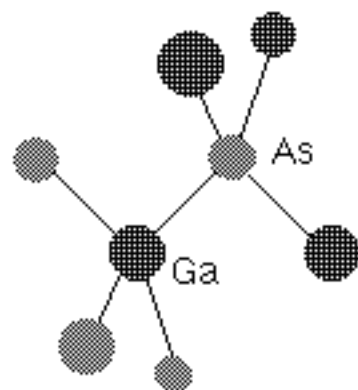


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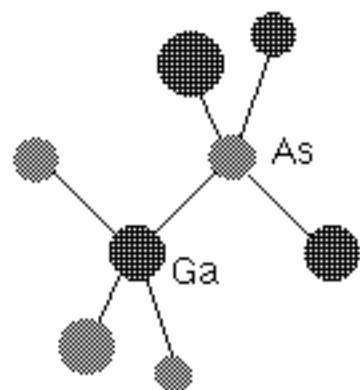
# WHAT IS HAPPENING?

structure



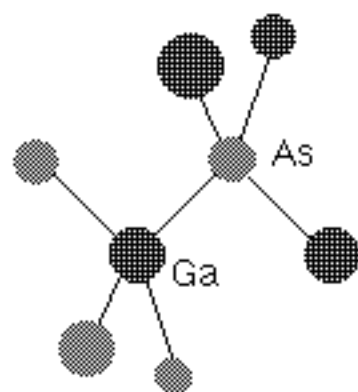
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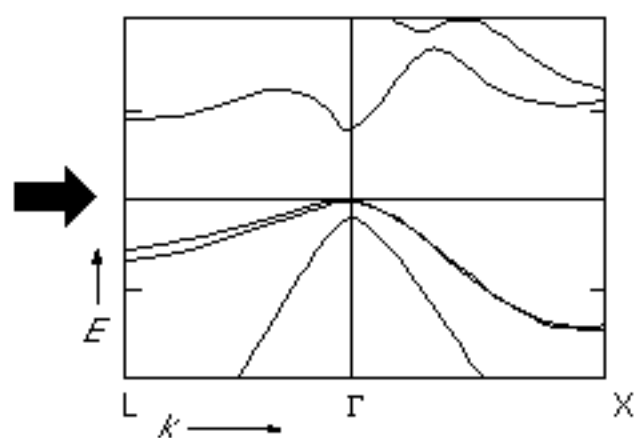


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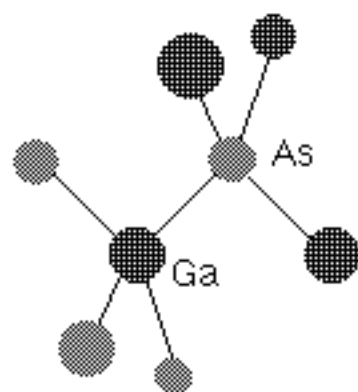


bandstructure

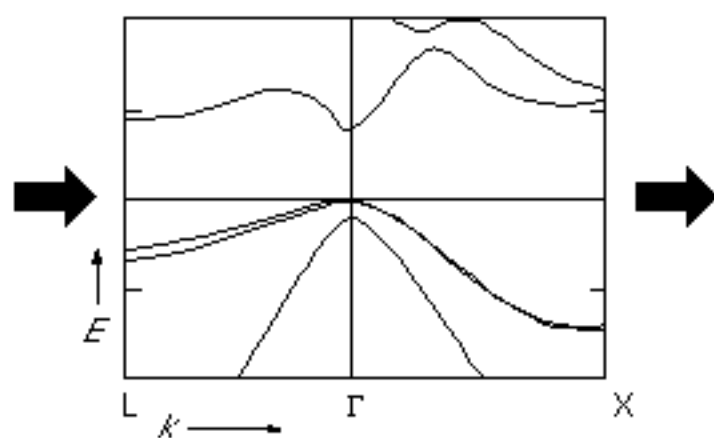


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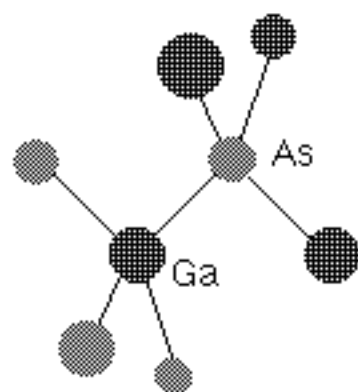
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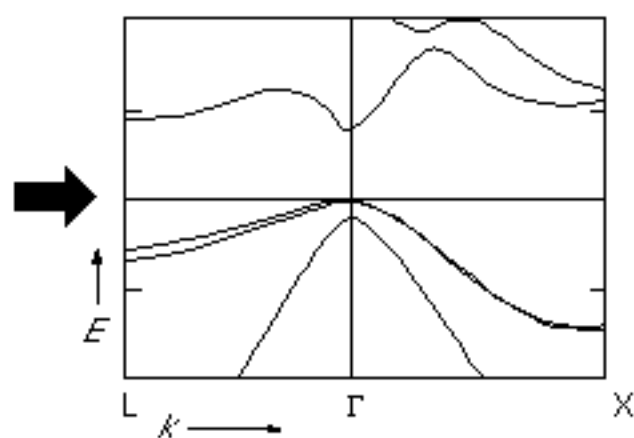


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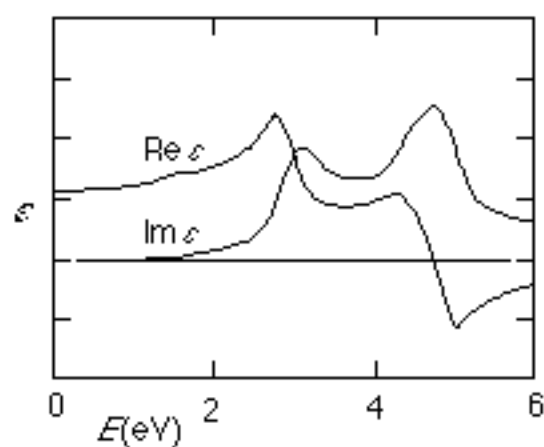
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bandstructure

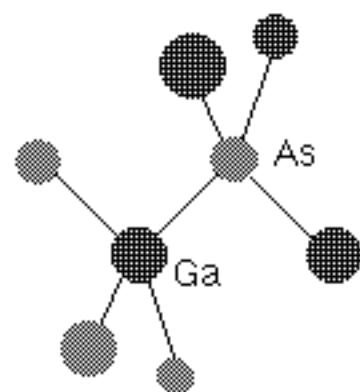


dielectric function

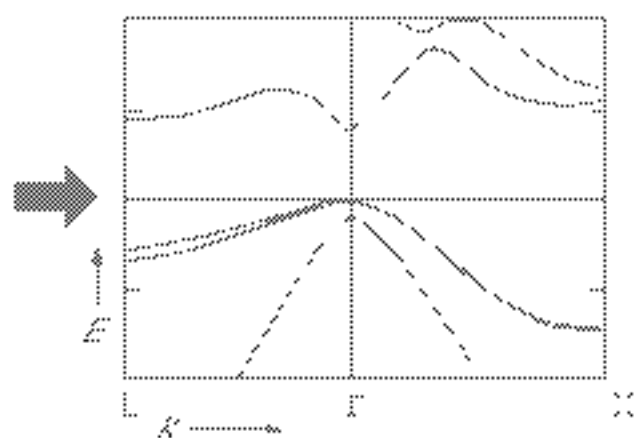


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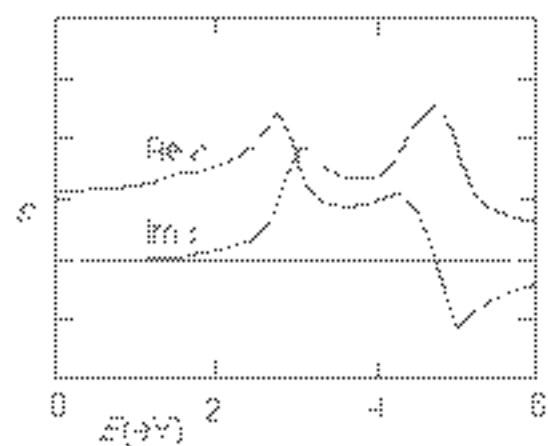
structure



bandstructure

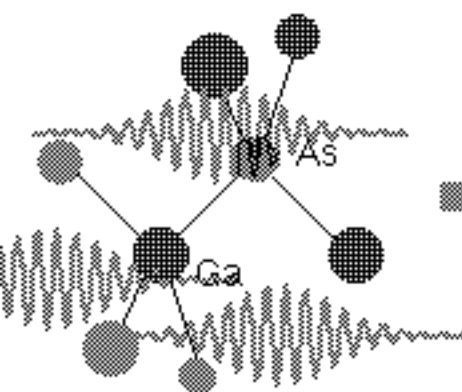


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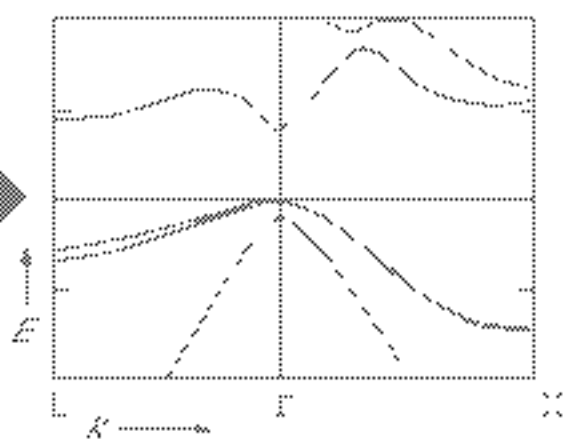


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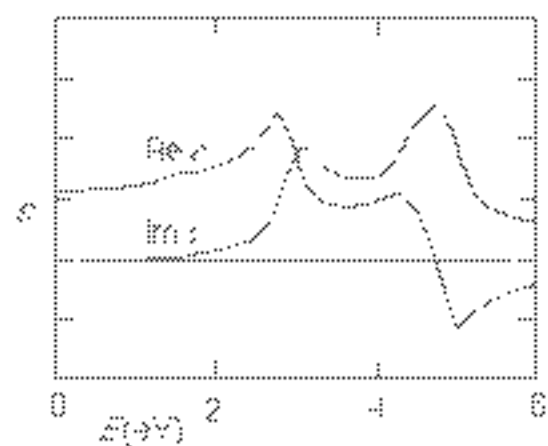
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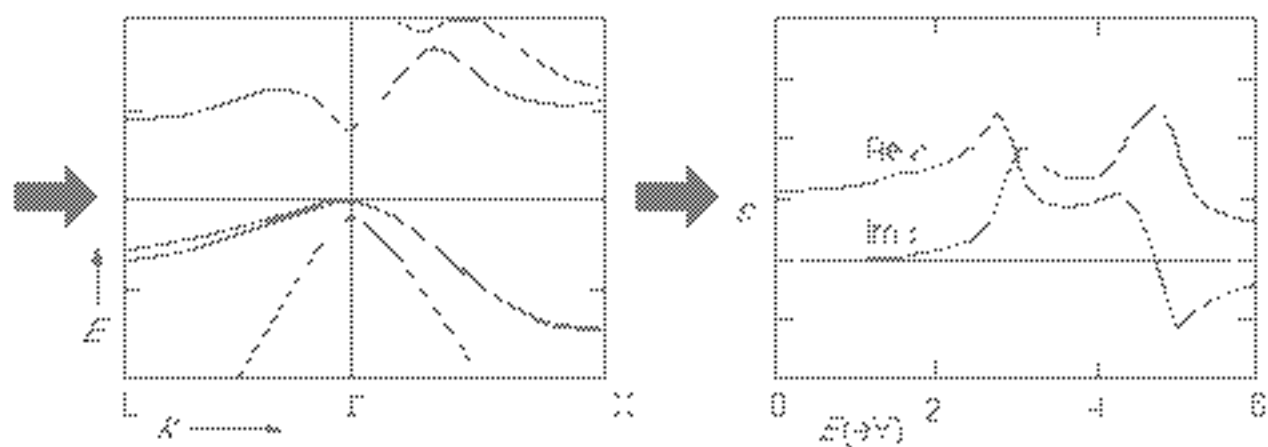
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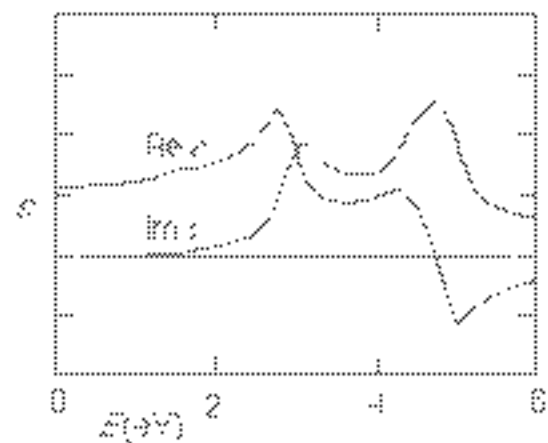
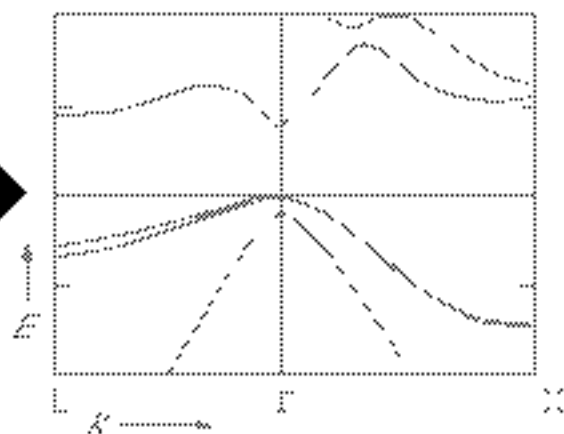
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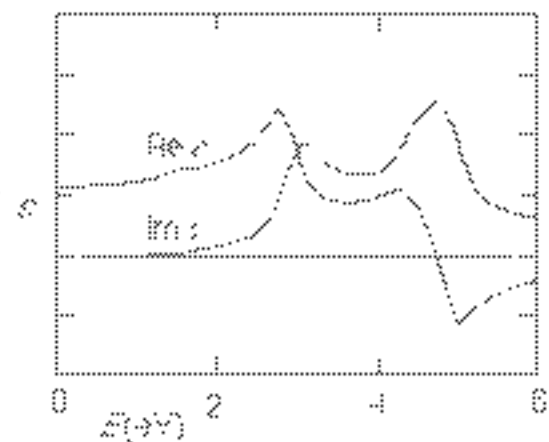
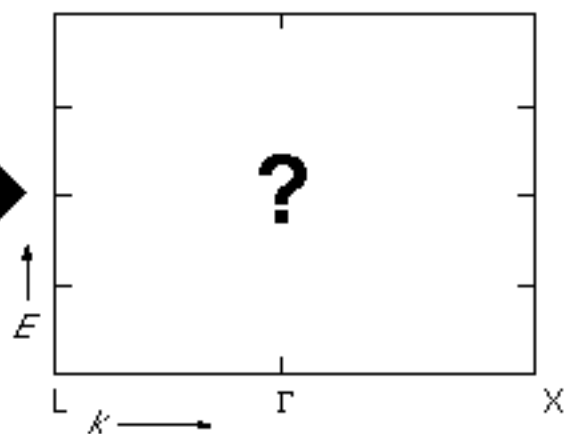


bandstructure

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dielectric function



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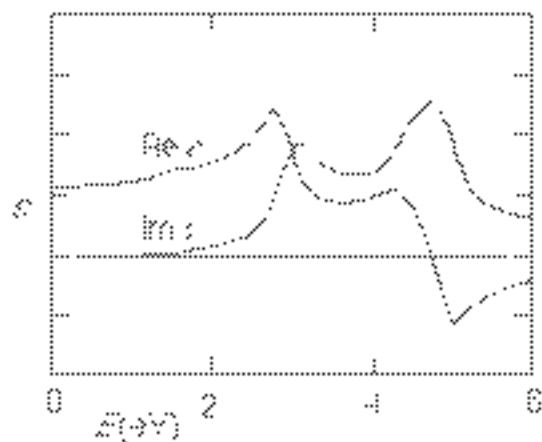
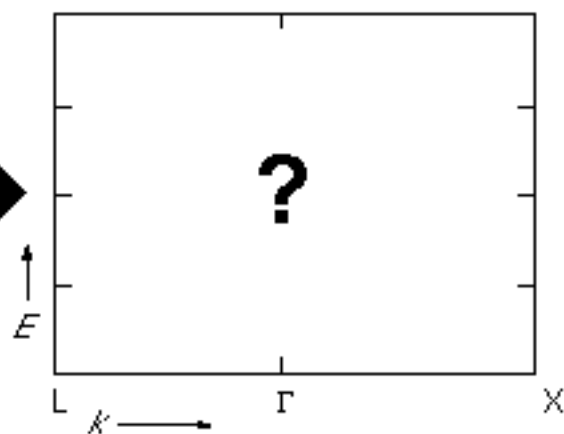


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dielectric function



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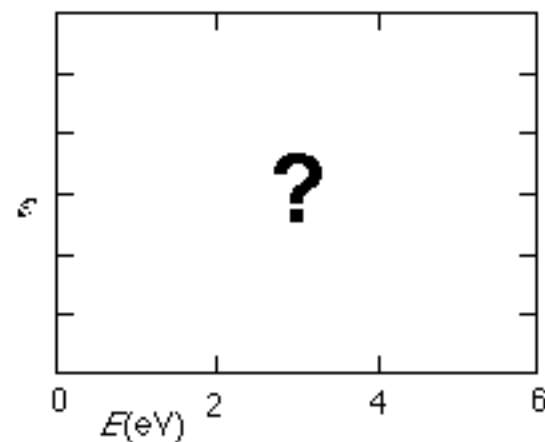
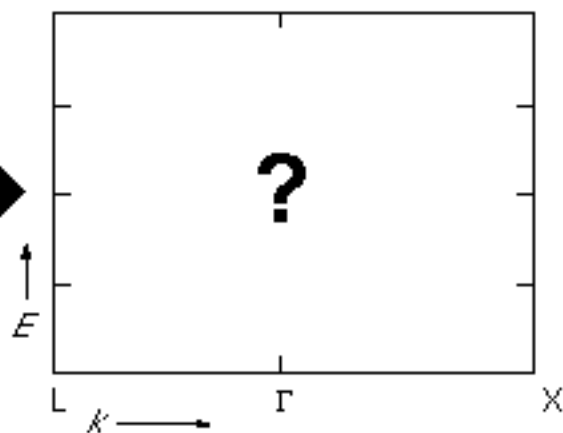
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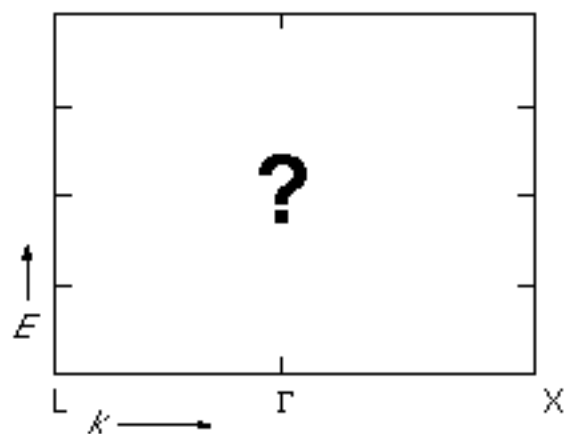


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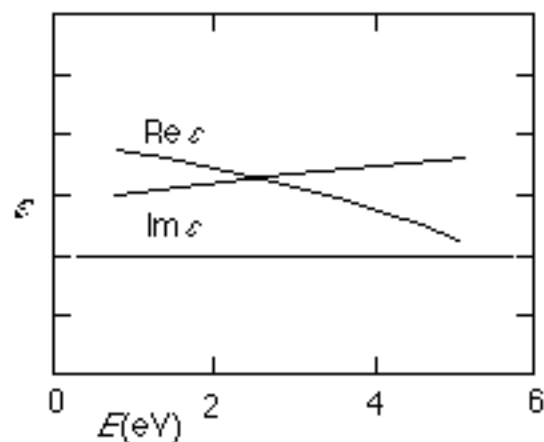
structure

?

bandstructure



dielectric function

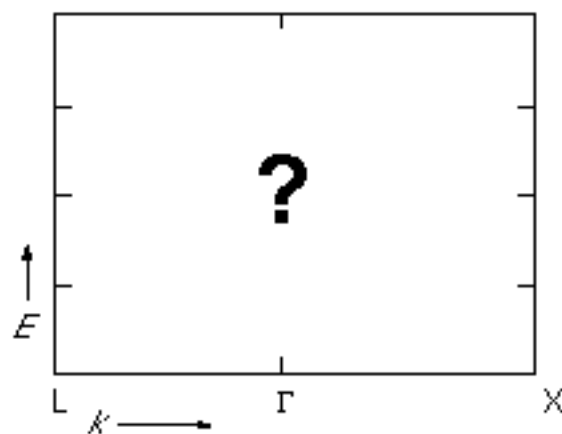


# WHAT IS HAPPENING?

structure

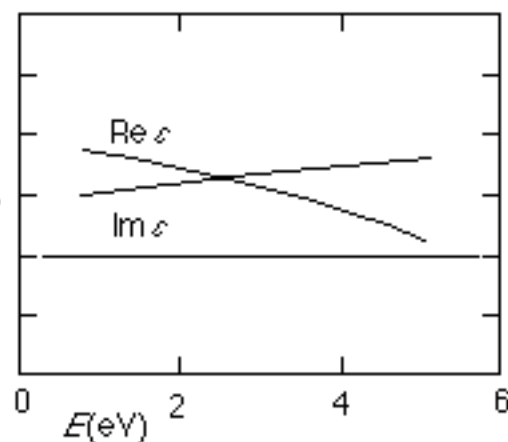
?

bandstructure



dielectric function

$\epsilon$



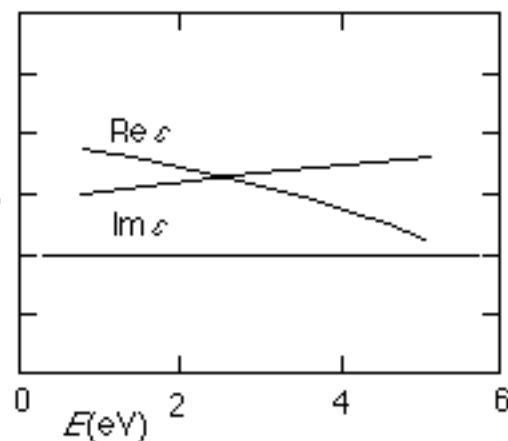
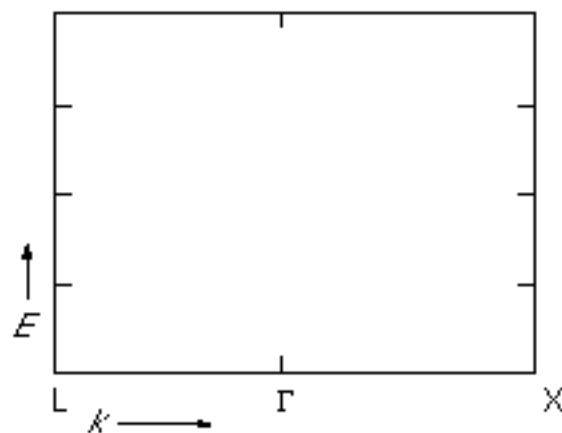
# WHAT IS HAPPENING?

structure

bandstructure

dielectric function

?



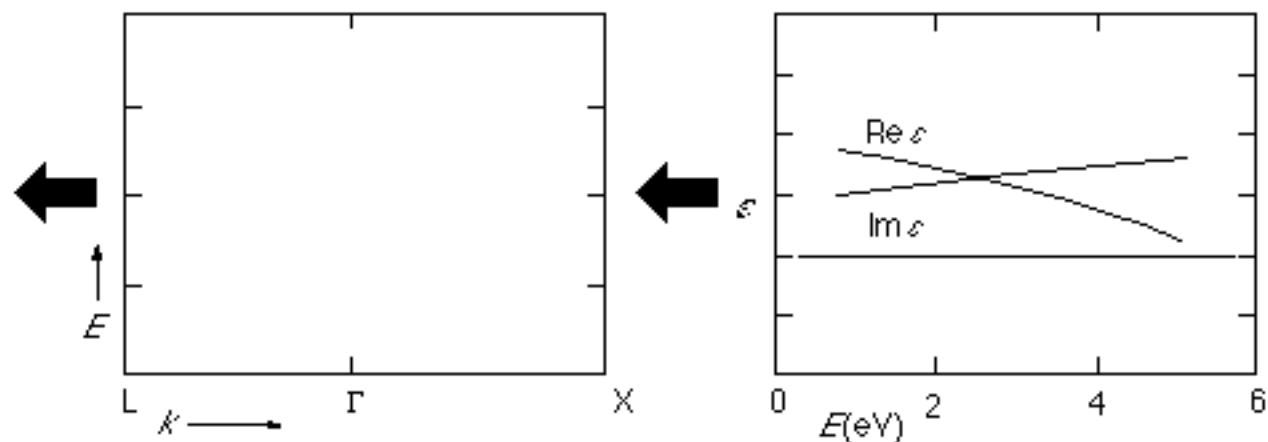
# WHAT IS HAPPENING?

structure

bandstructure

dielectric function

?

