



ULTRAFAST LASER-INDUCED STRUCTURAL AND ELECTRONIC CHANGES IN SOLIDS

Eli Glezer

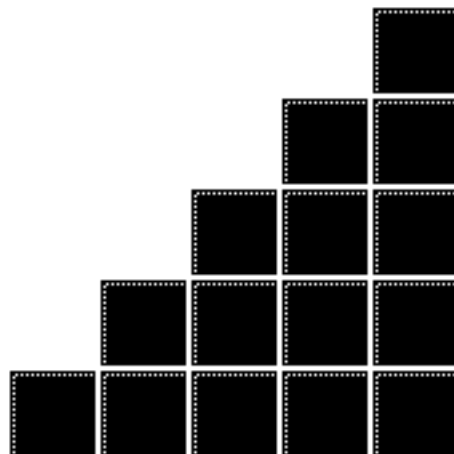
Li Huang

Paul Callan

PIERS 1997

Hong Kong

7 January 1997



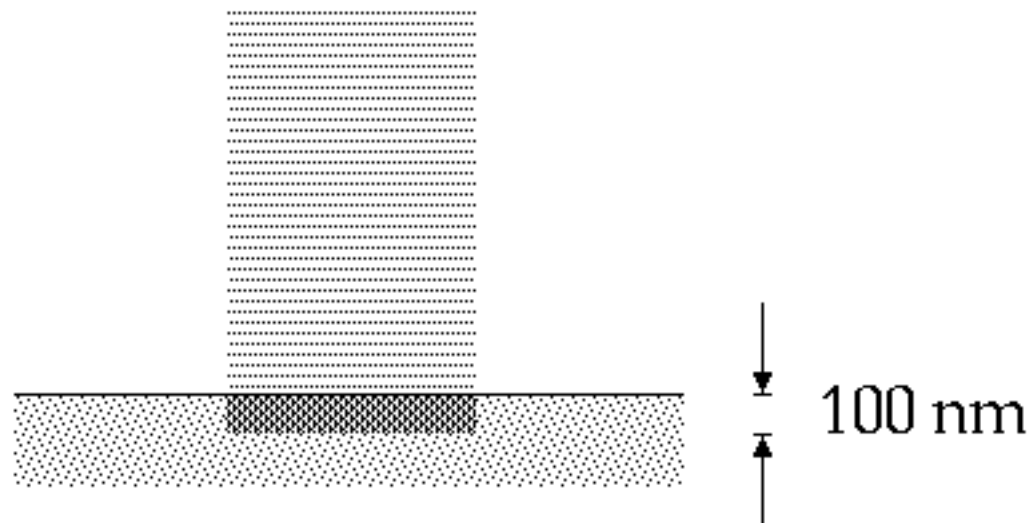
INTRODUCTION

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



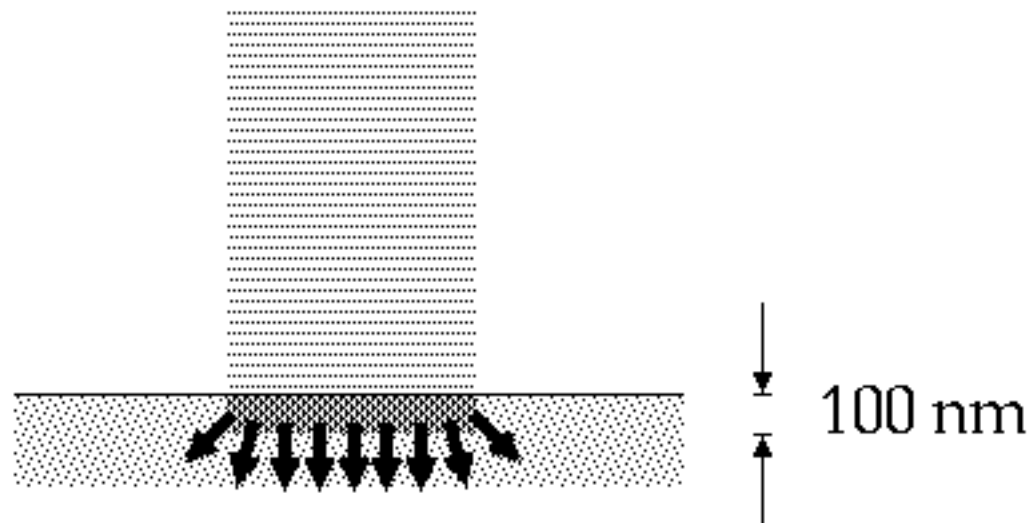
INTRODUCTION

laser deposits energy near surface...