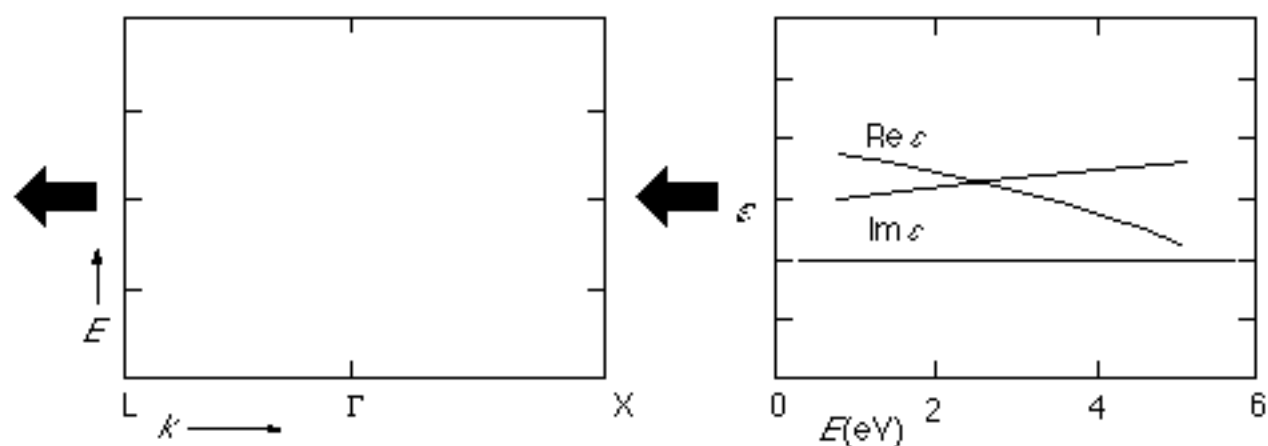


# WHAT IS HAPPENING?

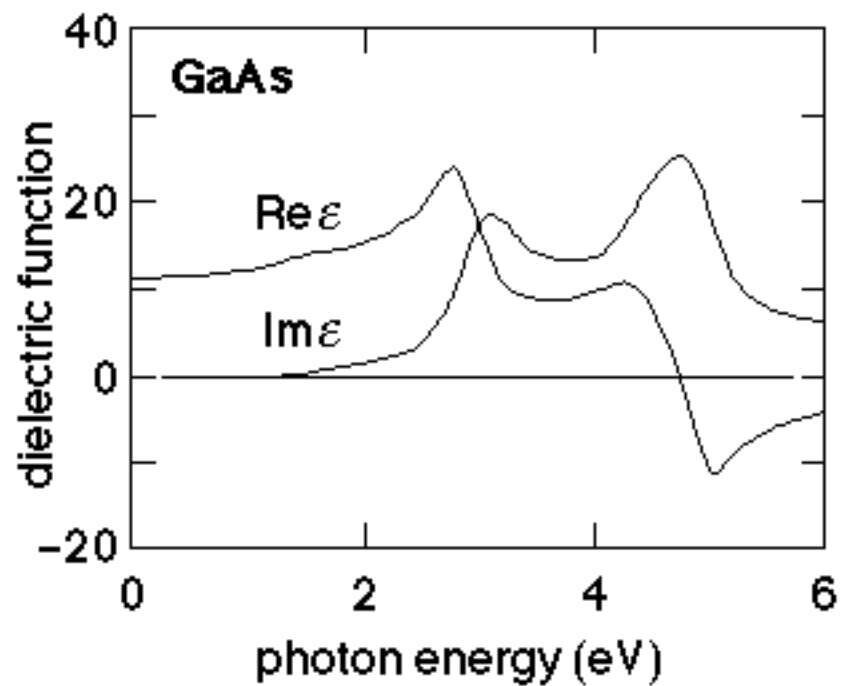
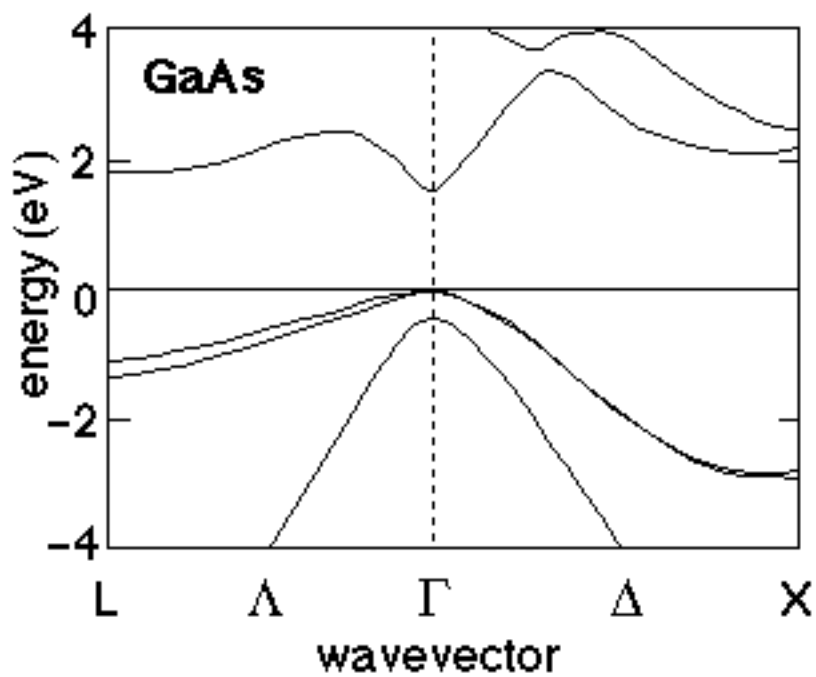
structure

bandstructure

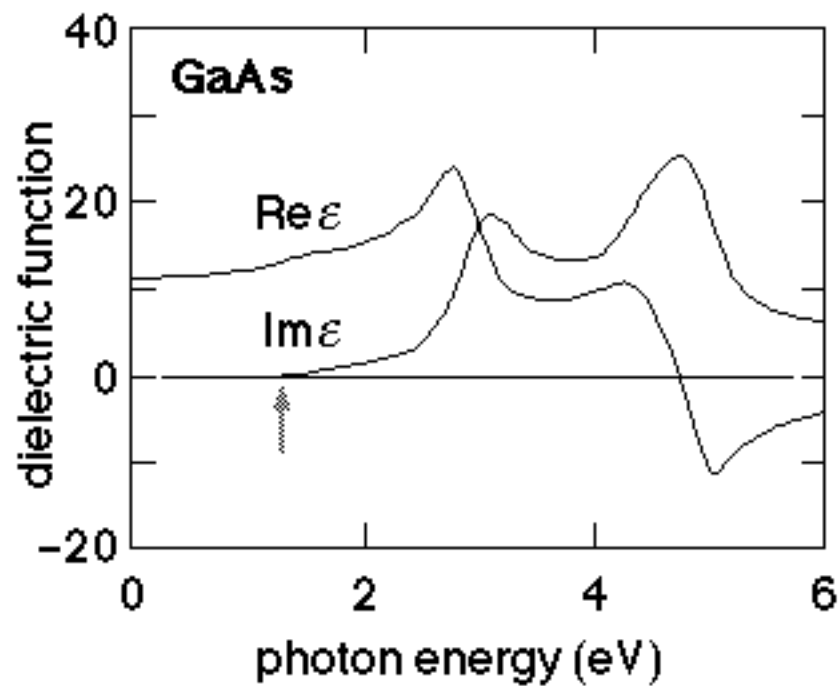
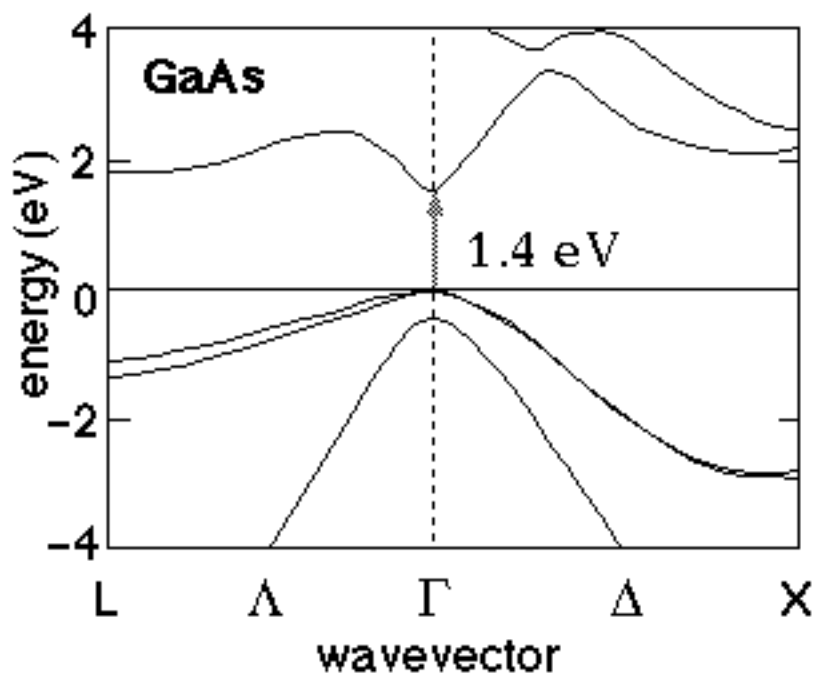
dielectric function



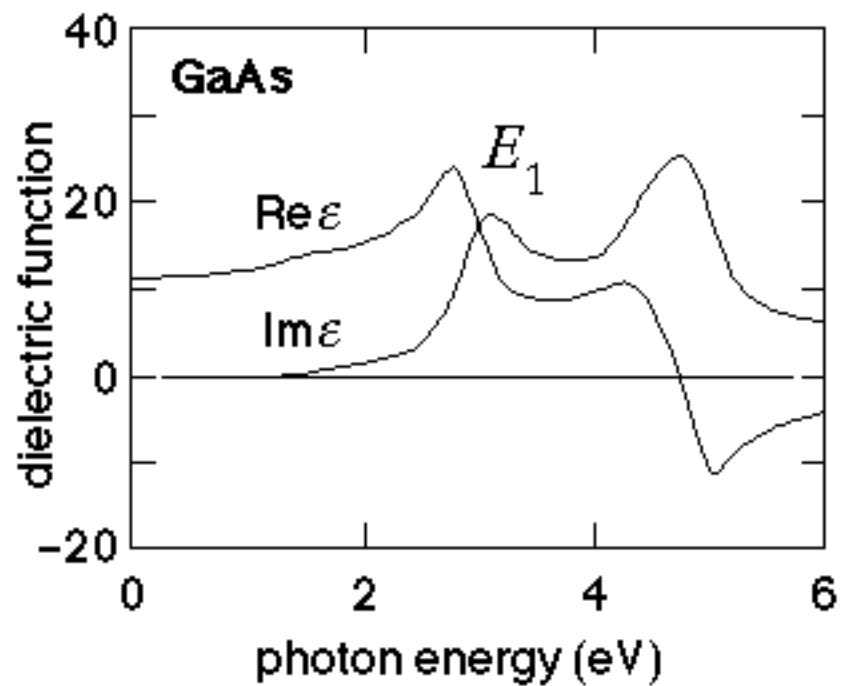
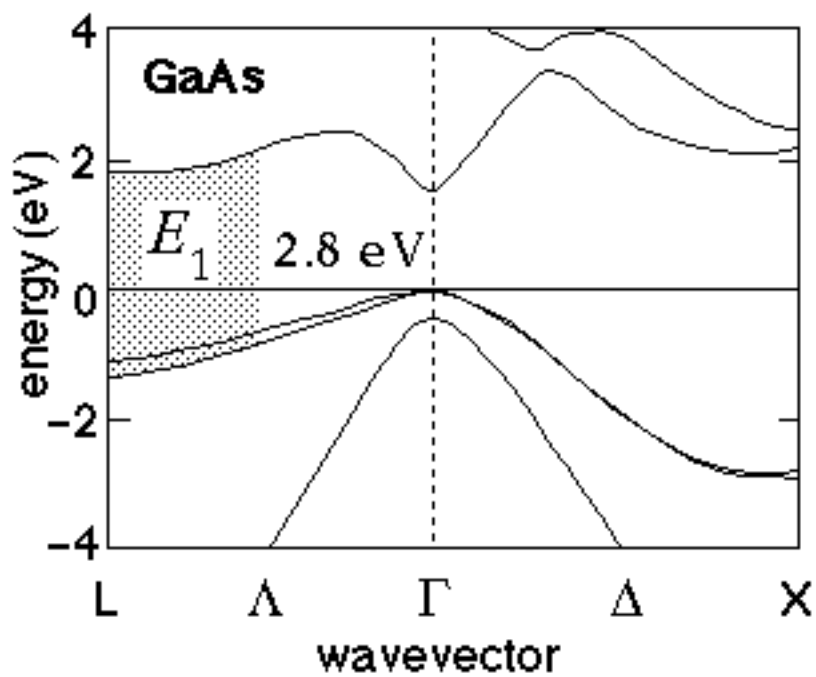
# *GaAs BANDSTRUCTURE*



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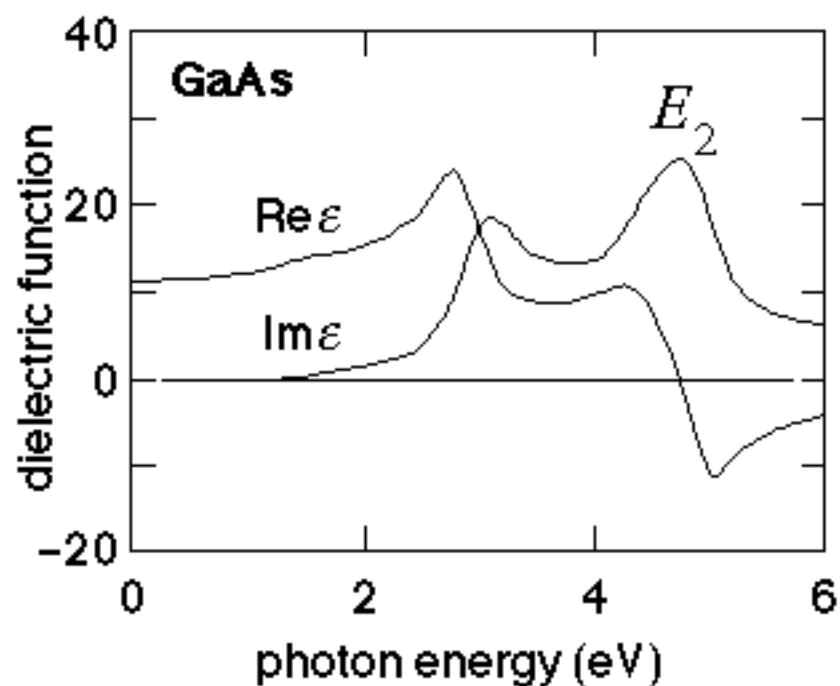
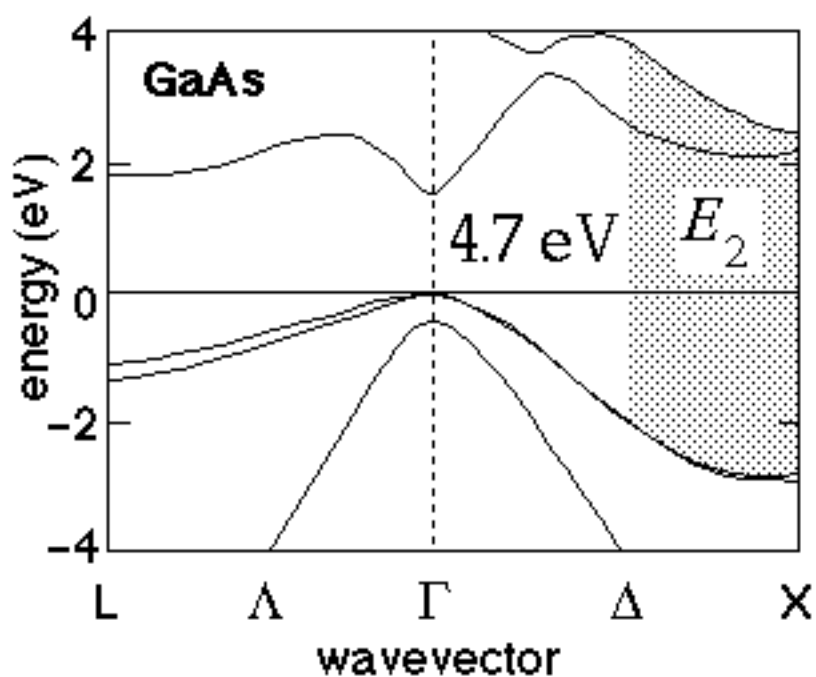


# GaAs BANDSTRUCTURE

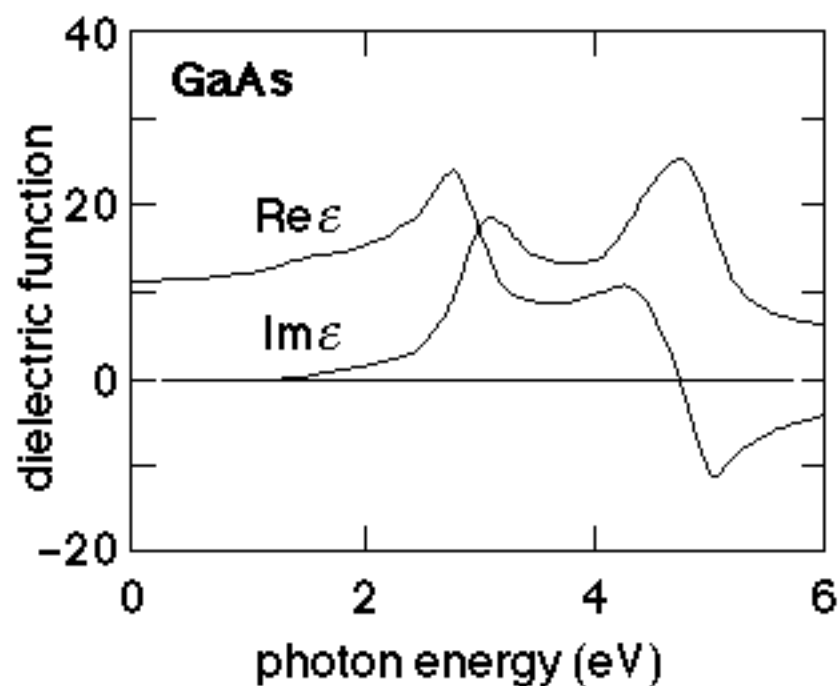
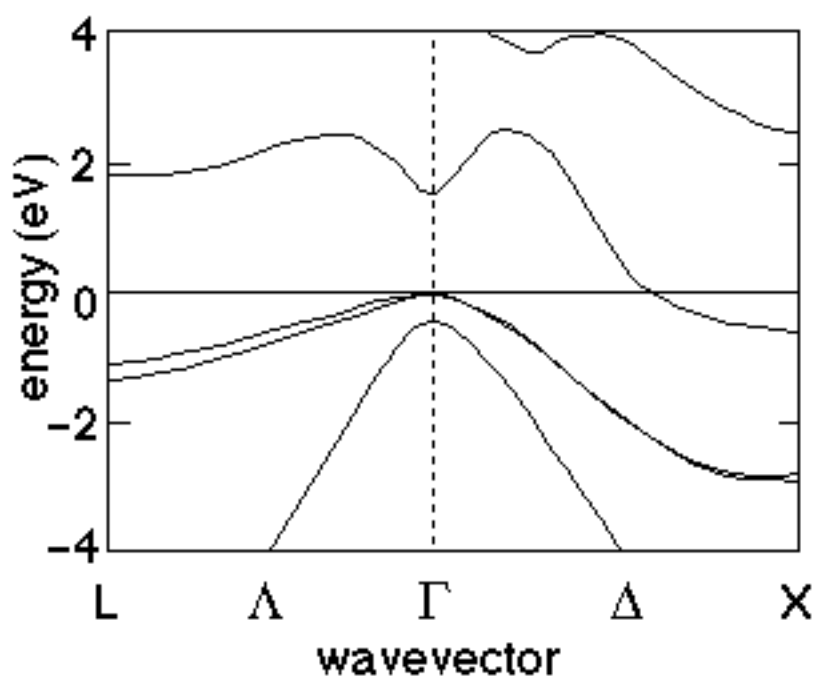




# *GaAs BANDSTRUCTURE*



# GaAs BANDSTRUCTURE

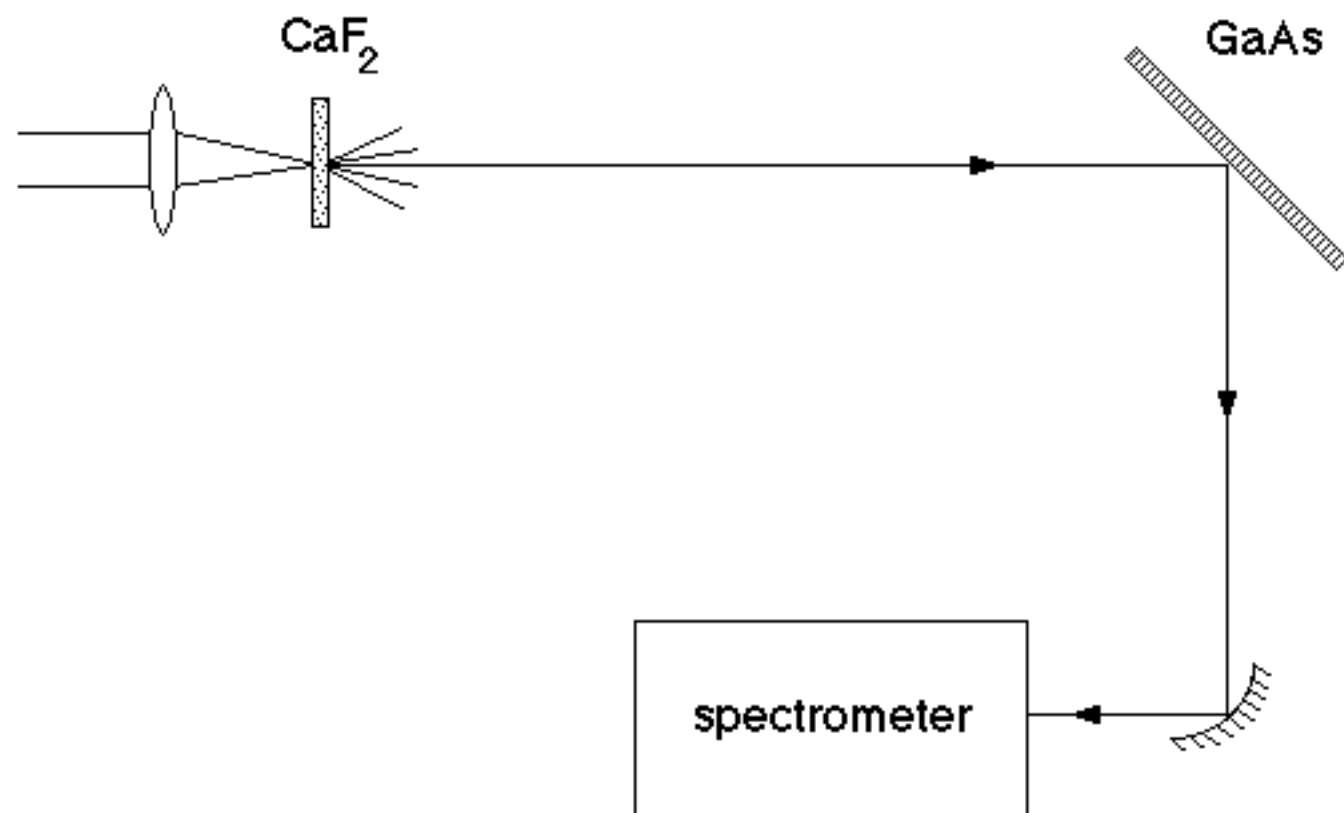


S. Froyen *et al.*, *Phys Rev. B* **28** (1983) 3258

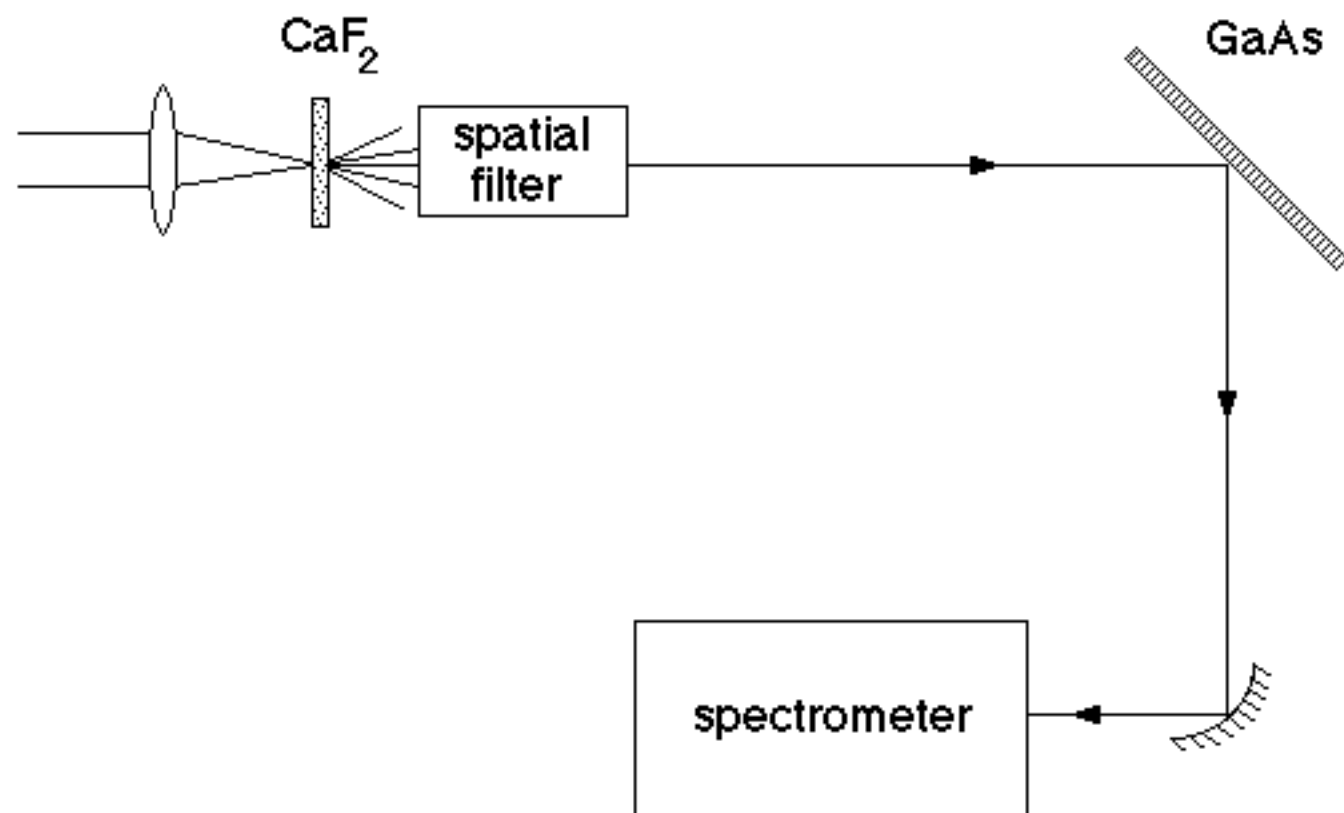
D.H. Kim *et al.*, *Sol. State. Comm.* **89** (1994) 119



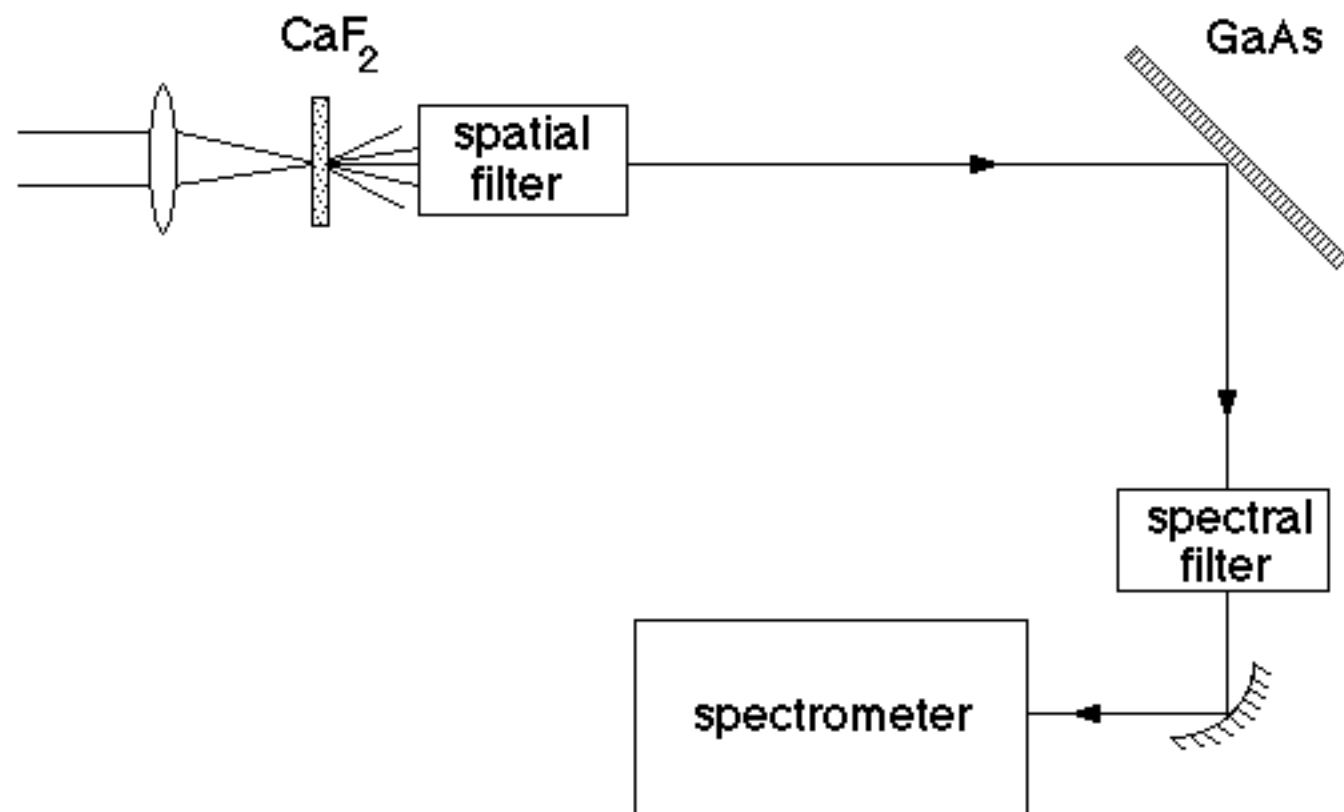
# ***BROADBAND PROBE***



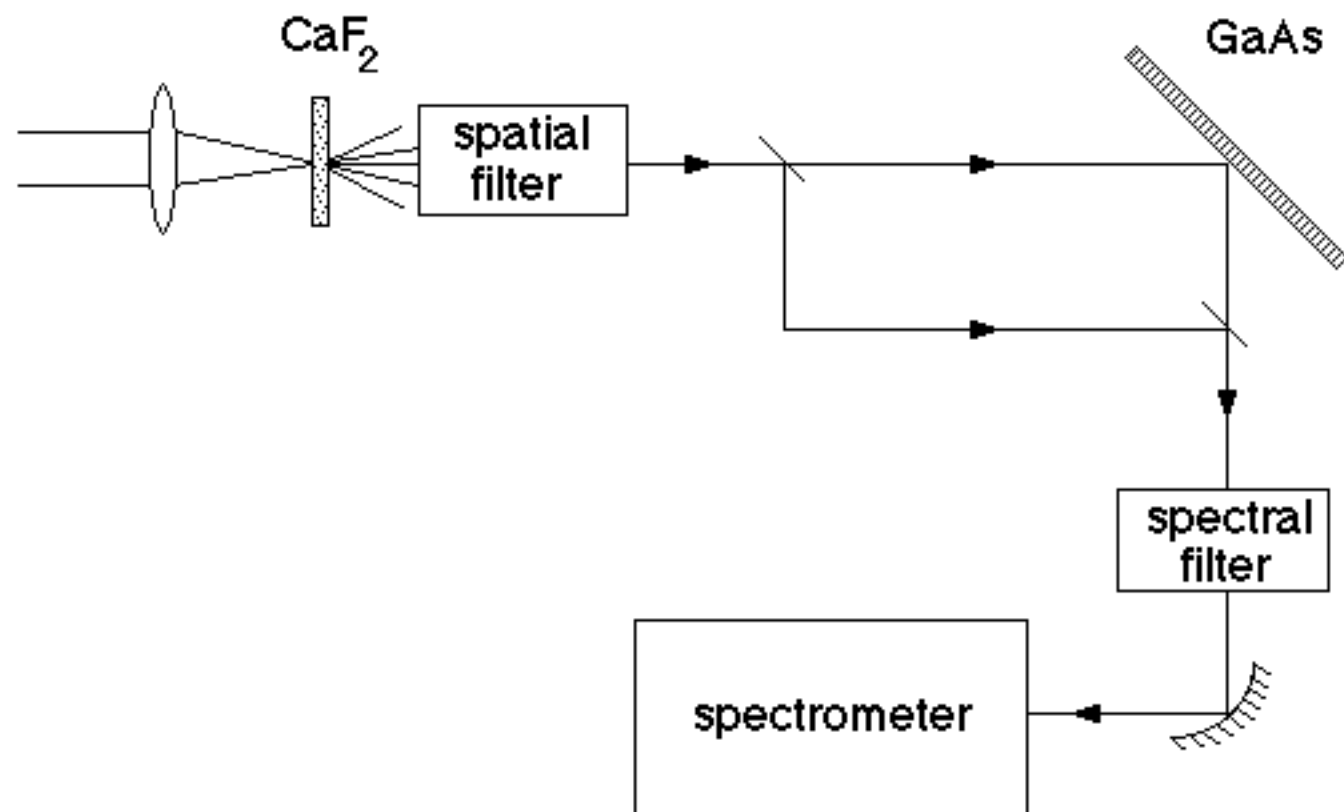
# *BROADBAND PROBE*



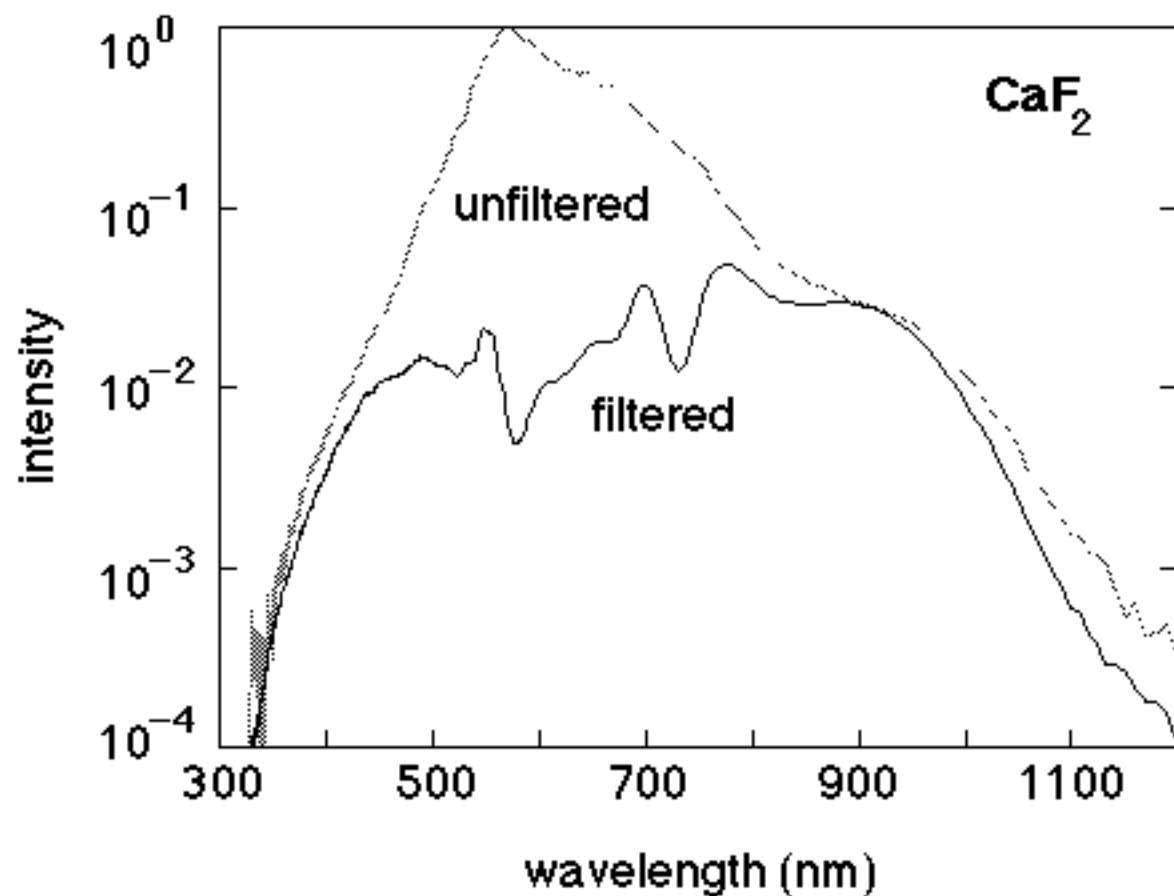
# *BROADBAND PROBE*



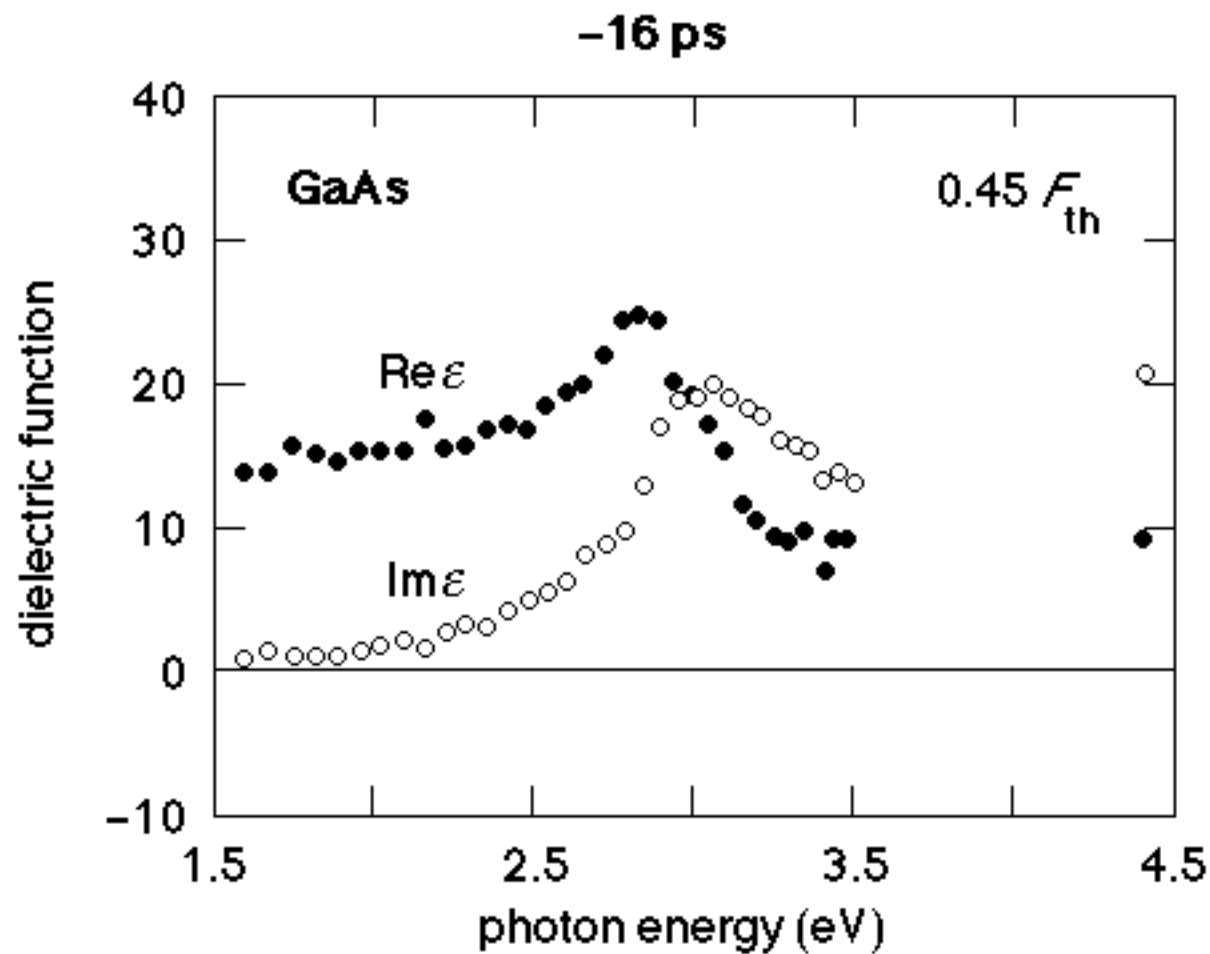
# ***BROADBAND PROBE***



# WHITE LIGHT SPECTRUM

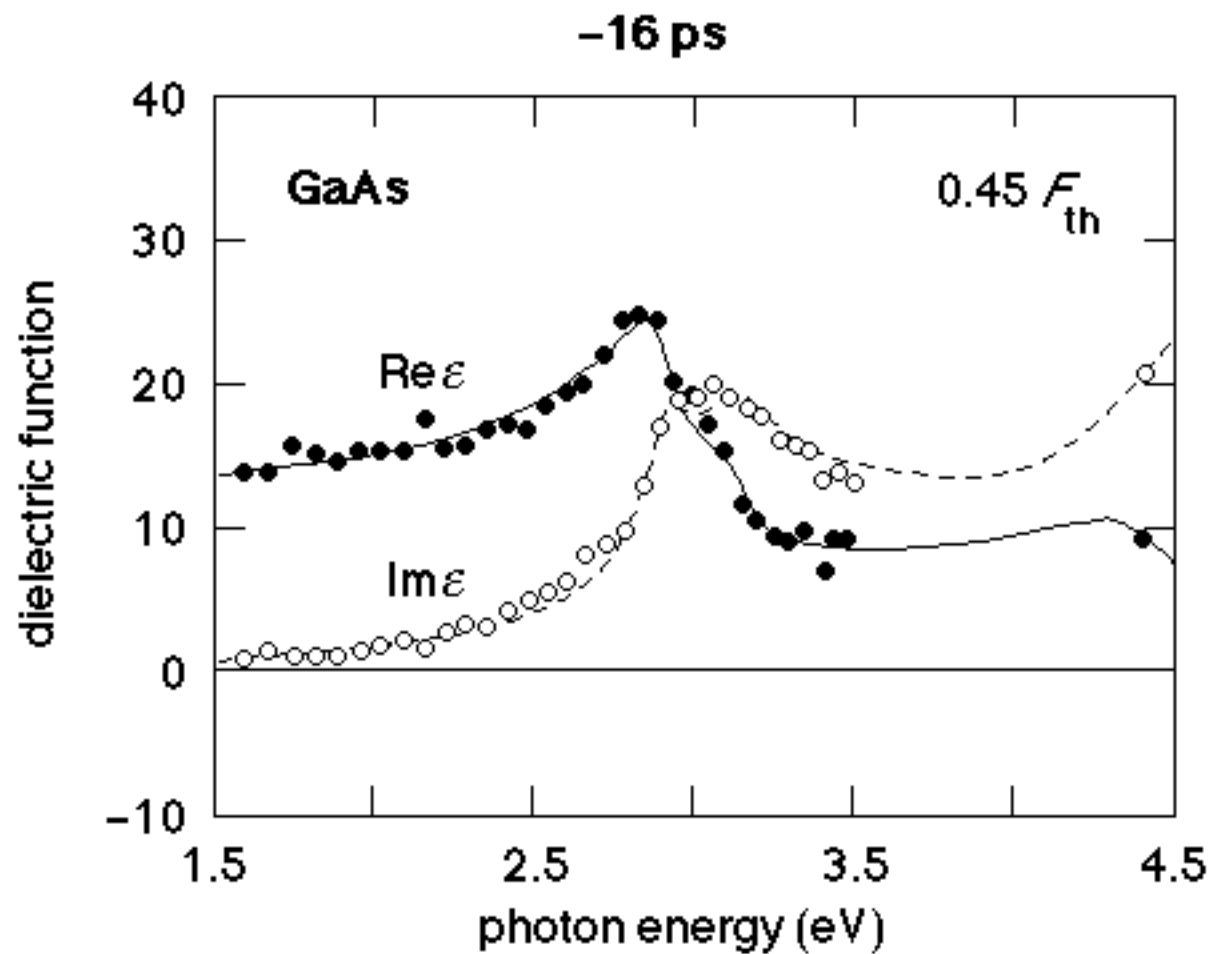


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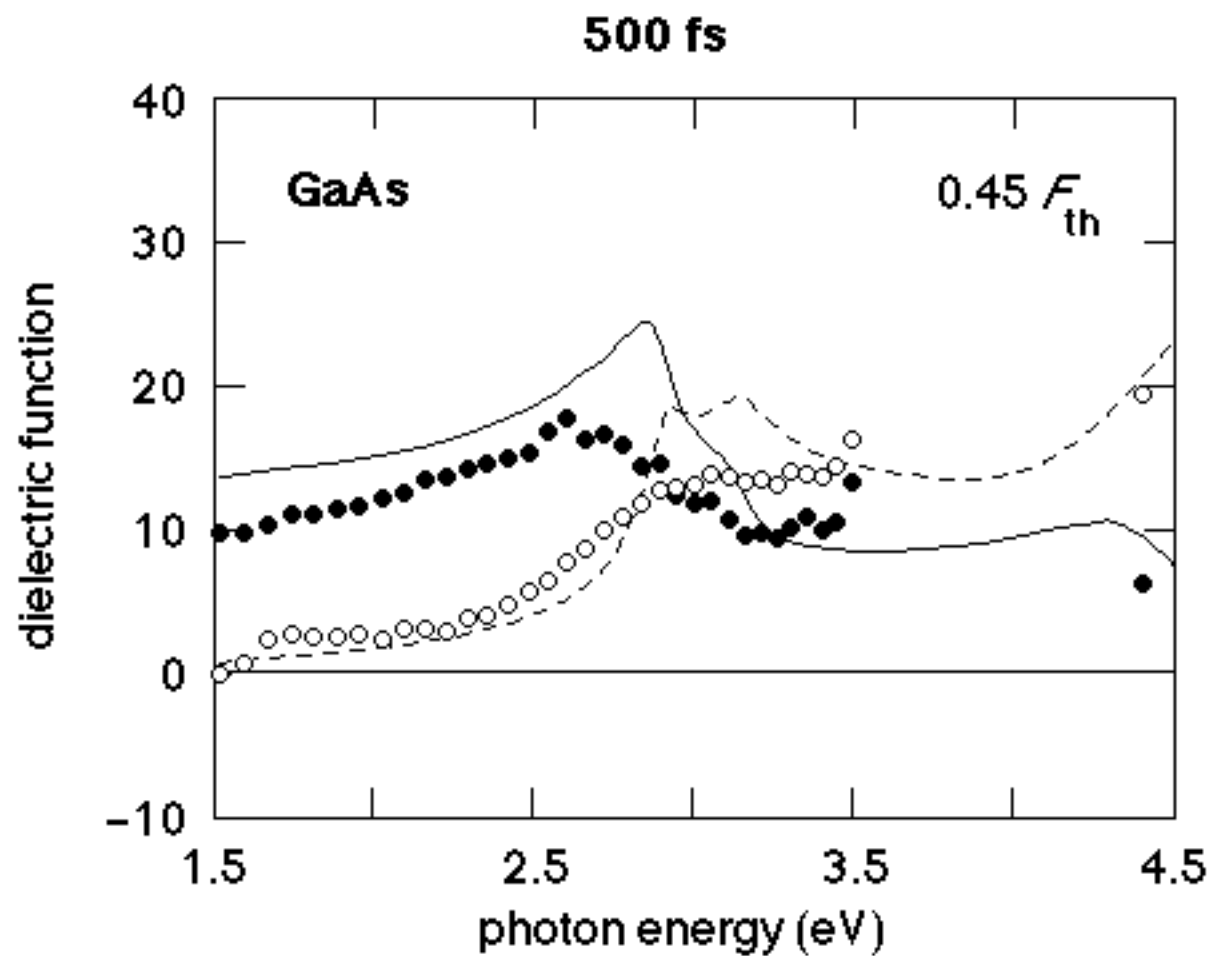