INTRODUCTION

energy rapidly diffuses into bulk...

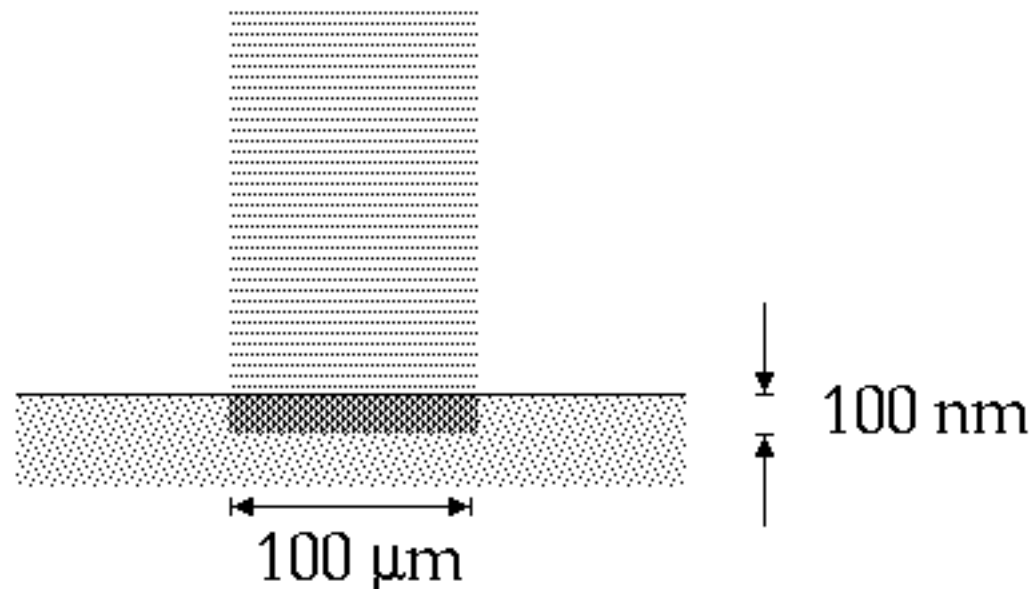


in 1 ns, diffusion length ≈ 100 nm



INTRODUCTION

For $t < 1$ ns can ignore diffusion



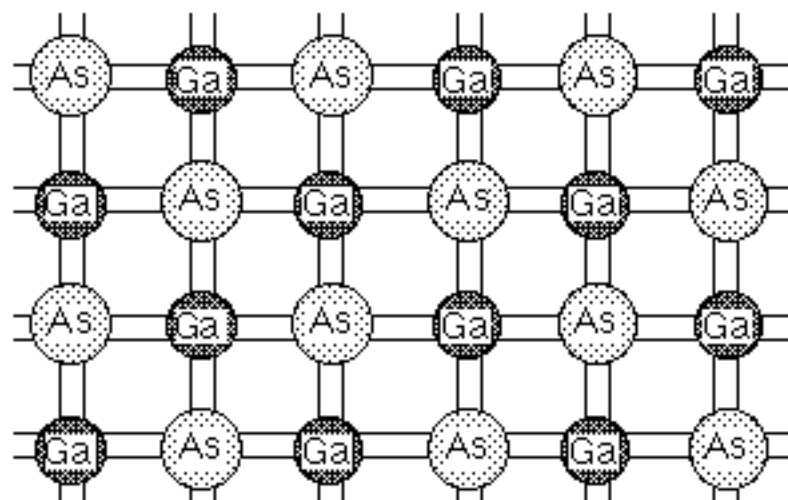
$10\ \mu\text{J}$ into a volume of about $10^{-15}\ \text{m}^3$!