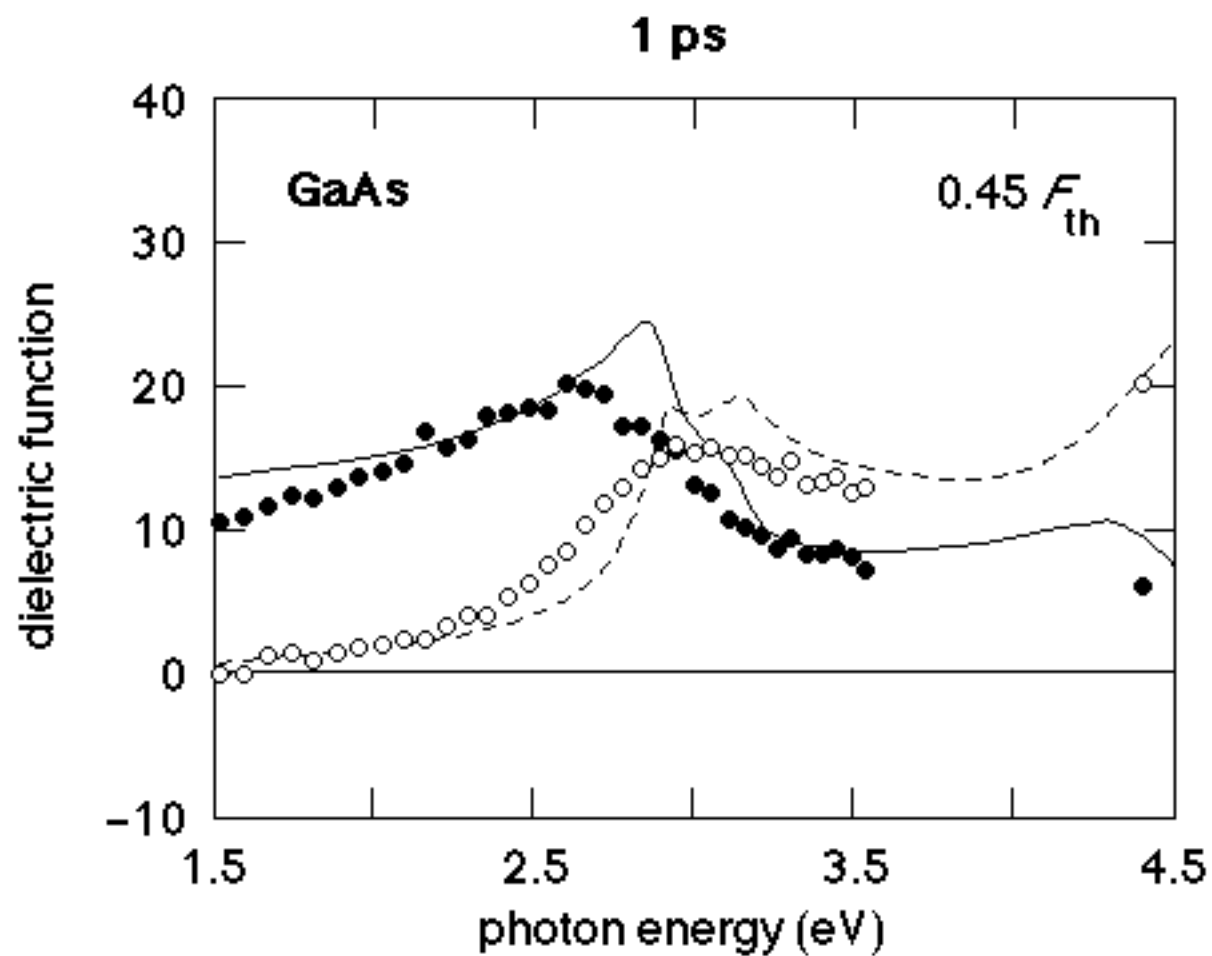
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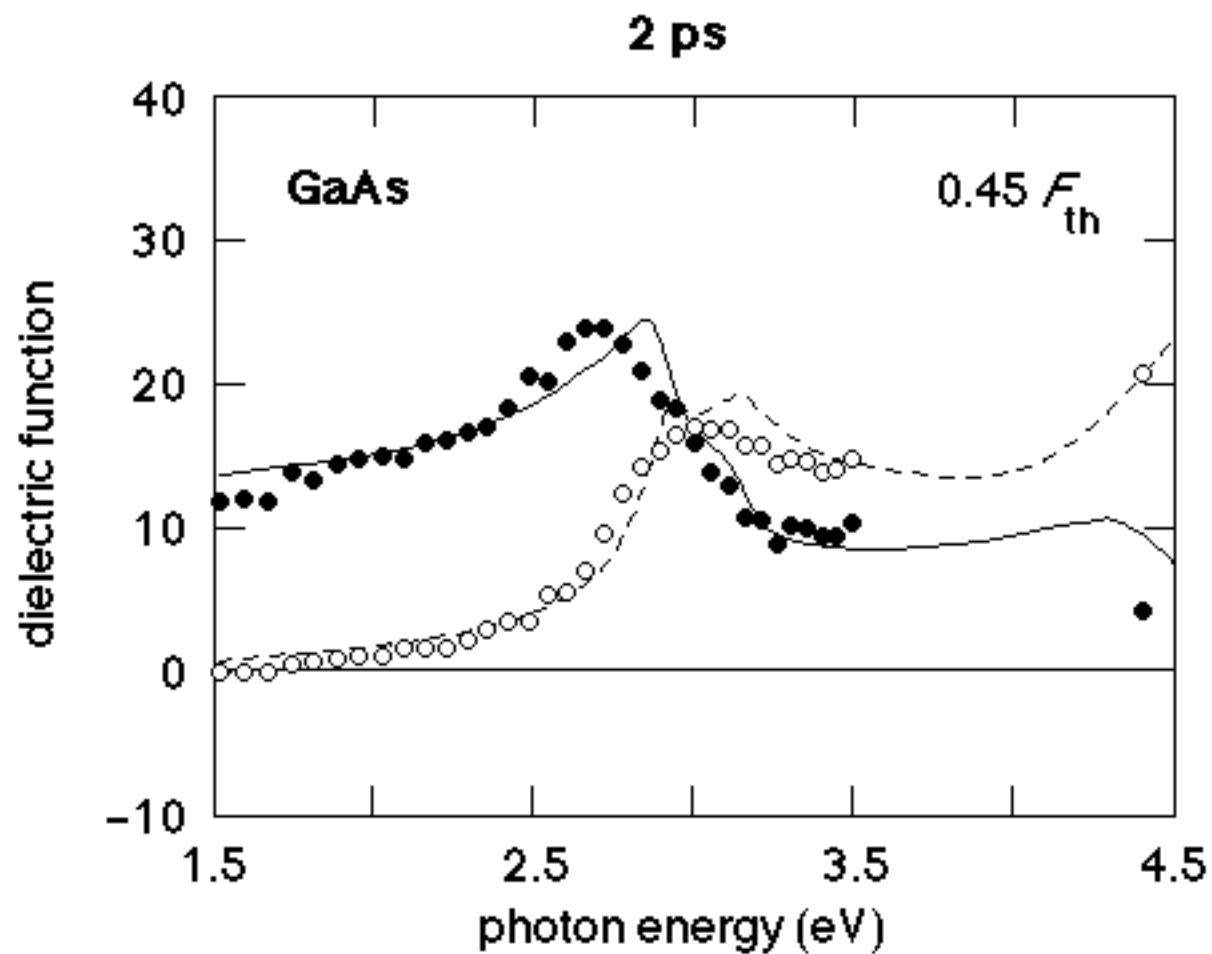
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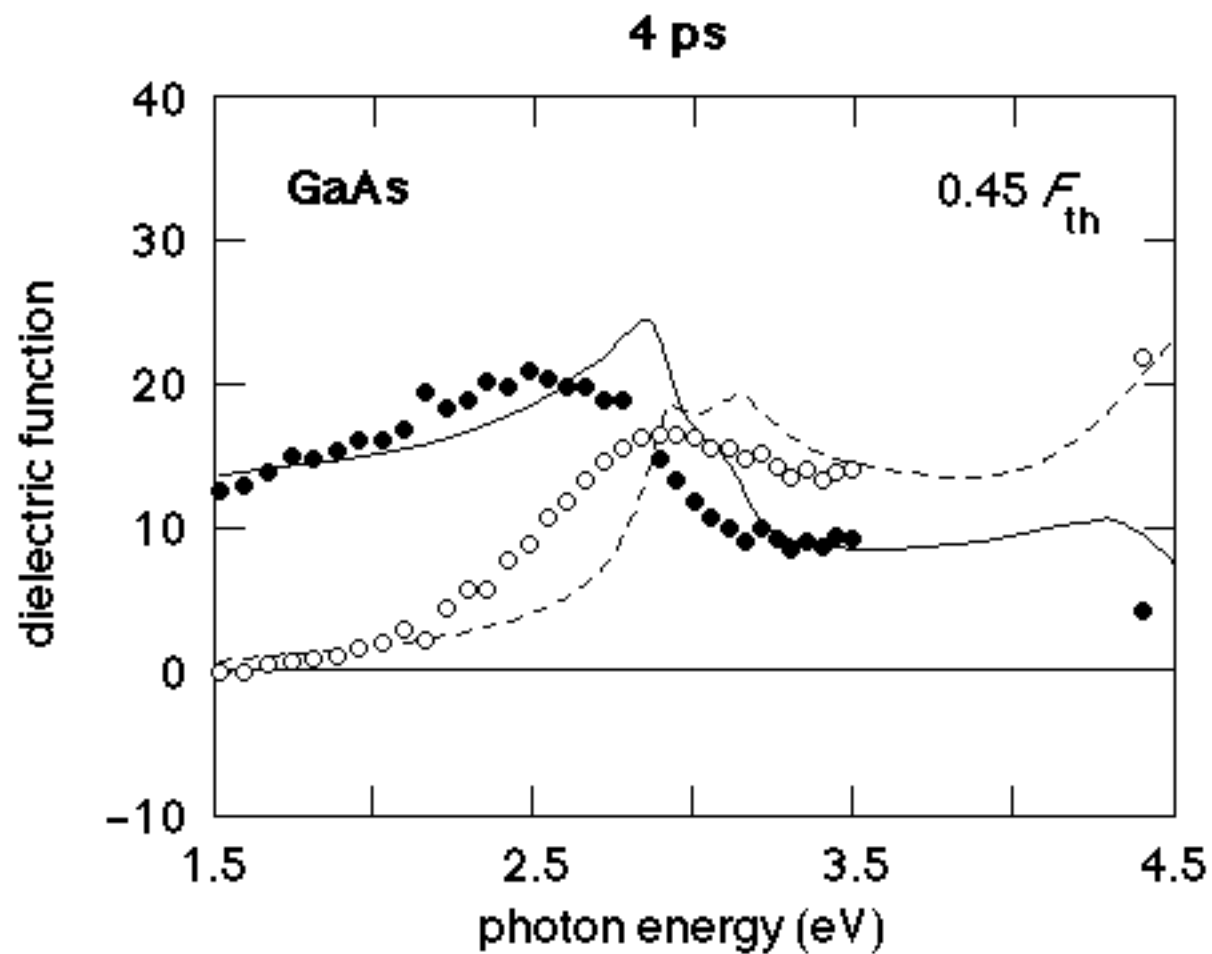
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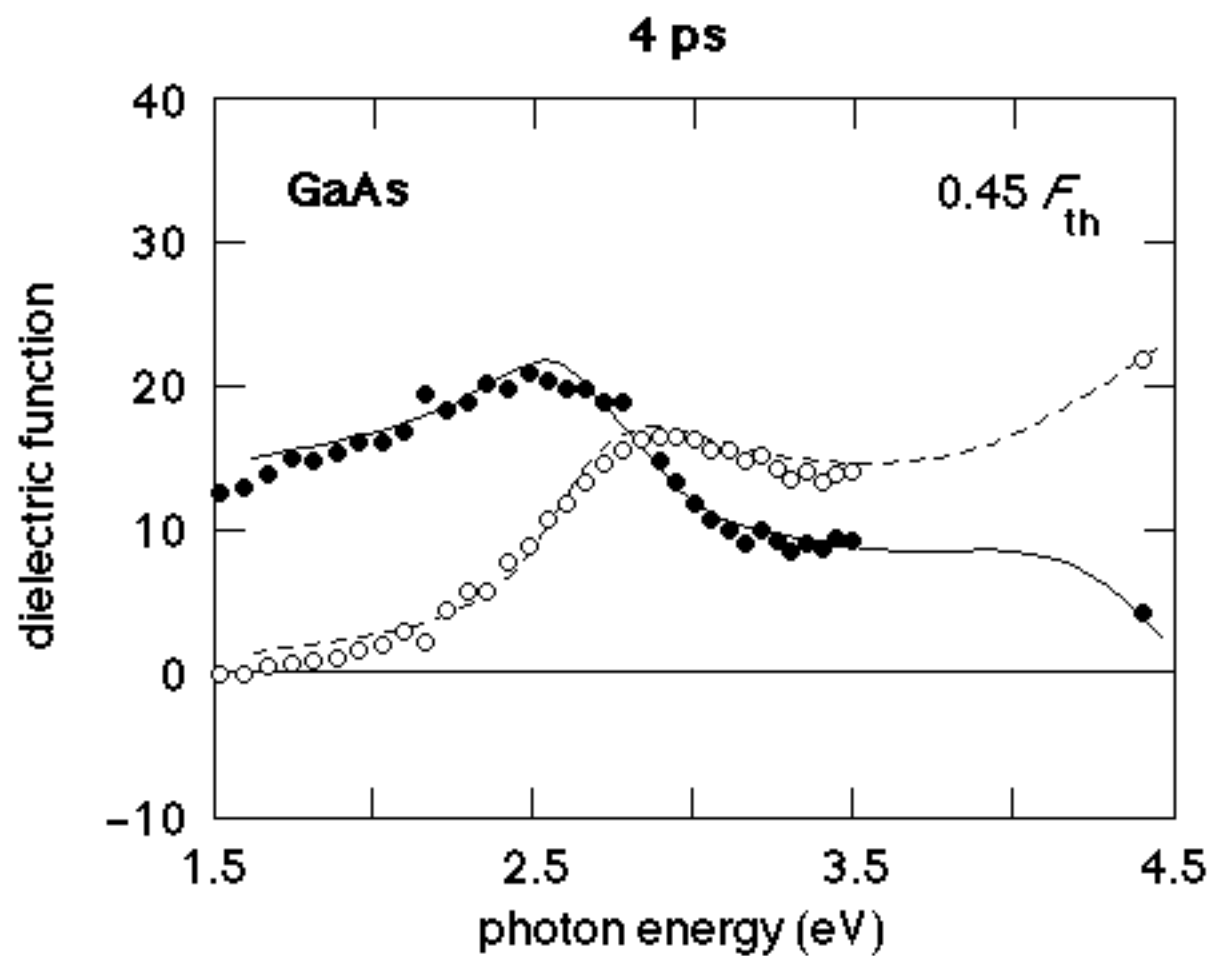
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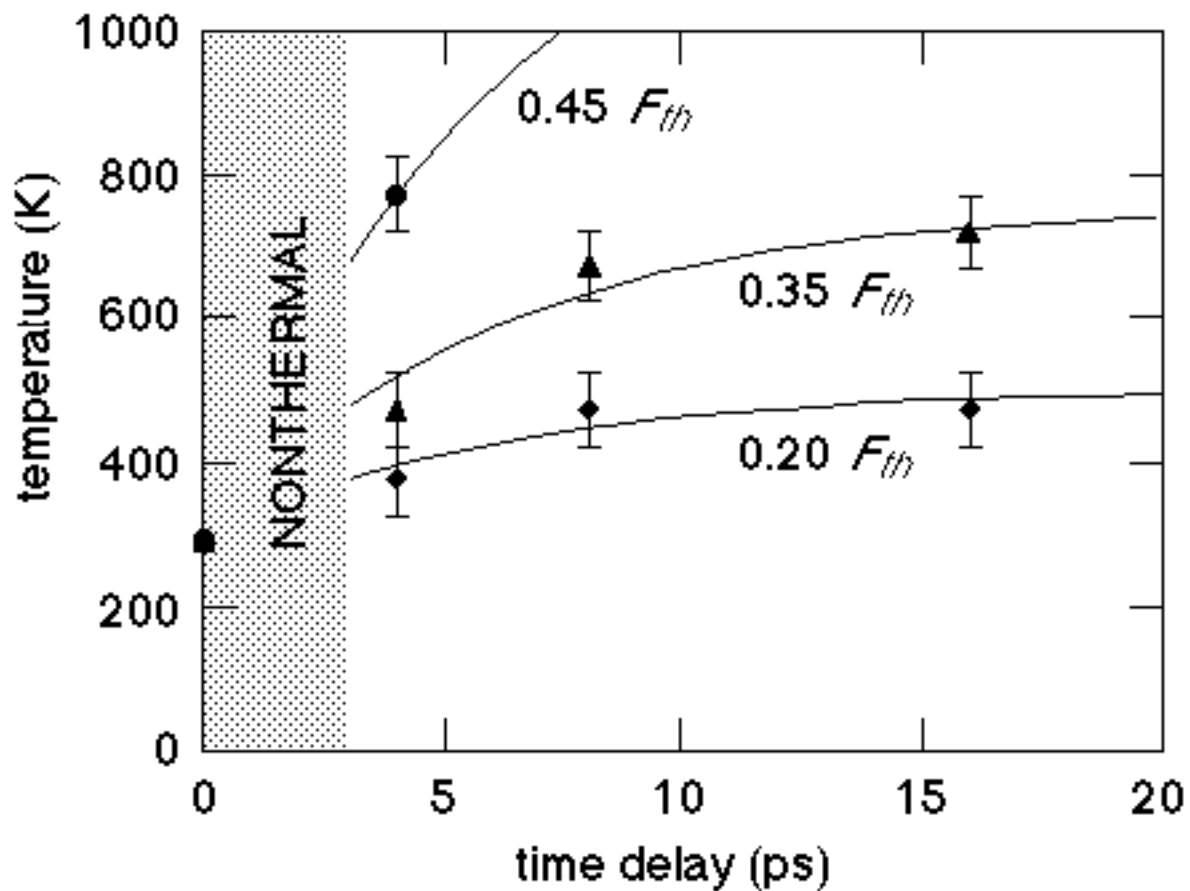
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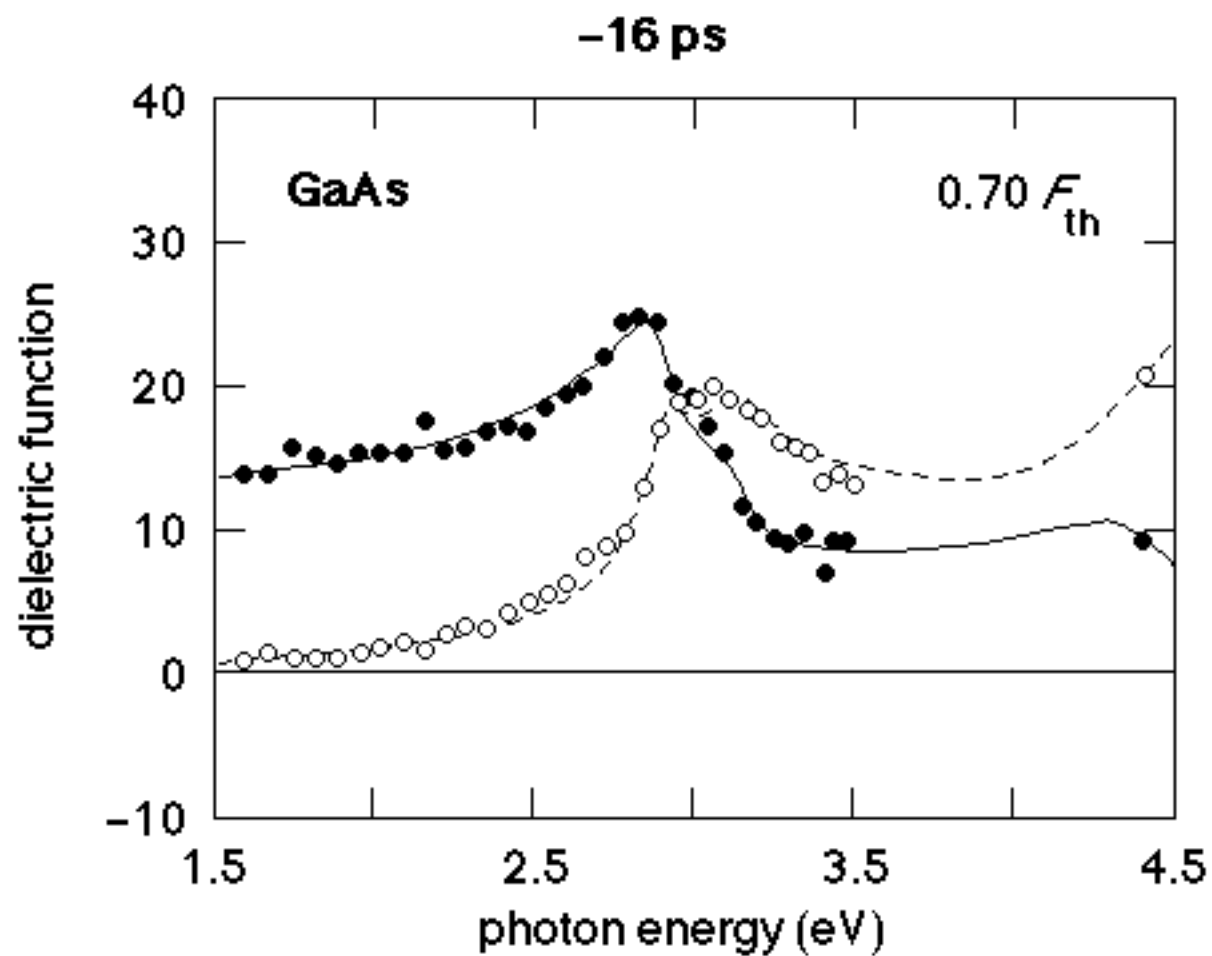
# BROADBAND DATA



# BROADBAND DATA

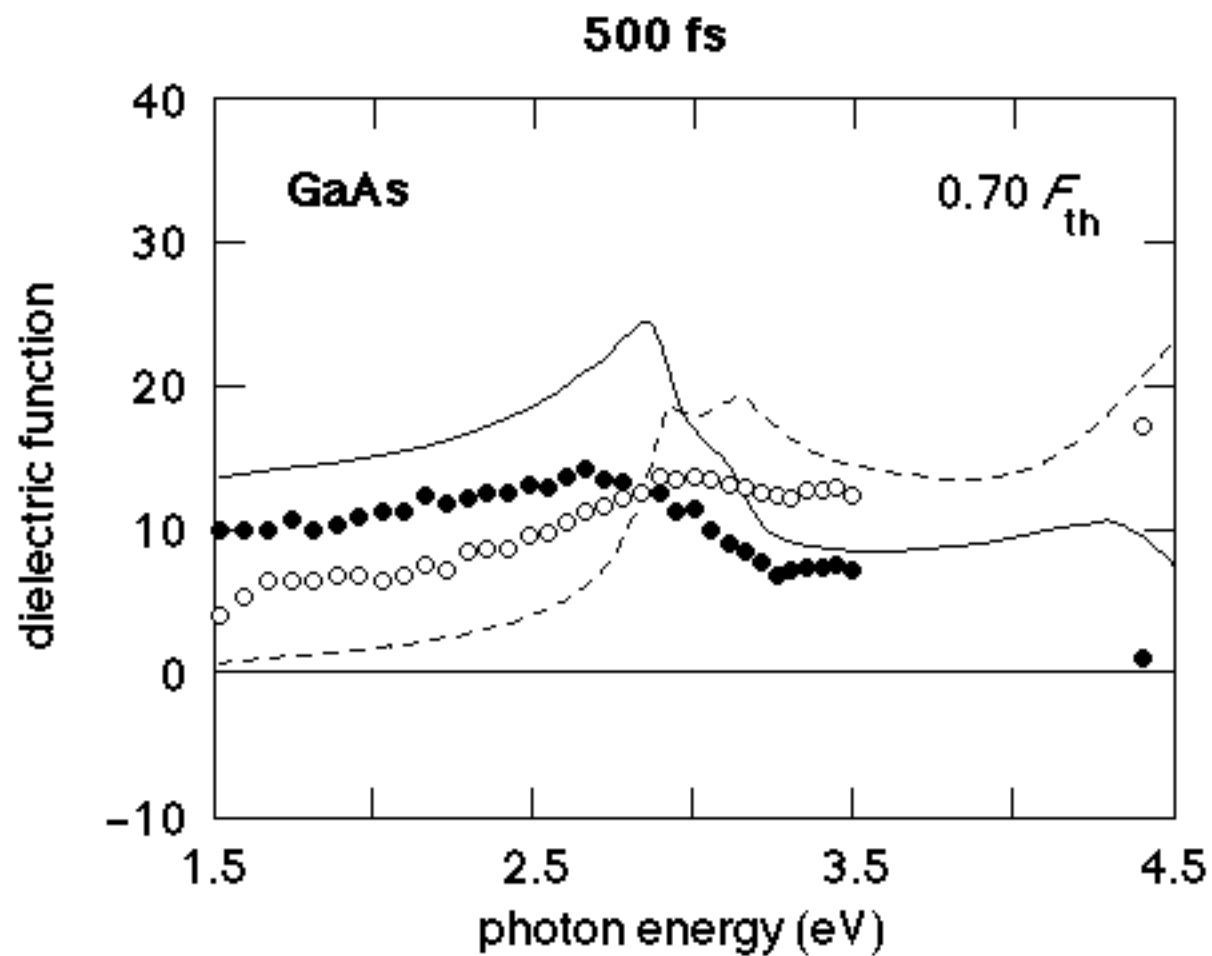


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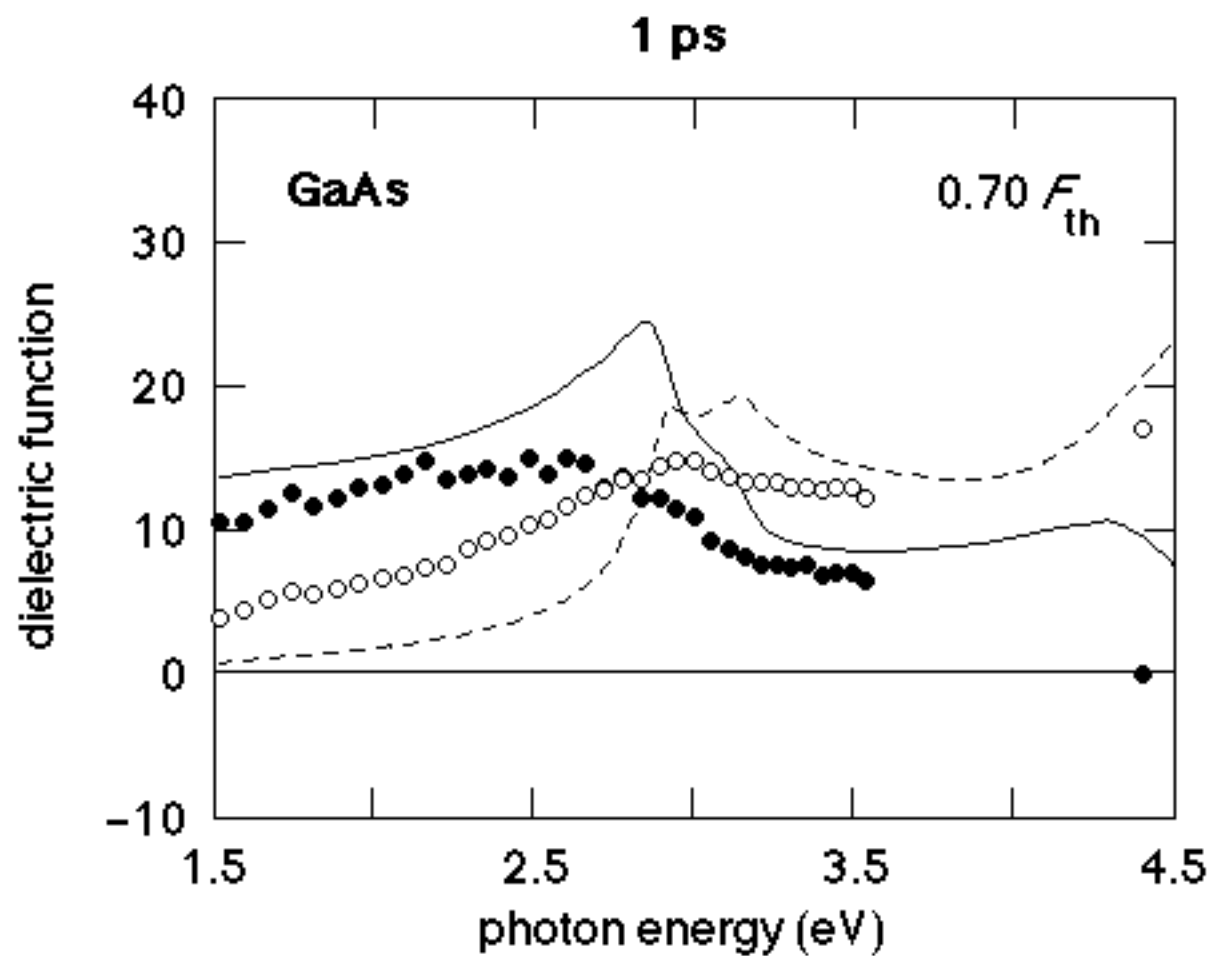




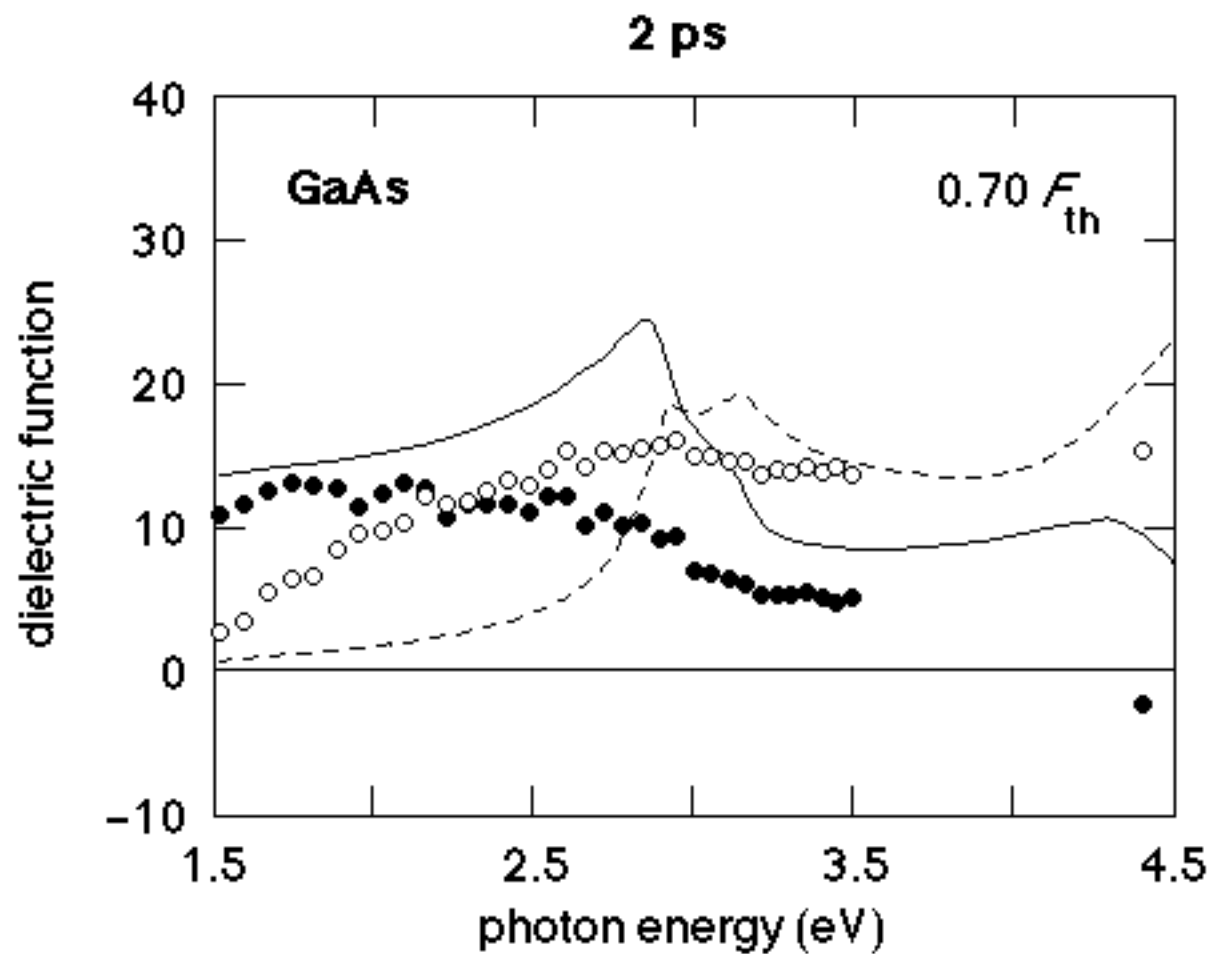
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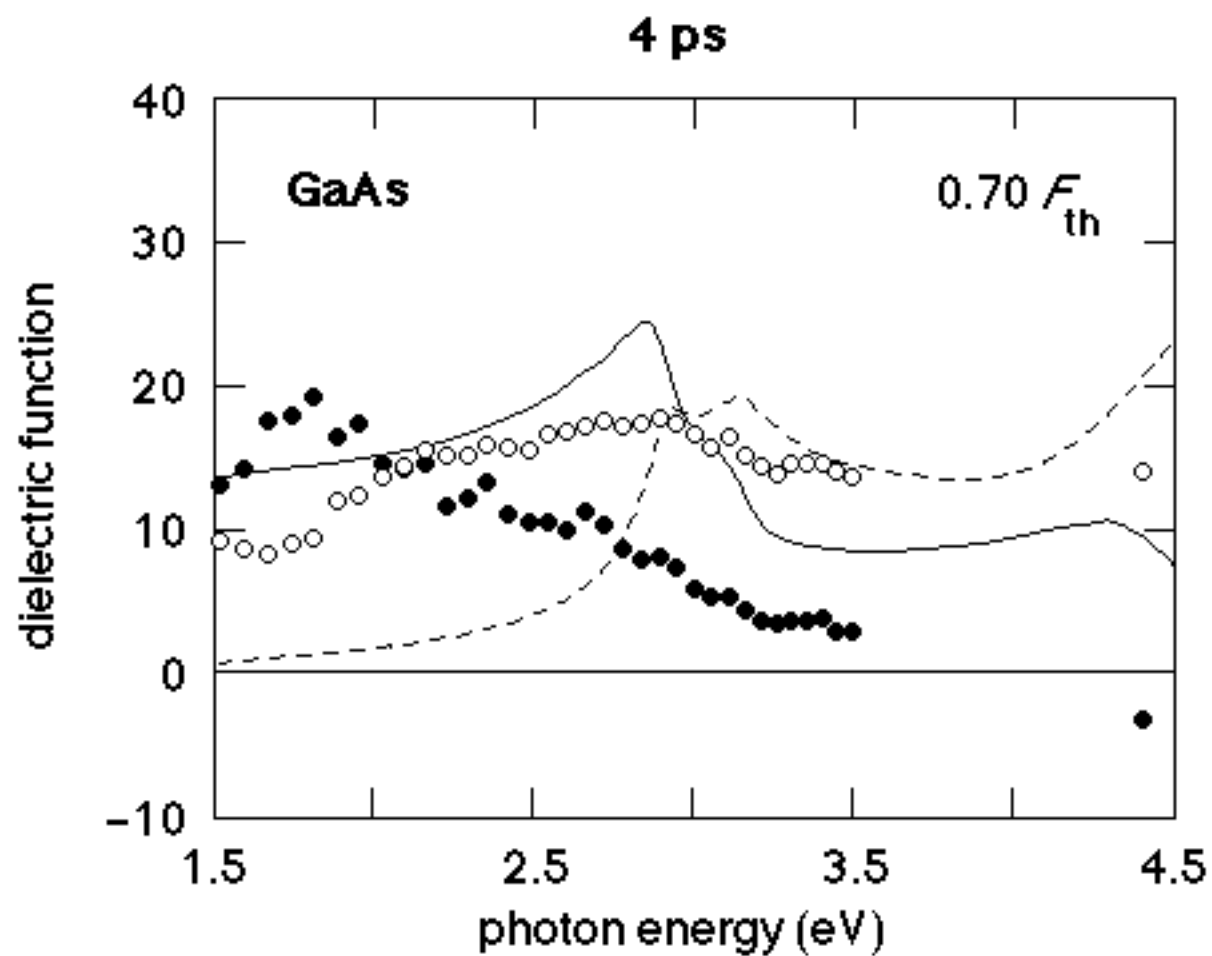
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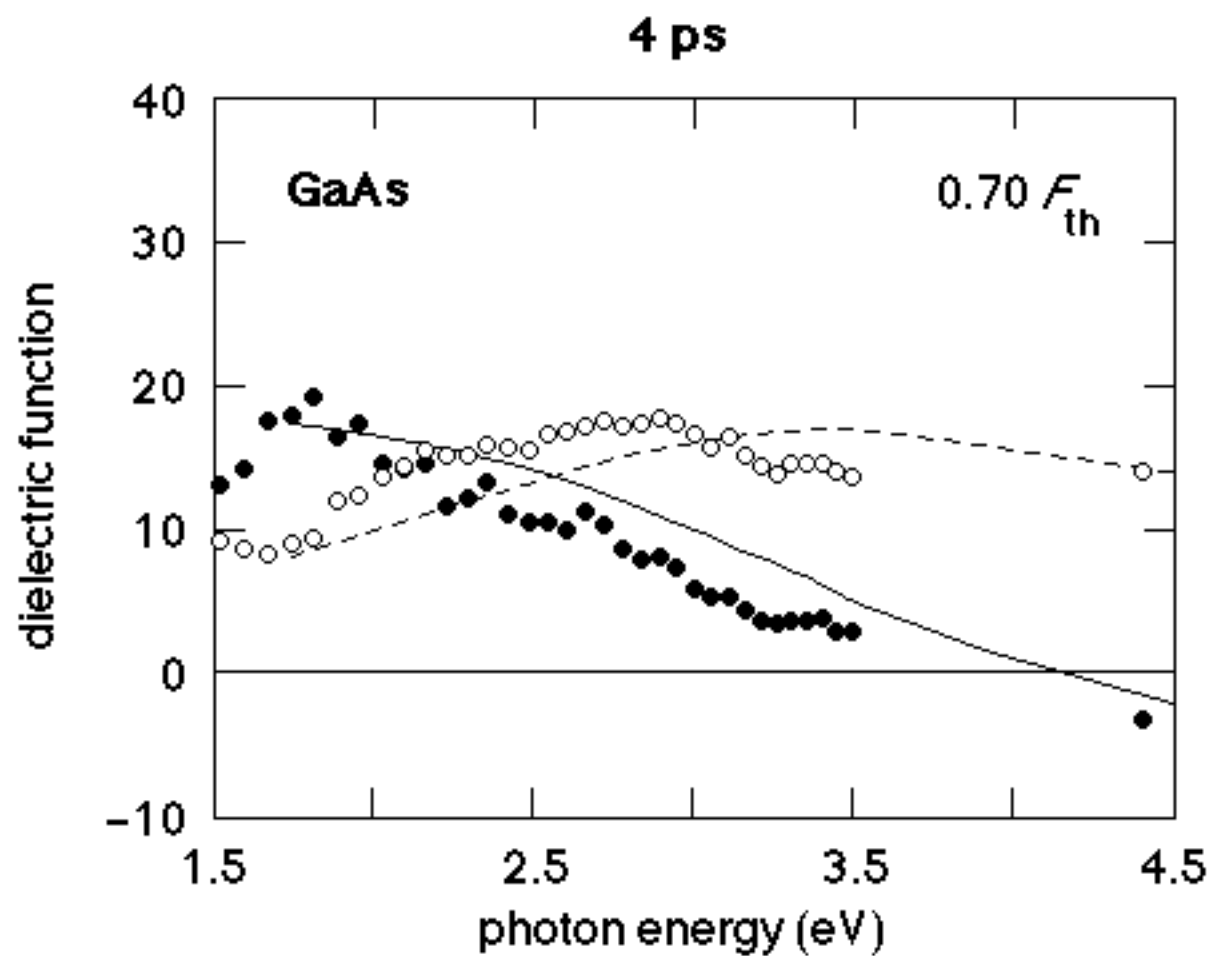
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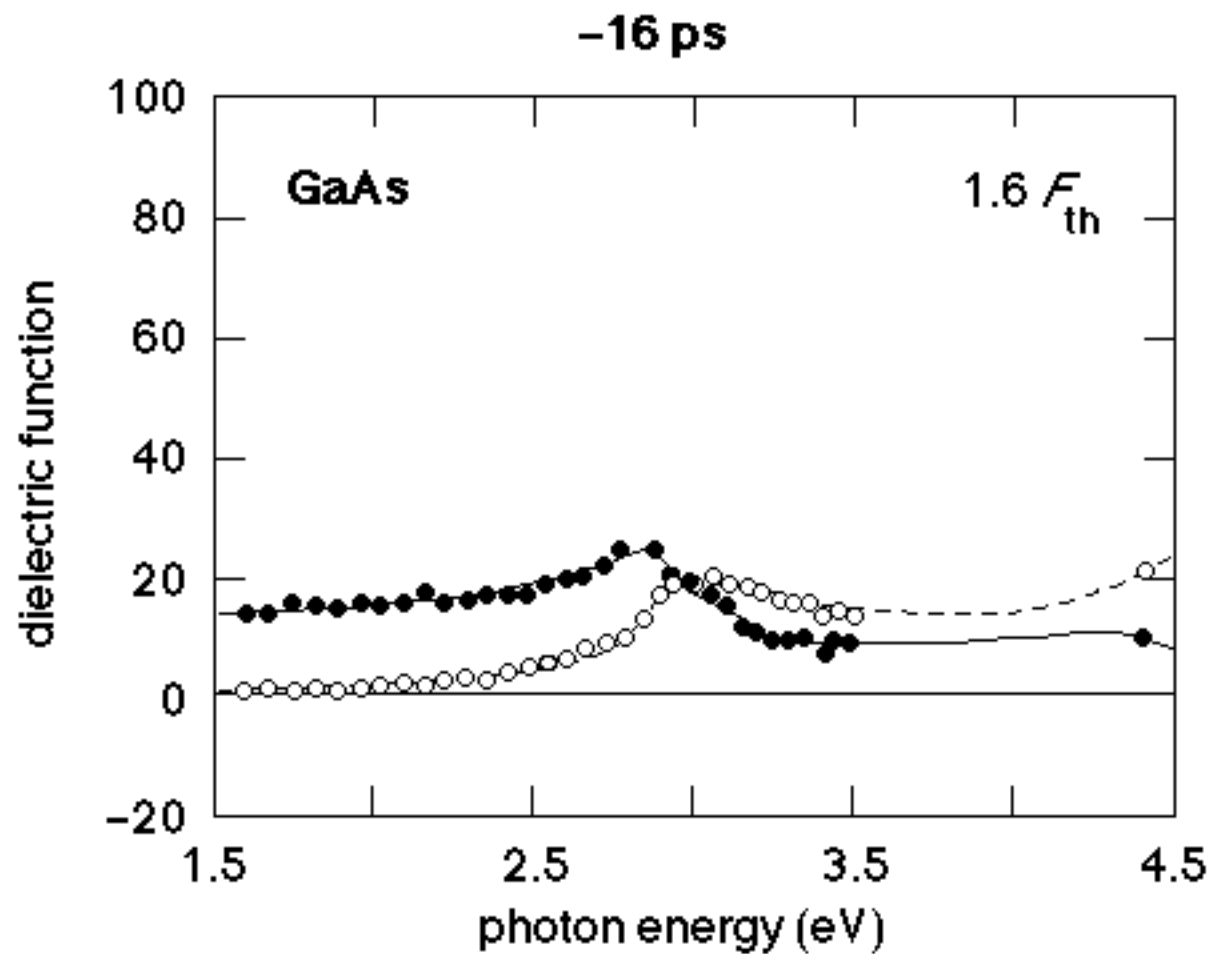
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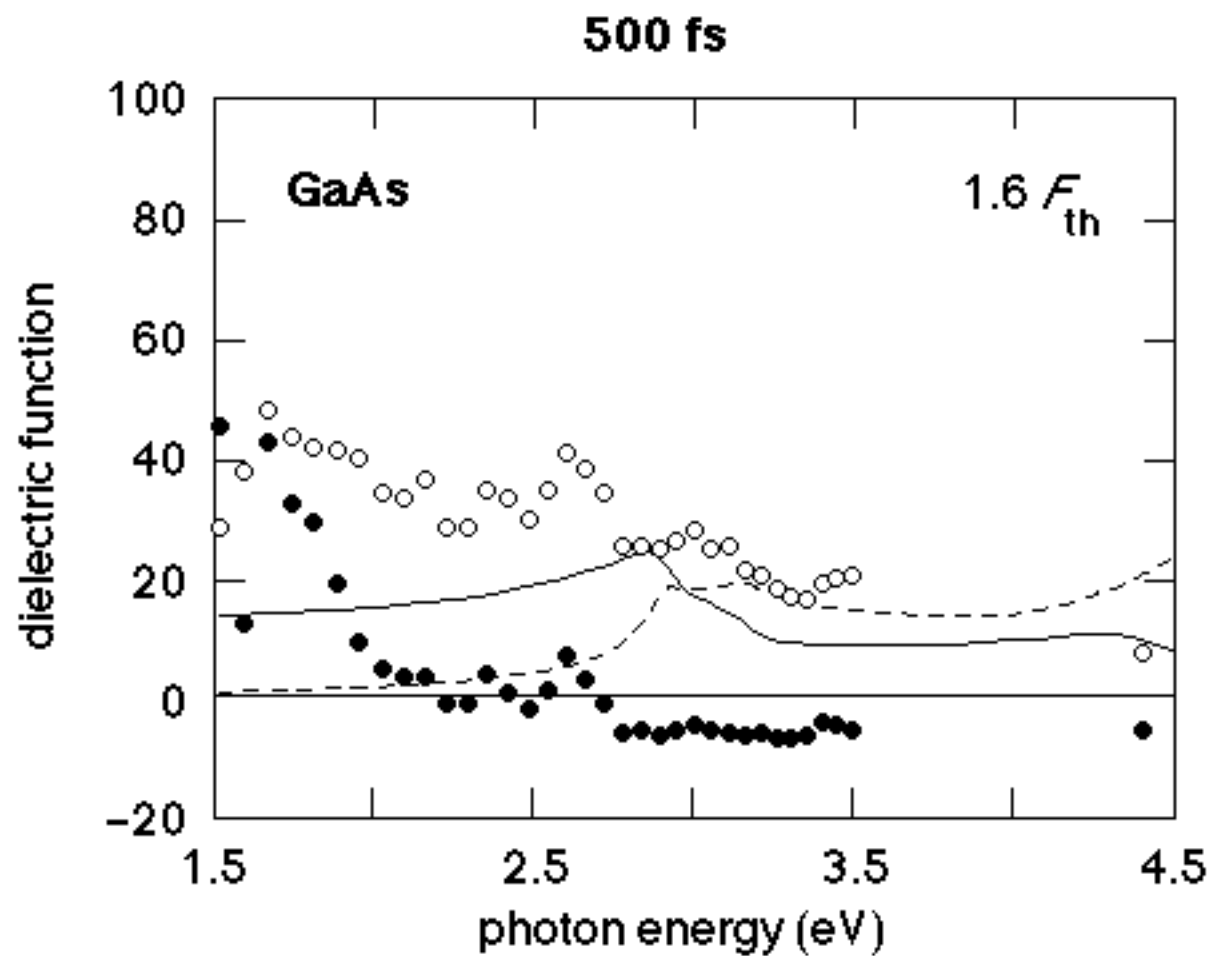
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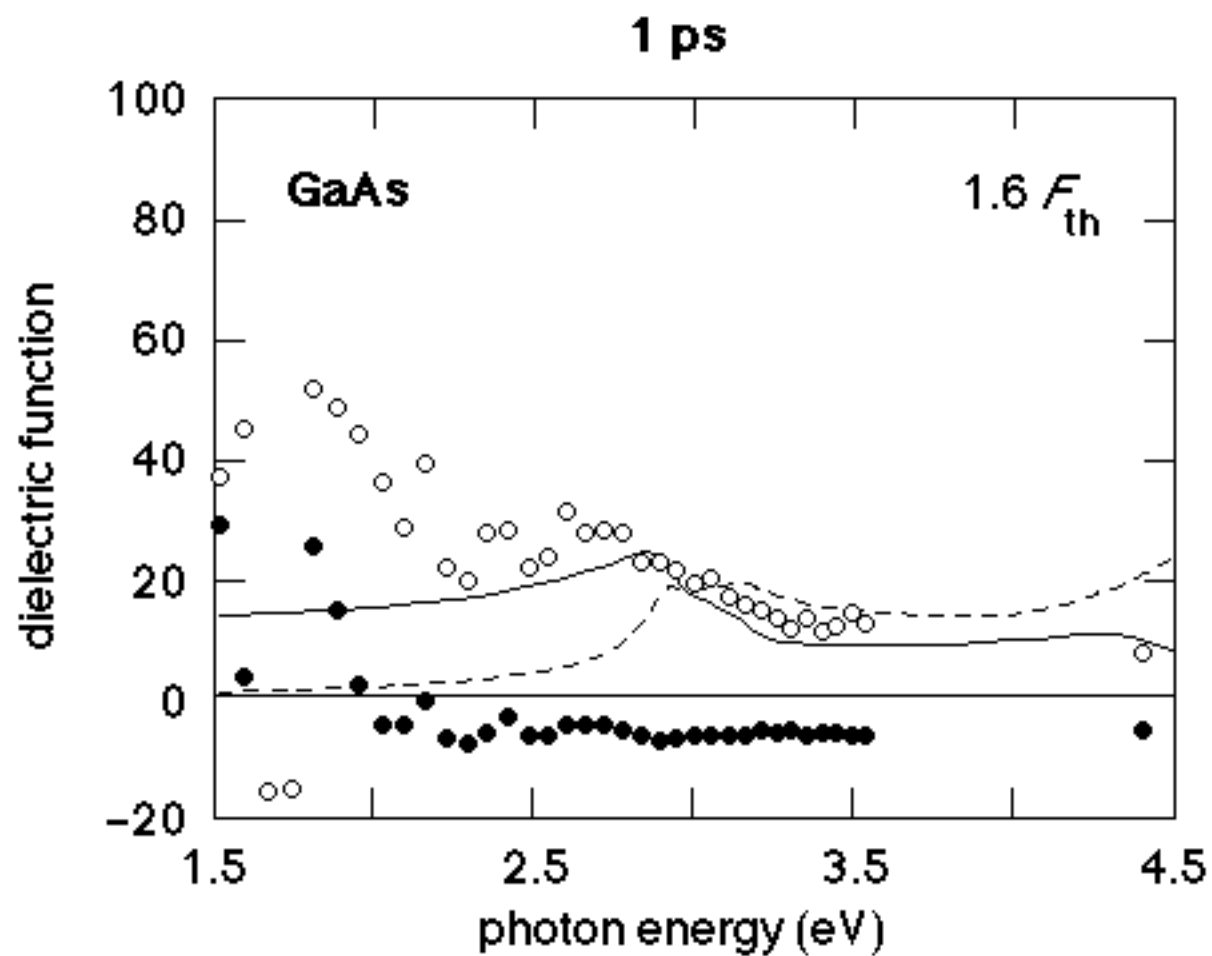
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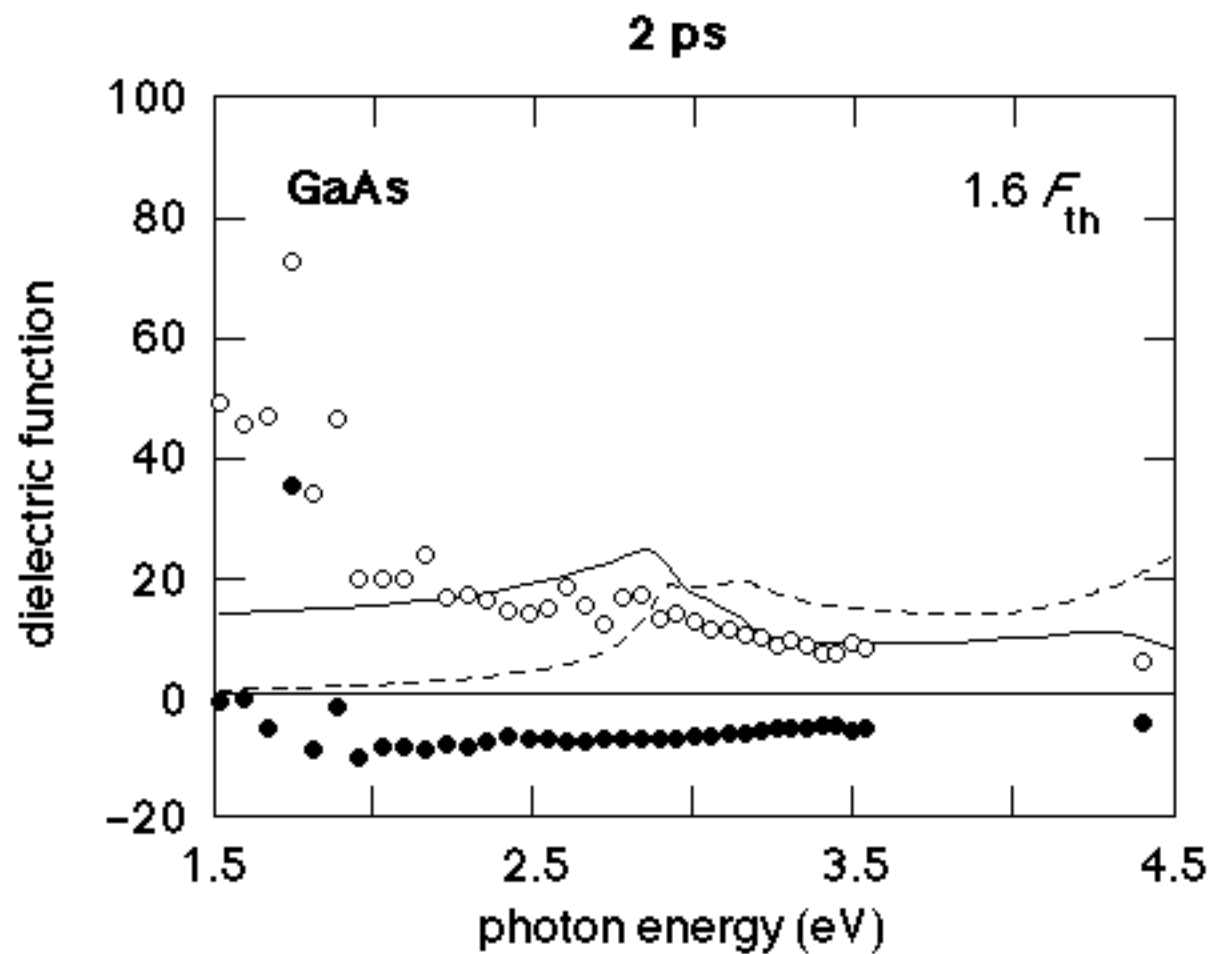