- ① Background
- ② Single frequency measurements
- ③ Detailed Picture



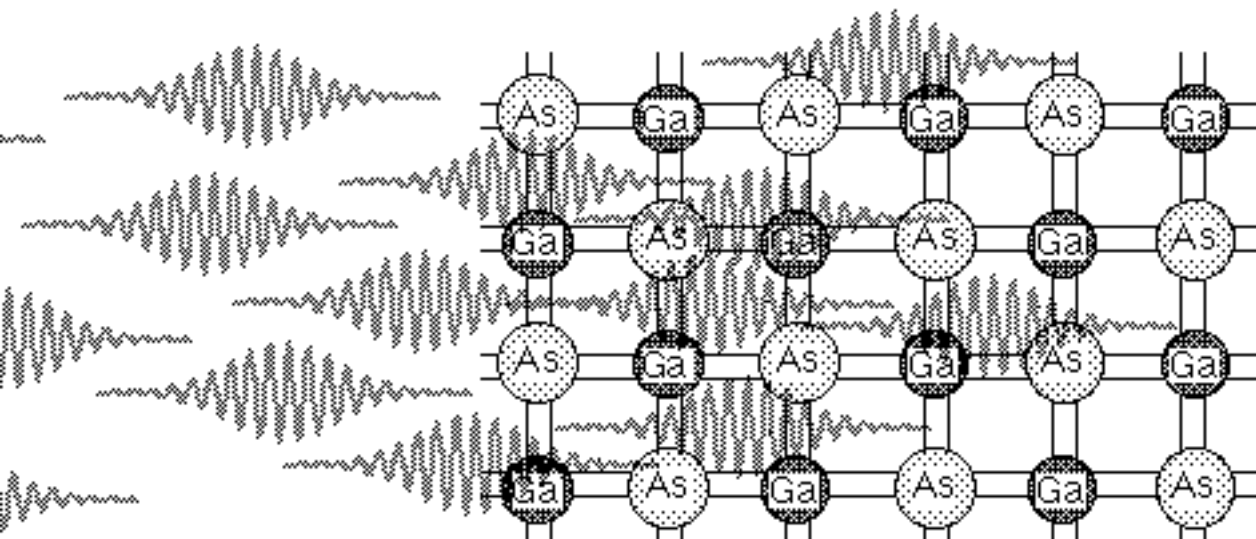
EARLY MODELS



how does light melt a solid?



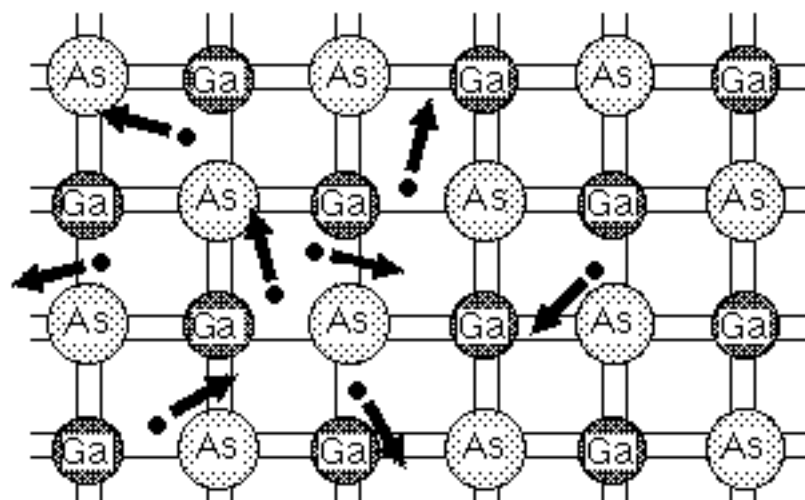
THERMAL MODEL



photons excite valence electrons...



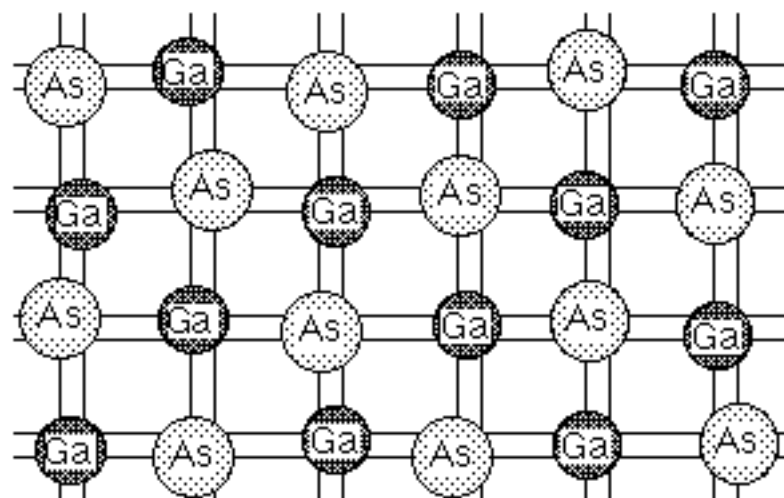
THERMAL MODEL



...and create hot electrons...