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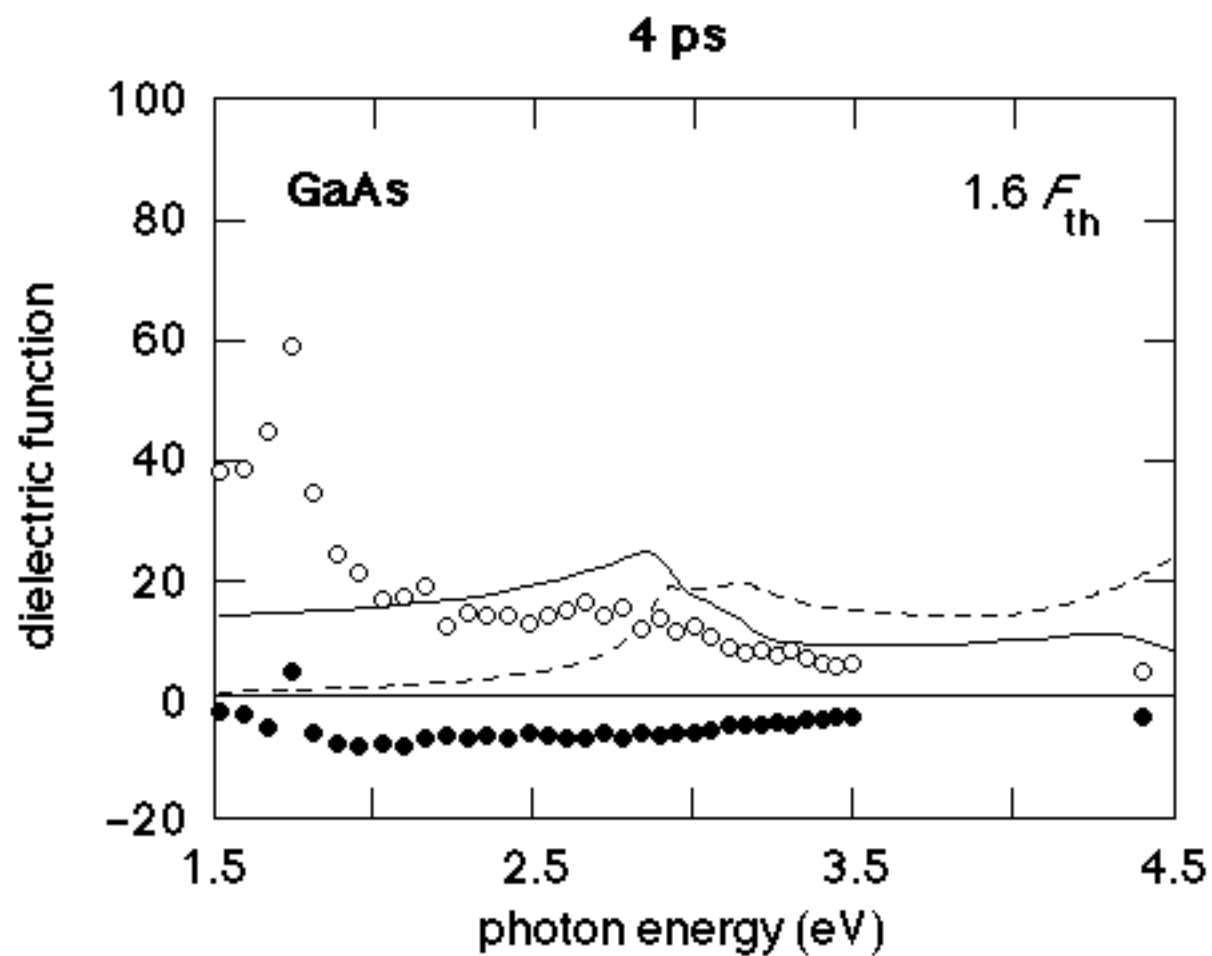




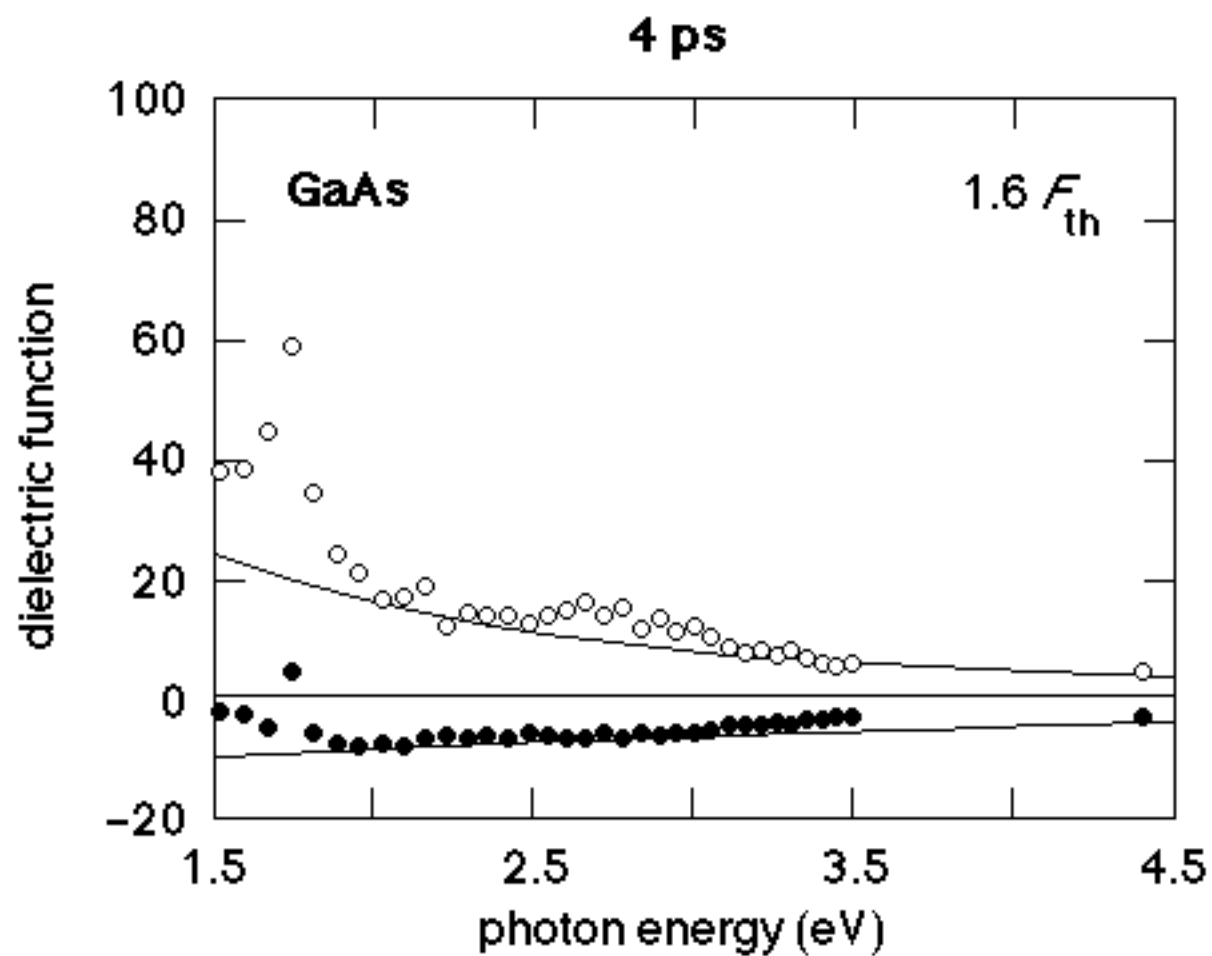
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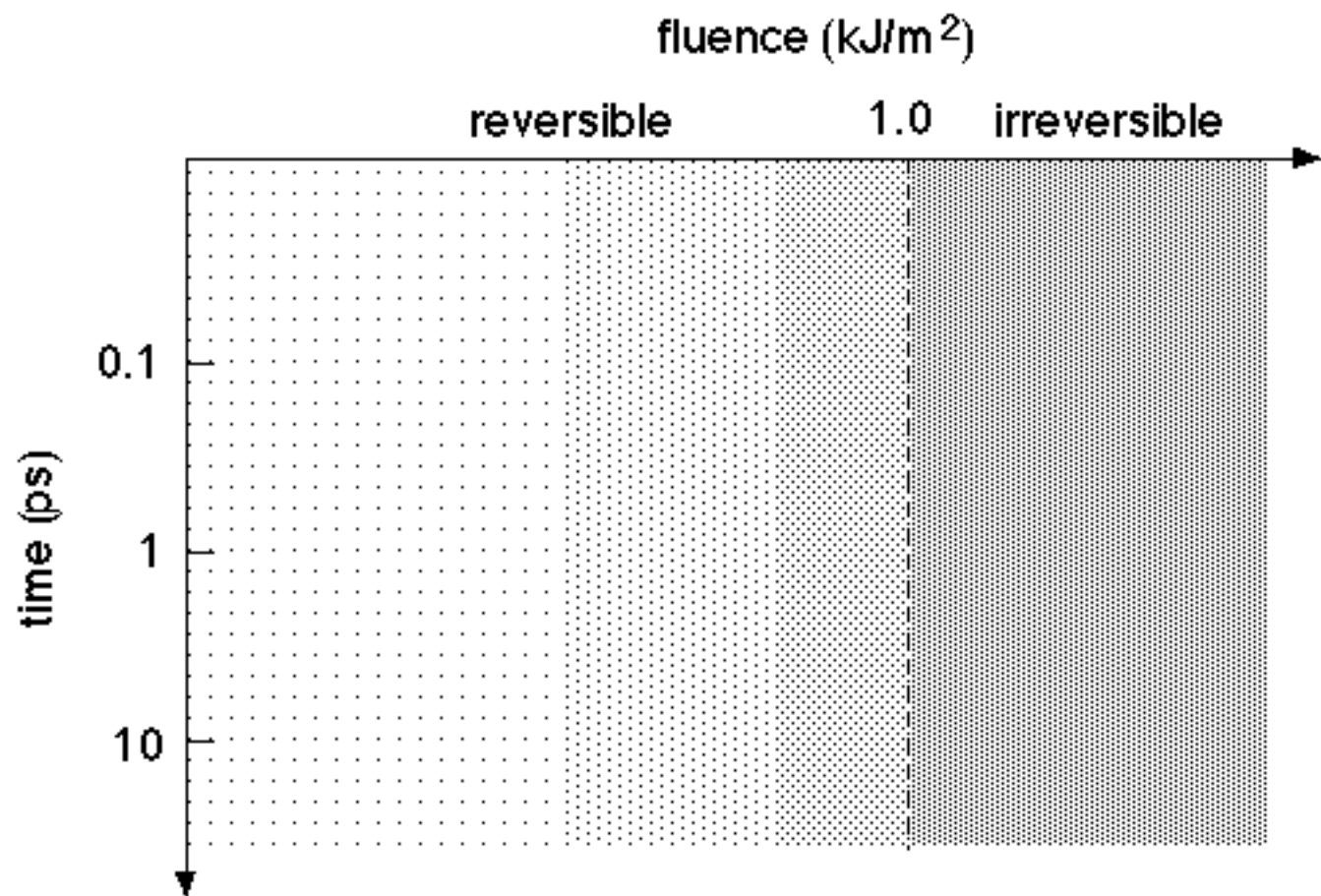
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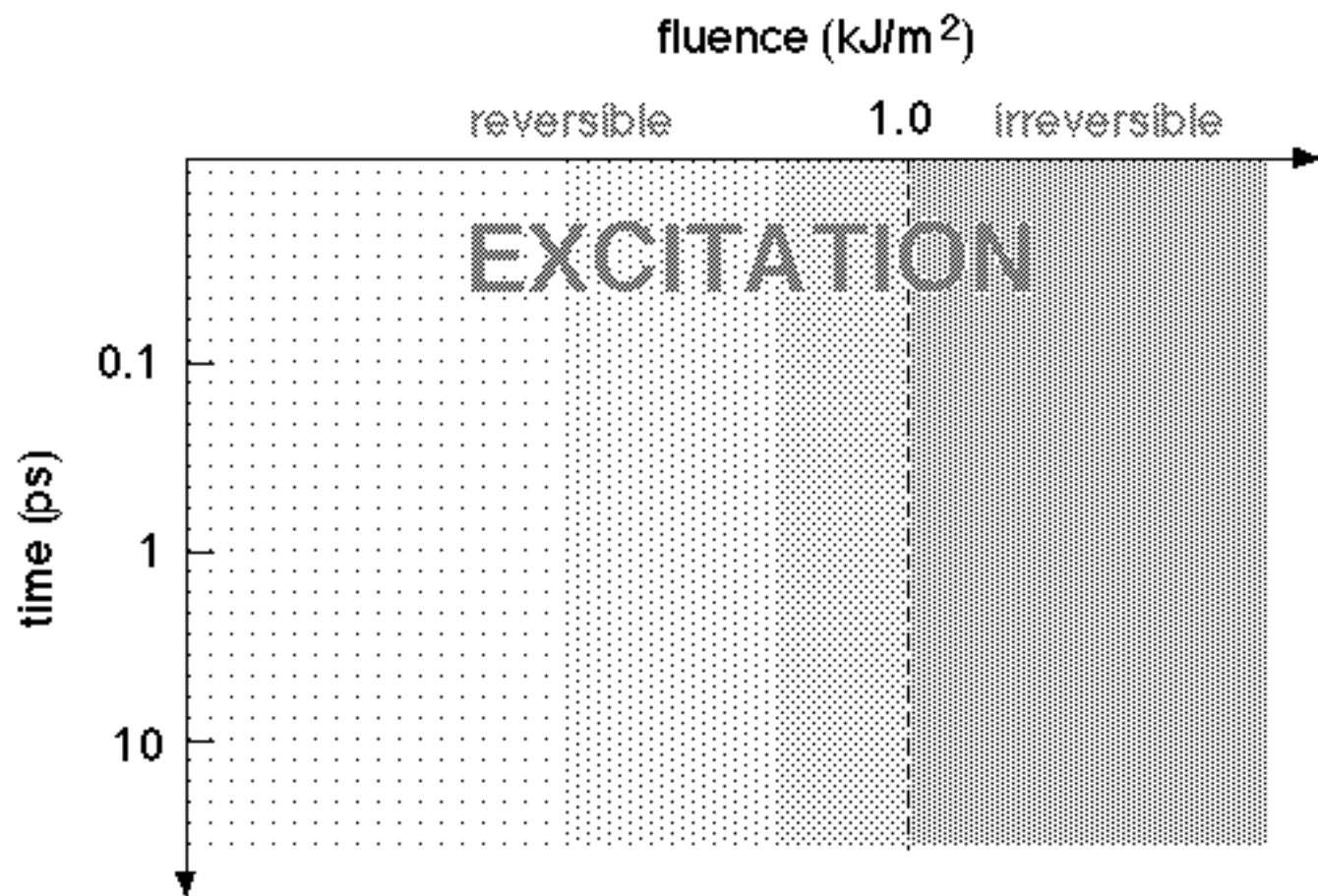
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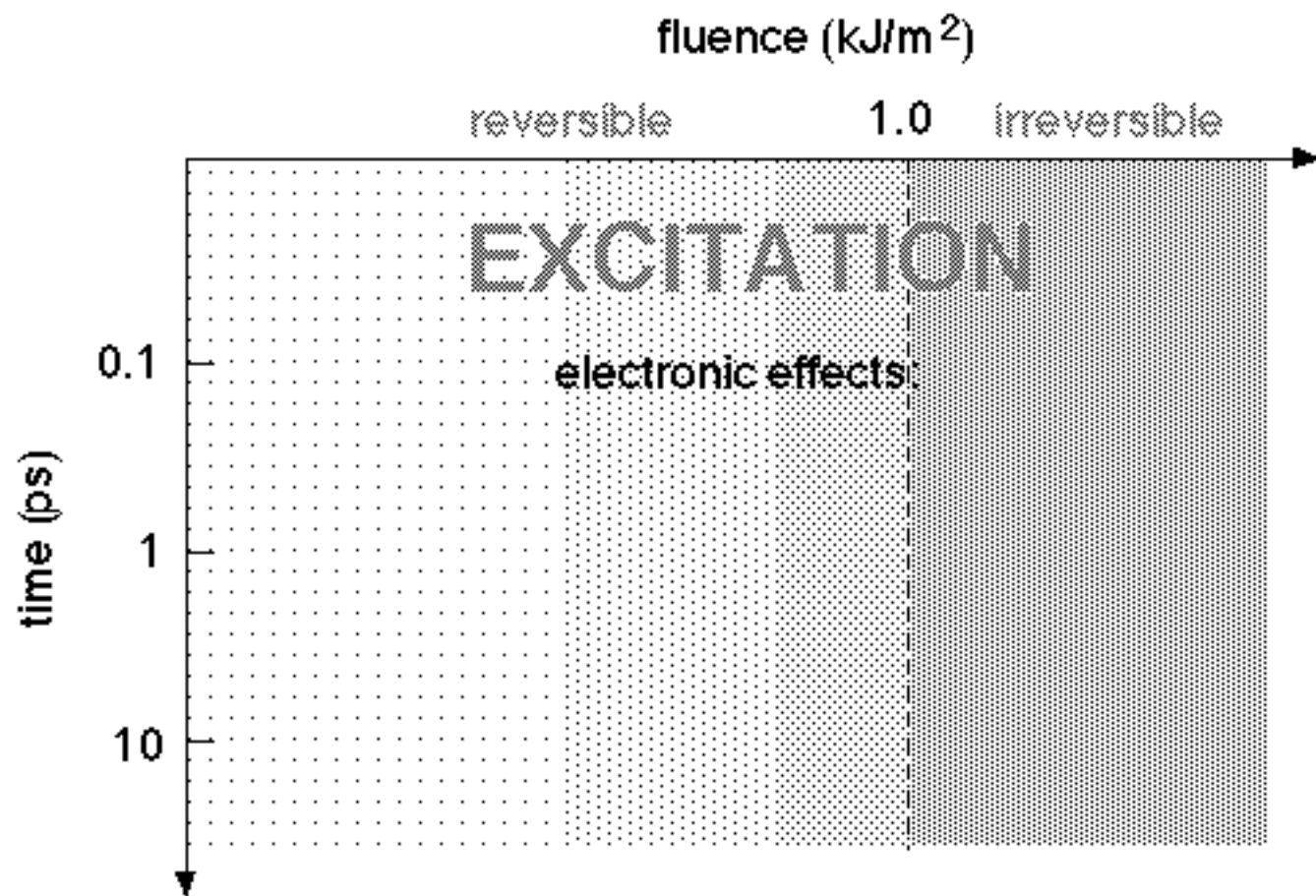
# ***BROADBAND DATA***



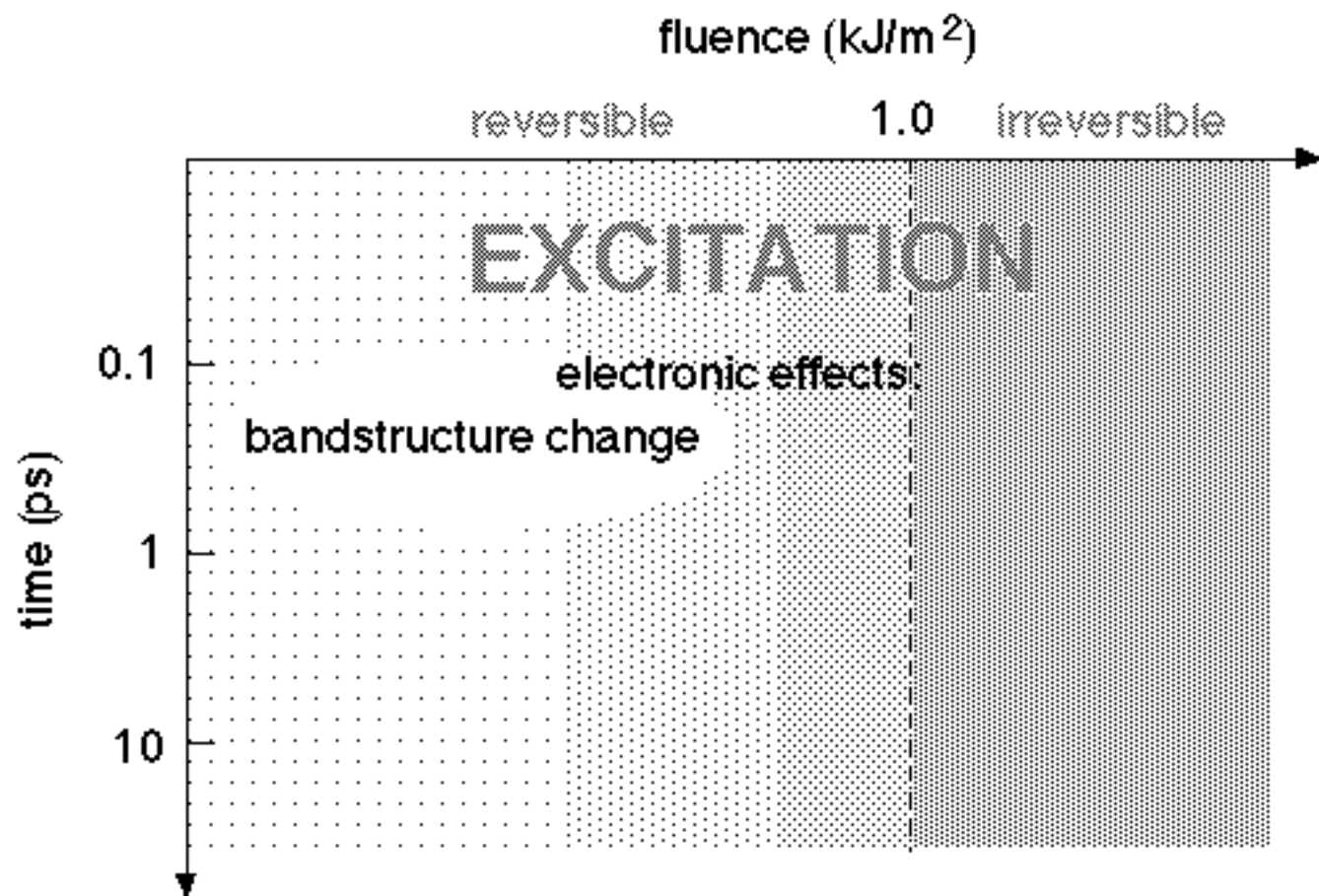
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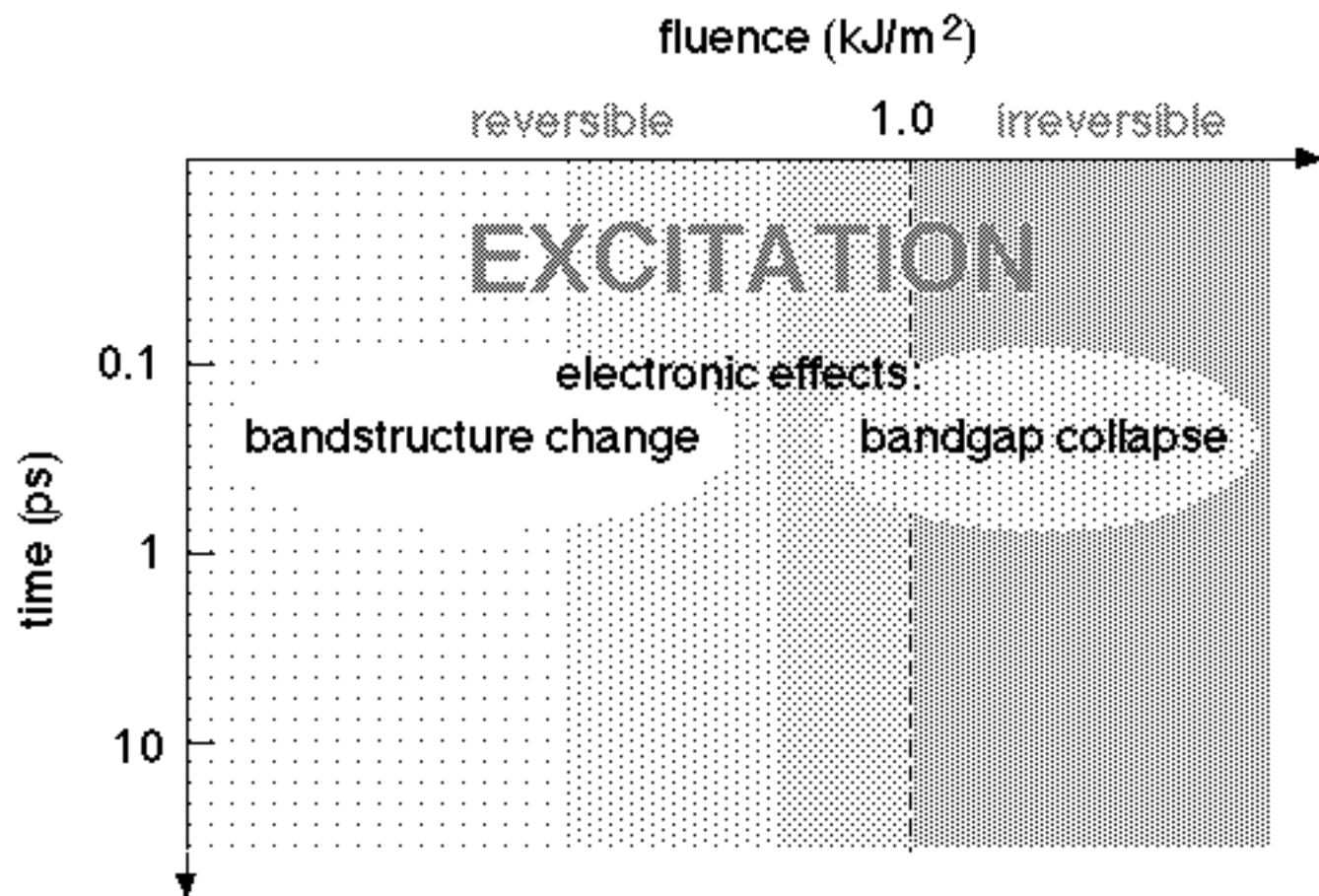
# ***BROADBAND DATA***



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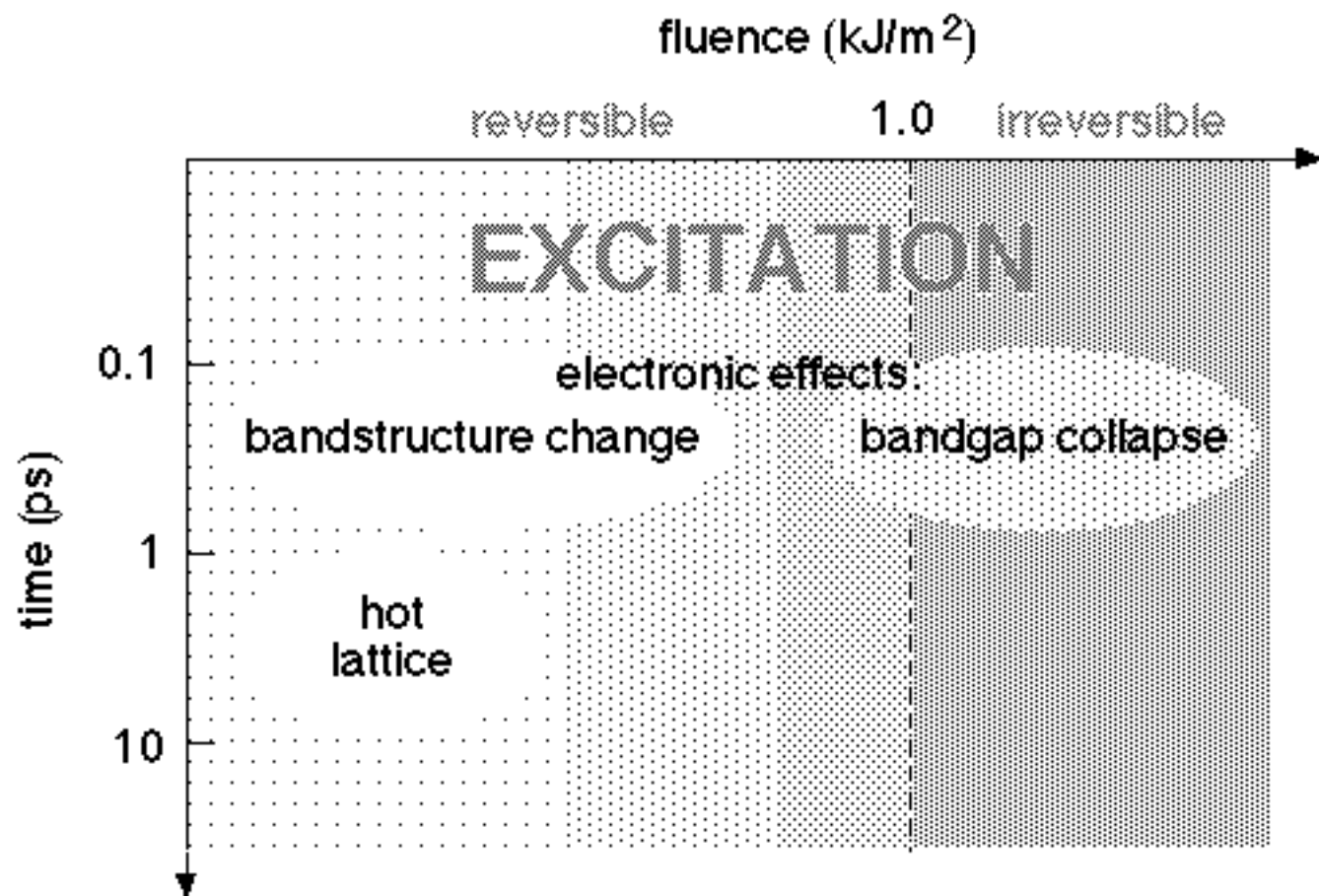


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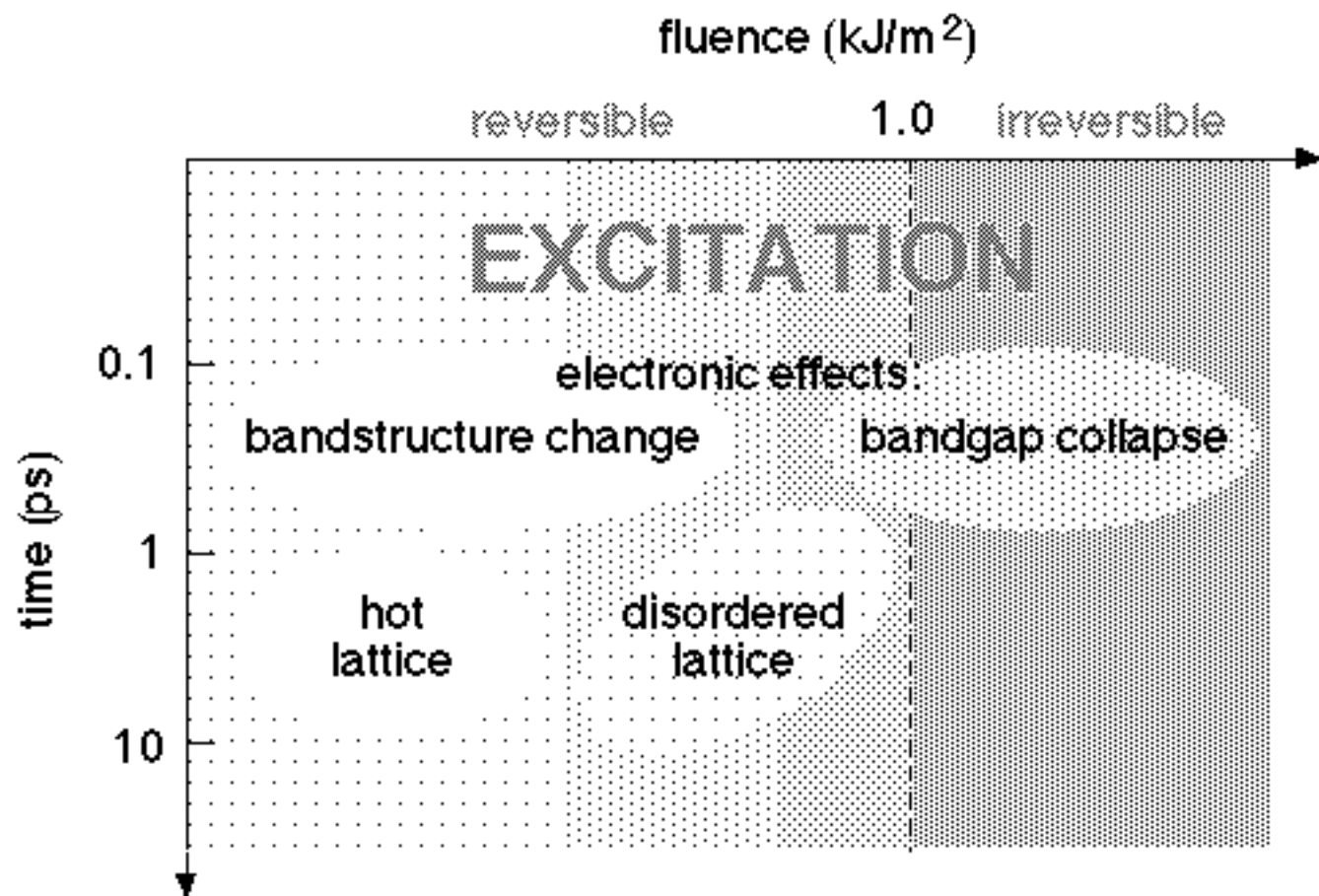




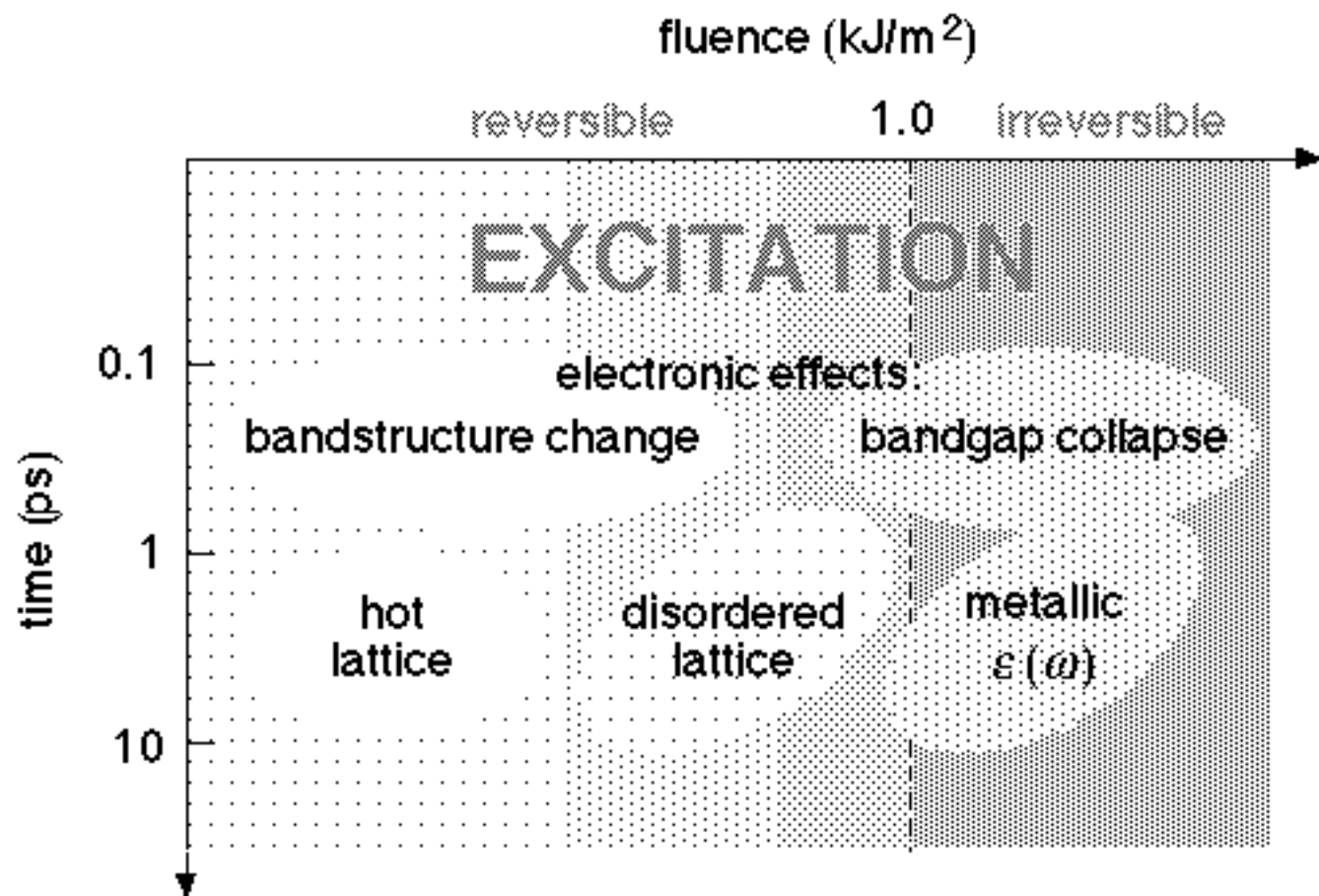
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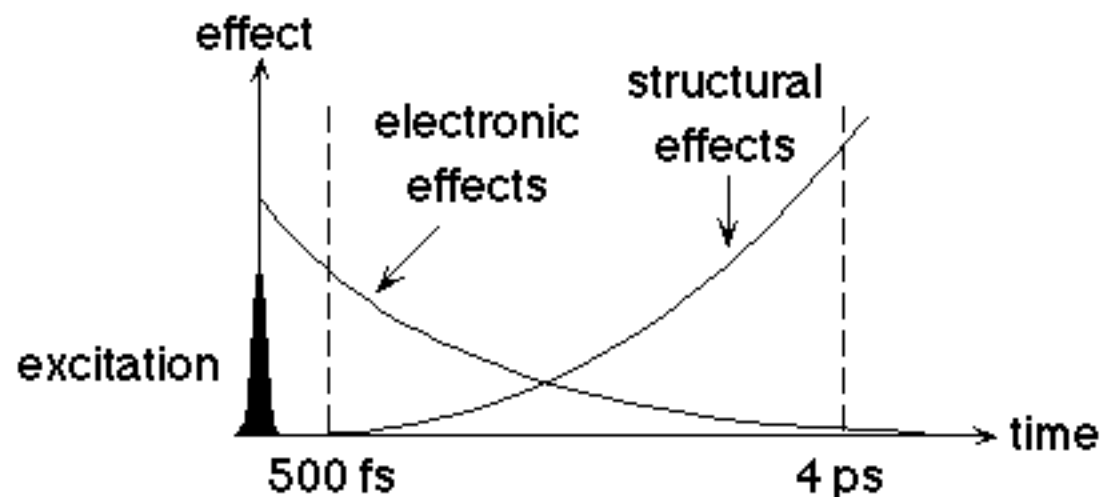
- ① Background
- ② Dielectric function
- ③ Nonlinear properties



## *CAUSE OF COLLAPSE*

What physical processes can change  $\epsilon(\omega)$ ?

- electronic effects: free carriers
- structural effects: atoms move



## *CAUSE OF COLLAPSE*

Electronic effects dominate at short times

- screening & many body effects
- direct free carrier effects (Drude model)

However,

changes in  $\epsilon(\omega)$  at short times do not fit a Drude model!



## *CAUSE OF COLLAPSE*

Is rapid structural change possible?

- excite 10% of valence electrons → lattice instability
- 10% change in bond length → semiconductor to metal transition

Time-scale for structural change reasonable:  
moving 10% of bond length in 1 ps requires 25 m/s



## ***NONLINEAR OPTICS***

Field strength at second harmonic:

$$E_r(2\omega) = -4\pi P^{(2)}(2\omega) g[\epsilon(\omega), \epsilon(2\omega), \theta_i]$$





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Field strength at second harmonic:

$$E_r(2\omega) = -4\pi P^{(2)}(2\omega) g[\epsilon(\omega), \epsilon(2\omega), \theta_i]$$

where:

$$P^{(2)}(2\omega) = 2\chi^{(2)} E_i^2(\omega) f[\epsilon(\omega), \theta_i]$$

so:



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Field strength at second harmonic:

$$E_r(2\omega) = -4\pi P^{(2)}(2\omega) g[\epsilon(\omega), \epsilon(2\omega), \theta_i]$$

where:

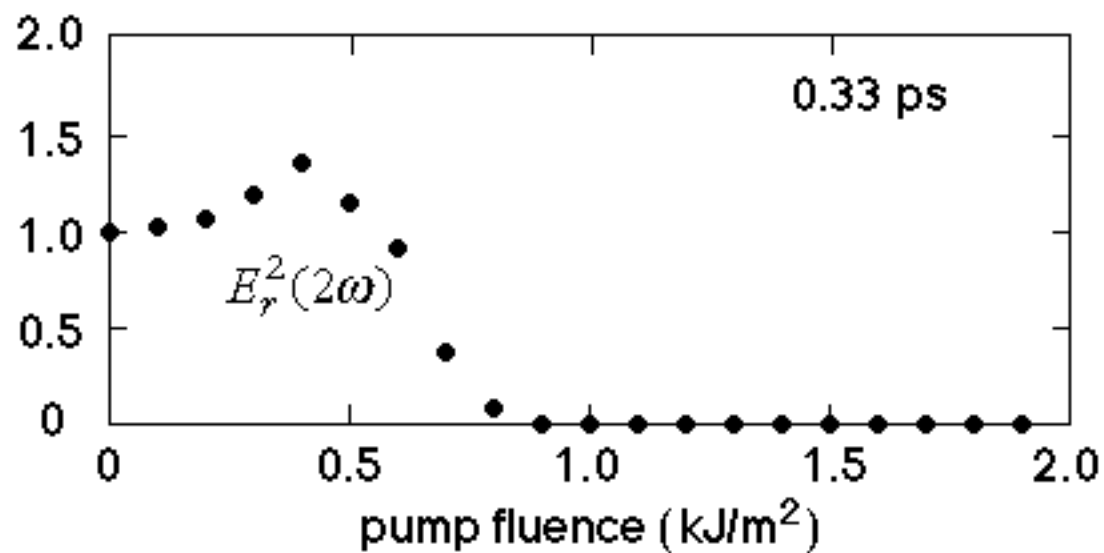
$$P^{(2)}(2\omega) = 2\chi^{(2)} E_i^2(\omega) f[\epsilon(\omega), \theta_i]$$

so:

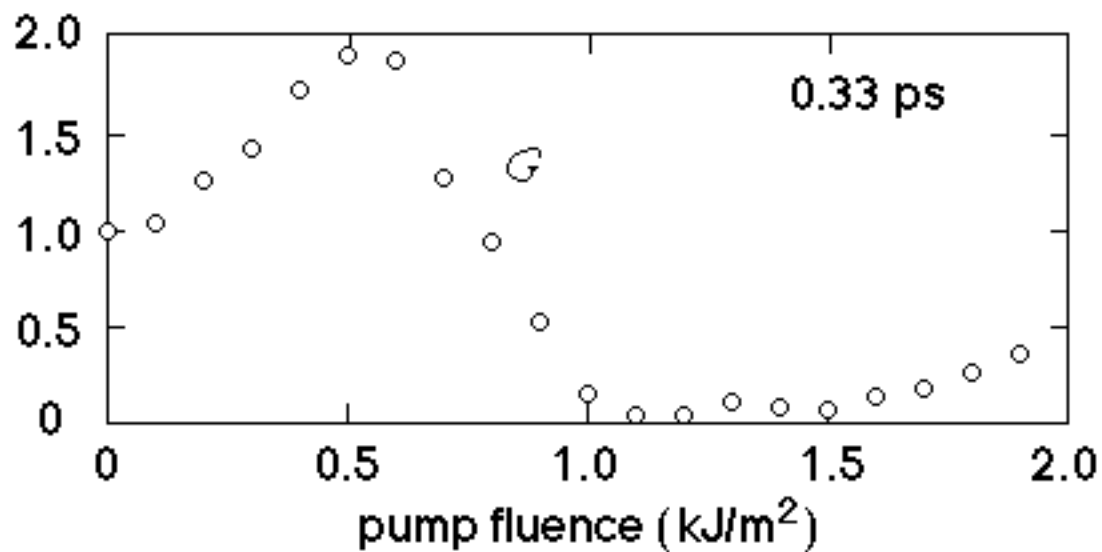
$$|\chi^{(2)}|^2 = \frac{E_r^2(2\omega)}{E_i^2(\omega) G[\epsilon(\omega), \epsilon(2\omega), \theta_i]}$$



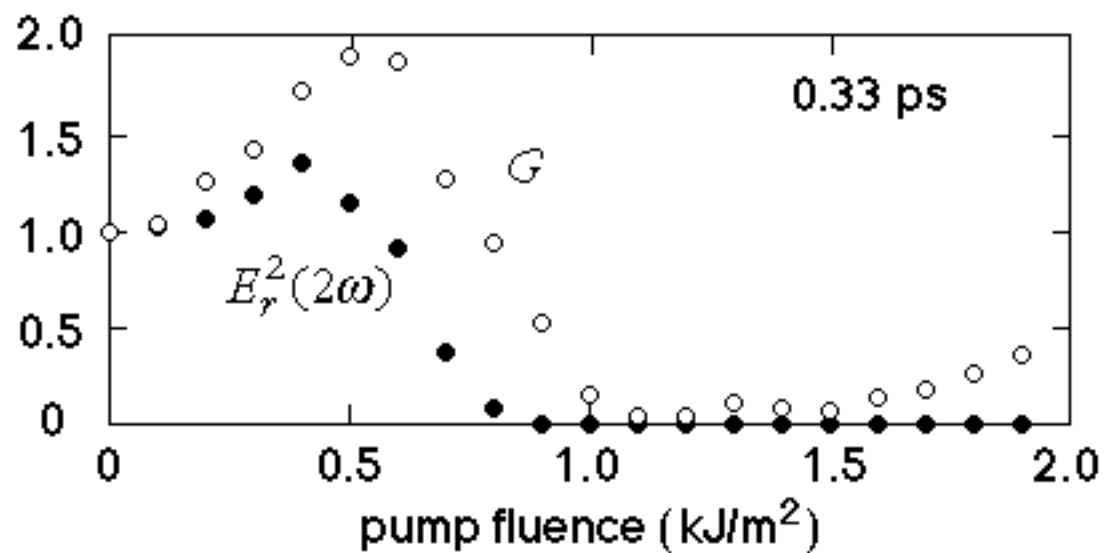
## SECOND HARMONIC DATA



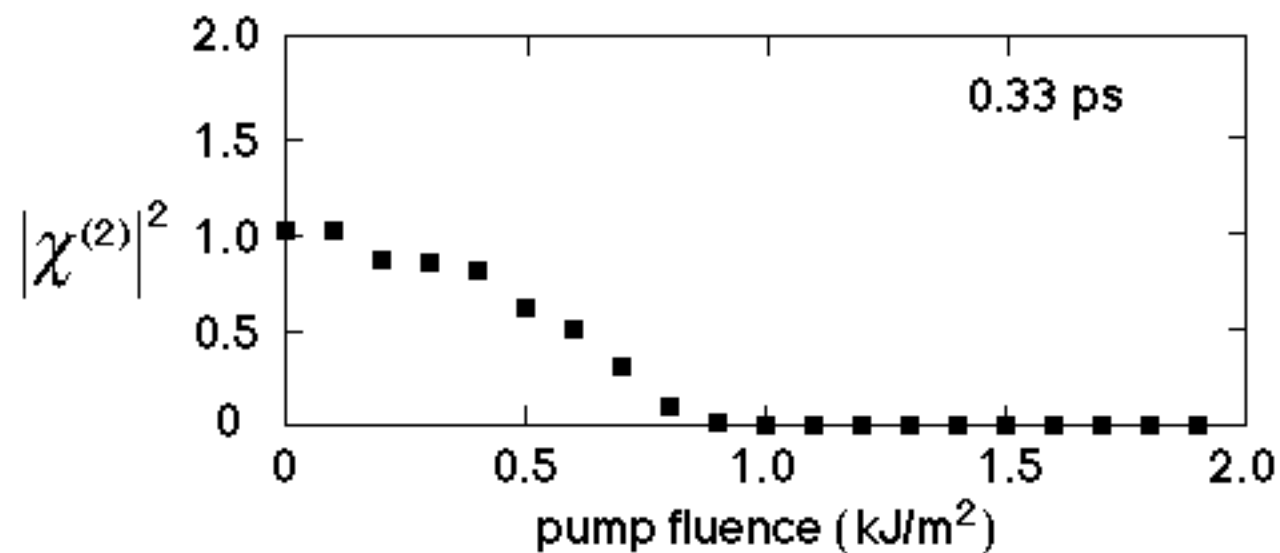
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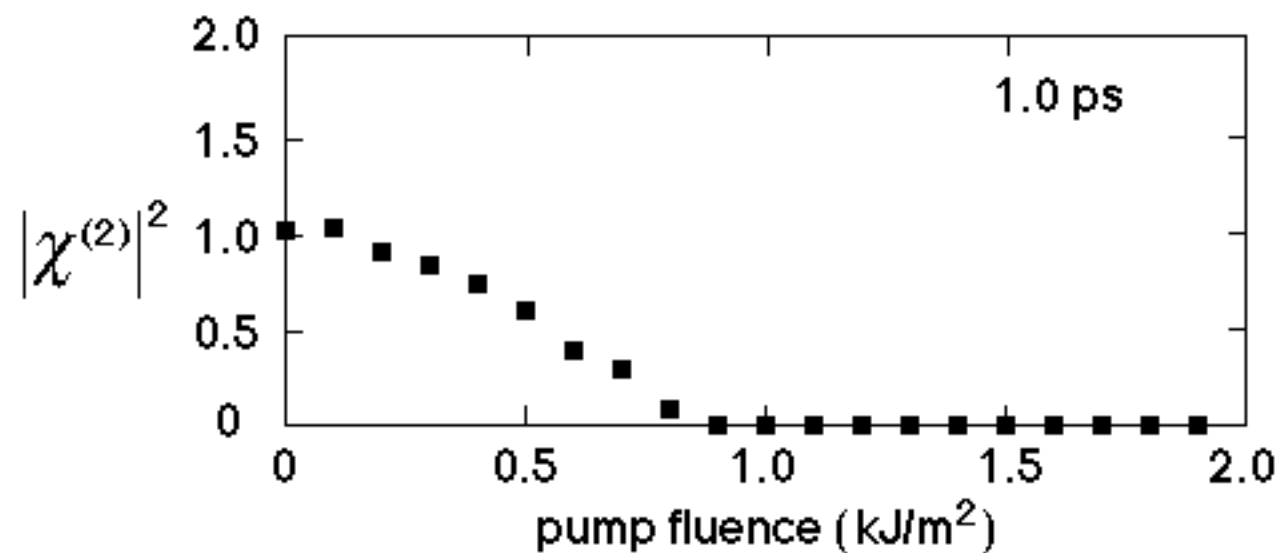
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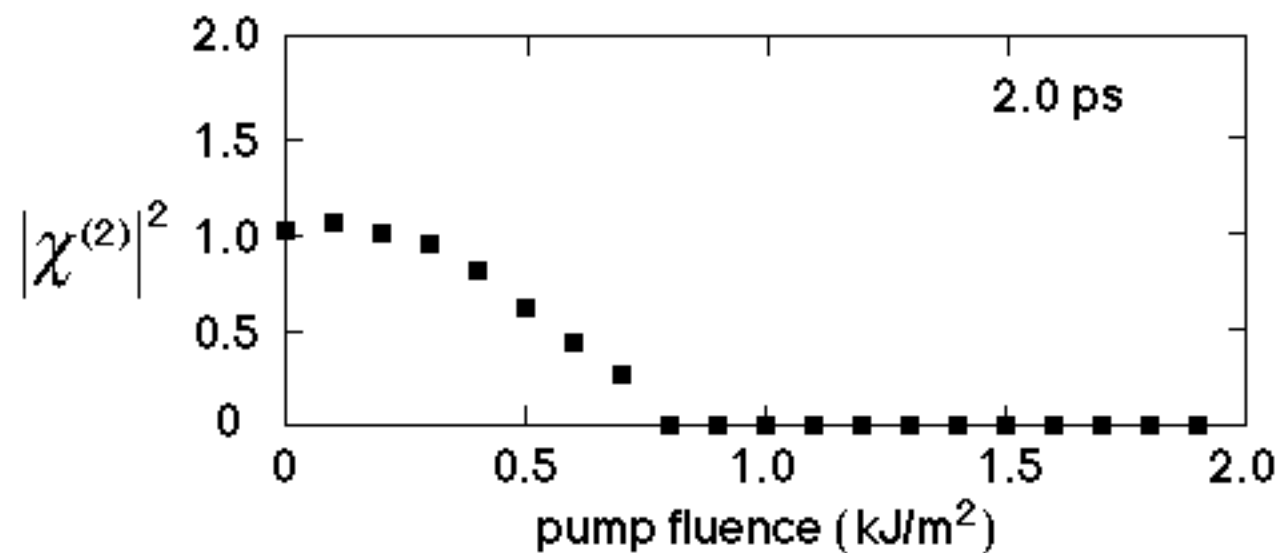
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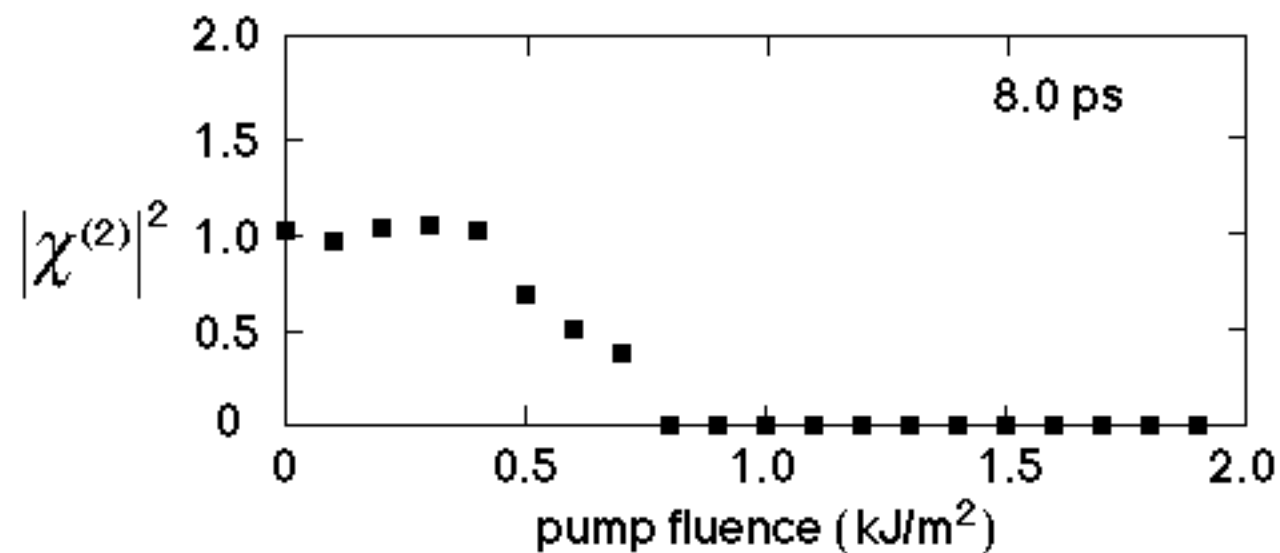


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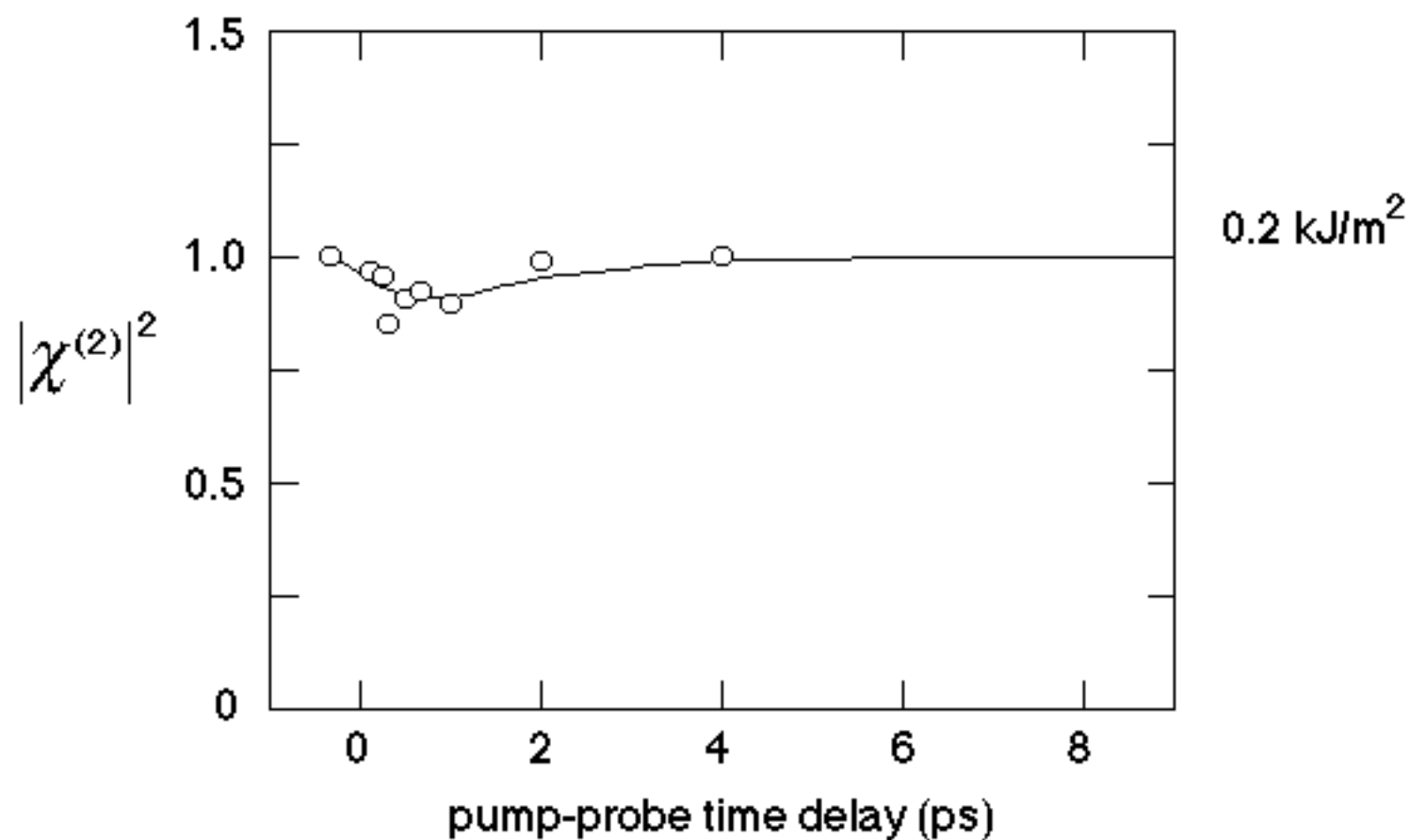




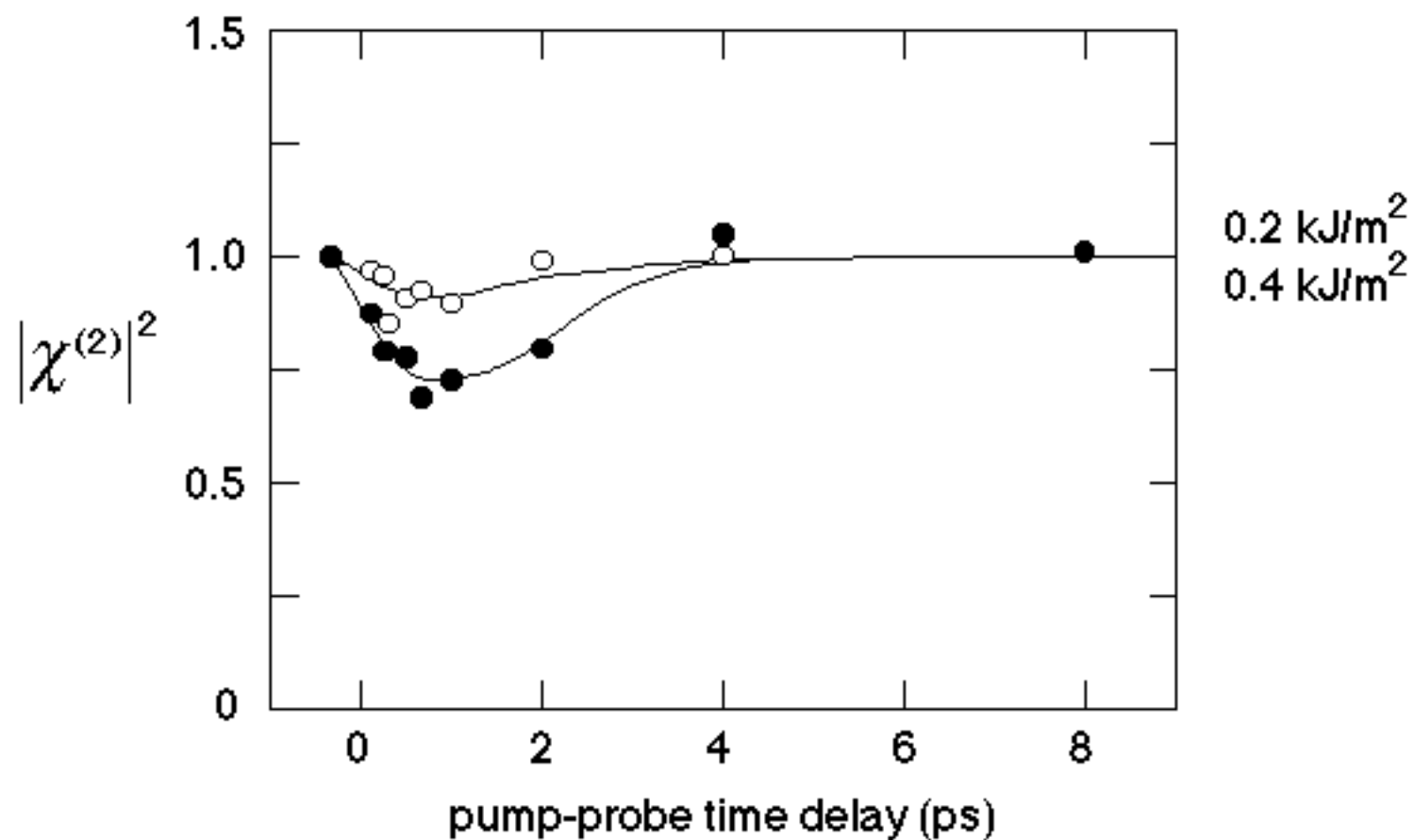
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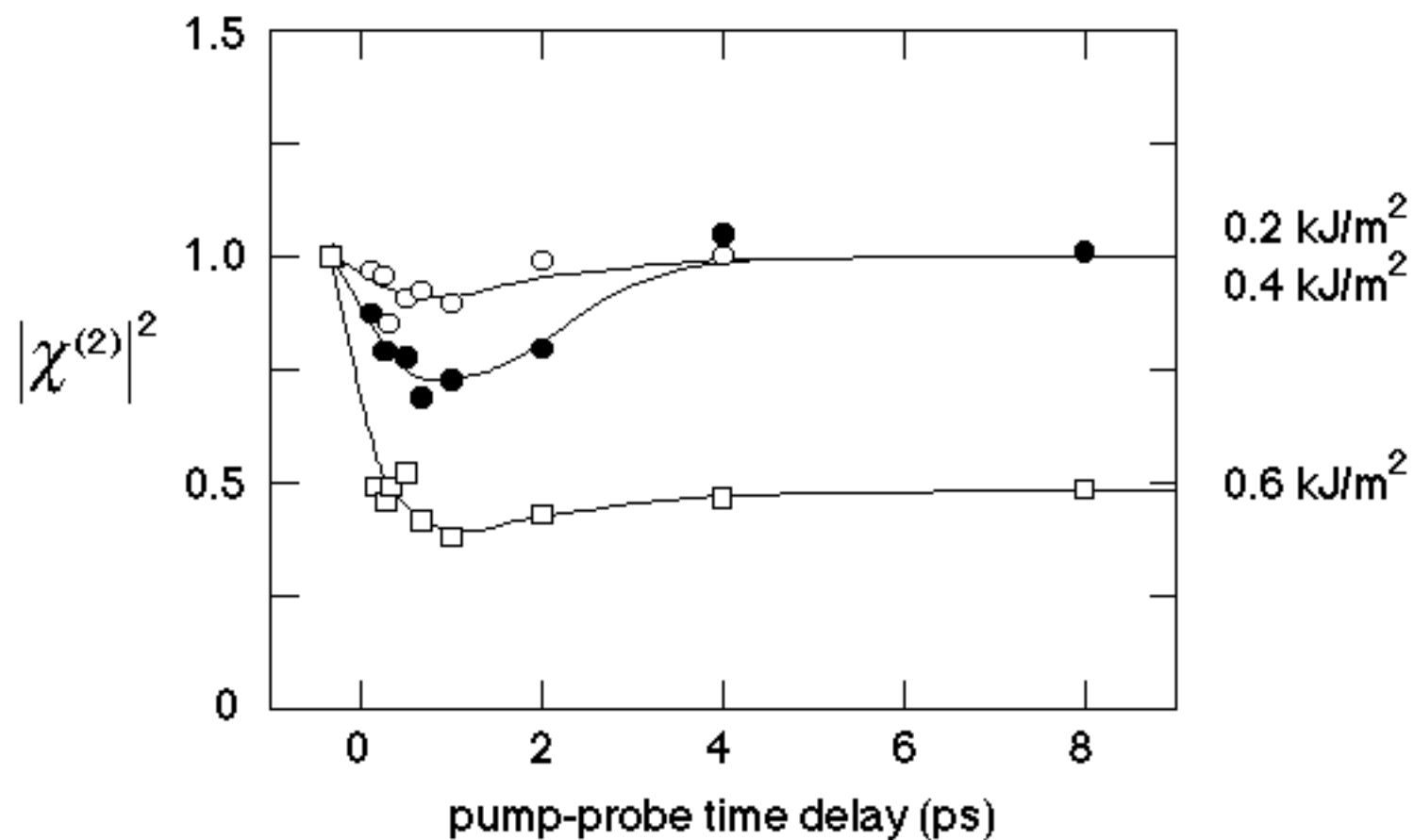
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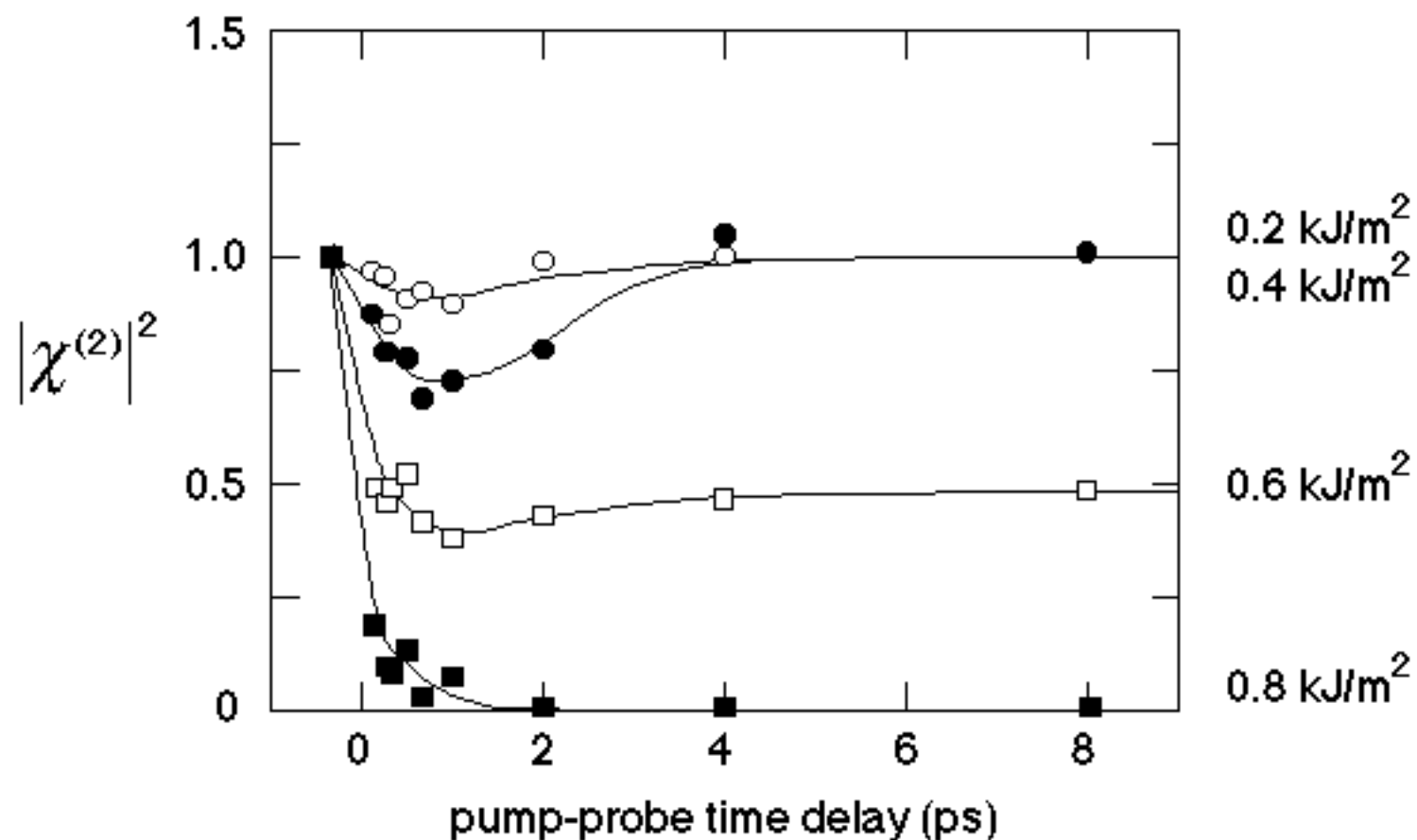
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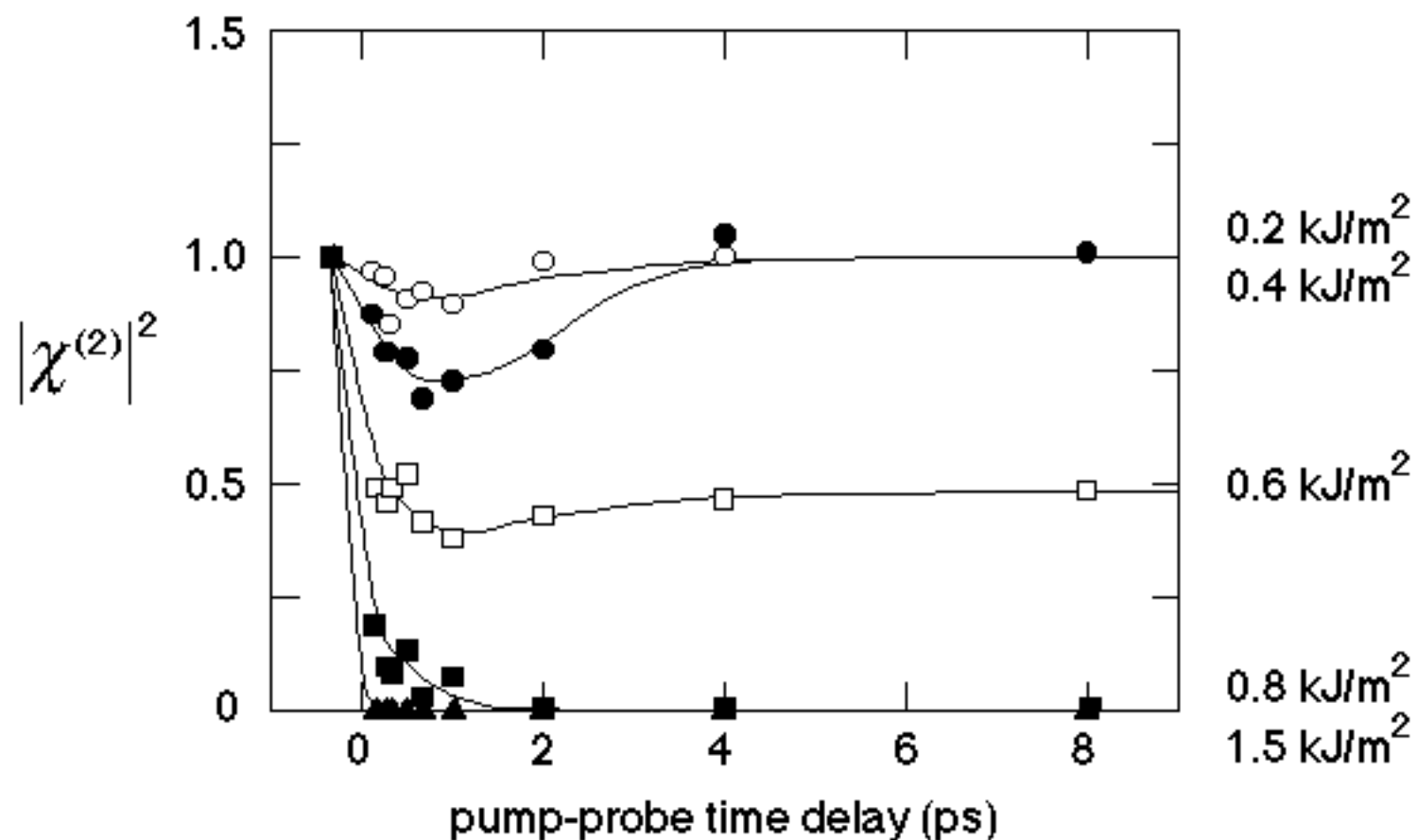
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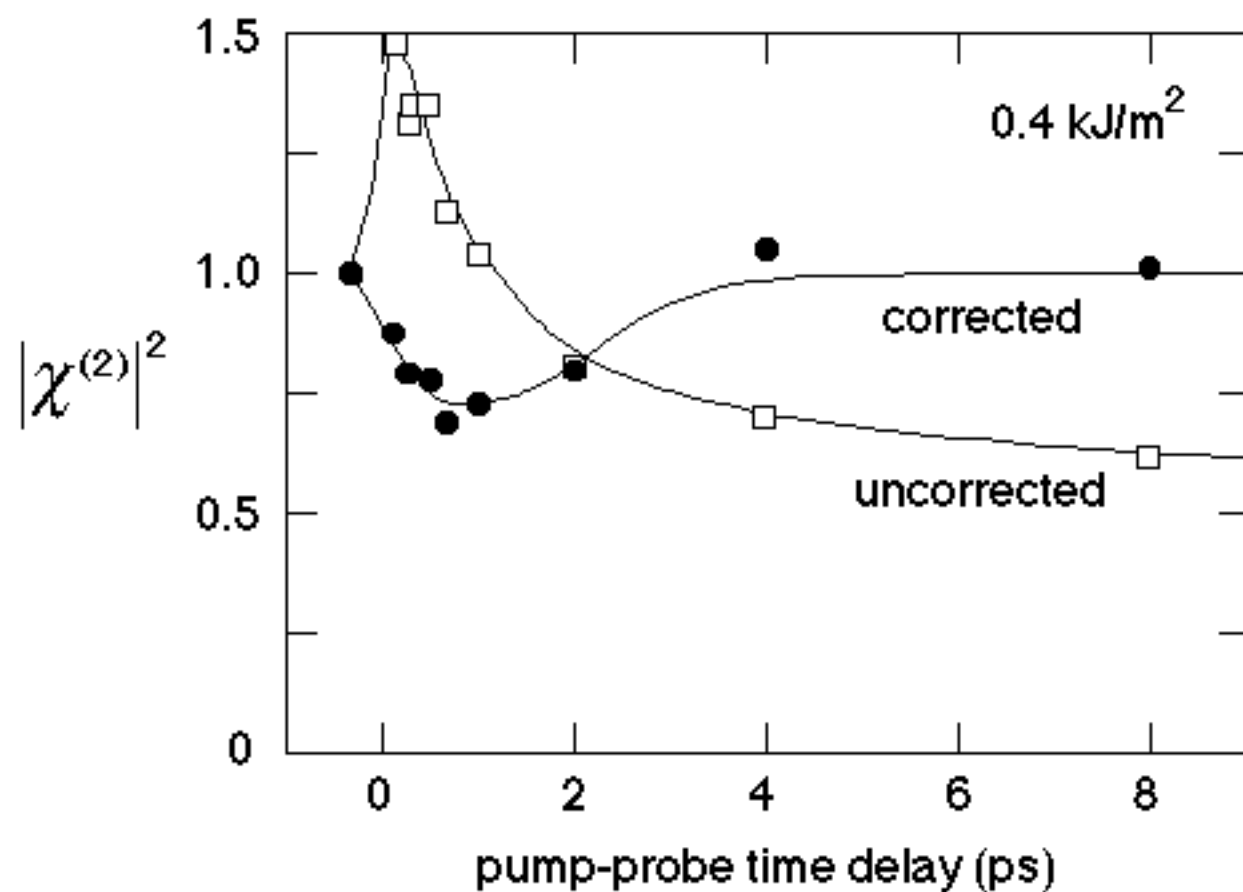
# SECOND HARMONIC DATA



# SECOND HARMONIC DATA



# EFFECT OF CHANGES IN $\epsilon$



## *NONLINEAR WORK*

- $\chi^{(2)}$  sensitive to loss of long-range order
- long-range order disappears even below threshold
- structural change due to destabilization of covalent bonds





- ① Background
- ② Dielectric function
- ③ Nonlinear properties



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



## SOME QUESTIONS

- ▶ how hot does the plasma inside the material get?
- ▶ is only long-range order lost, or also short-range order?
- ▶ how can we study the atomic-scale physics?



## Acknowledgments

Prof. Bloembergen  
Prof. Ehrenreich  
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Prof. Aziz

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NSF DMR 89-20490

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