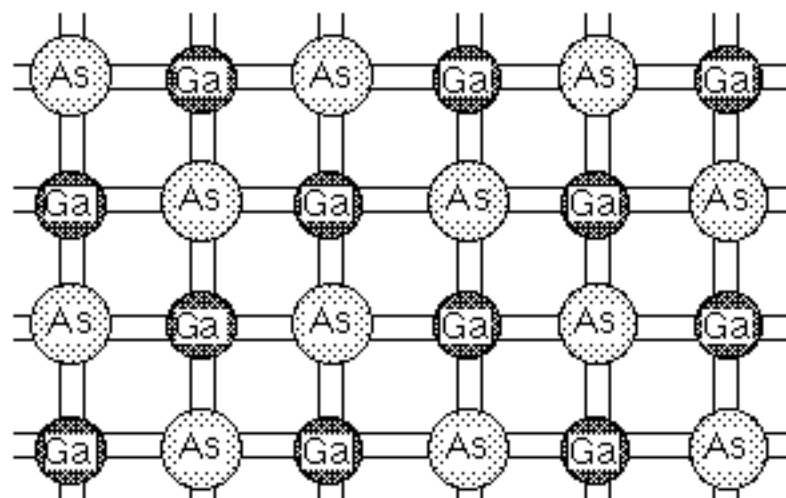
THERMAL MODEL



...which heat lattice



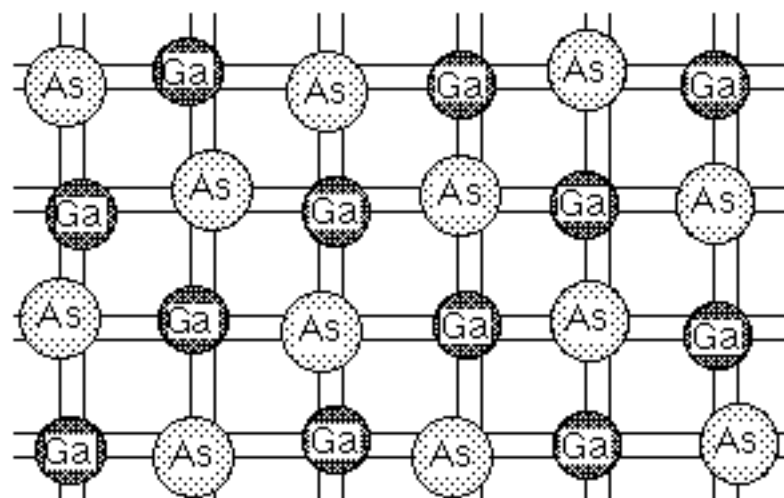
THERMAL MODEL



...which heat lattice



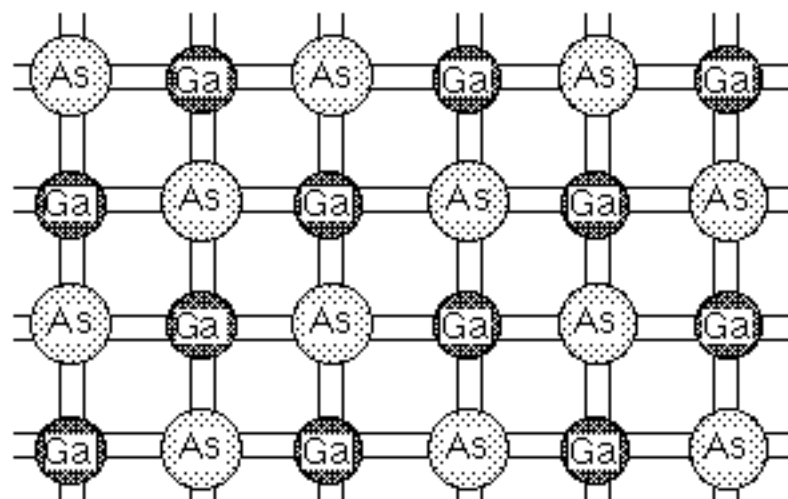
THERMAL MODEL



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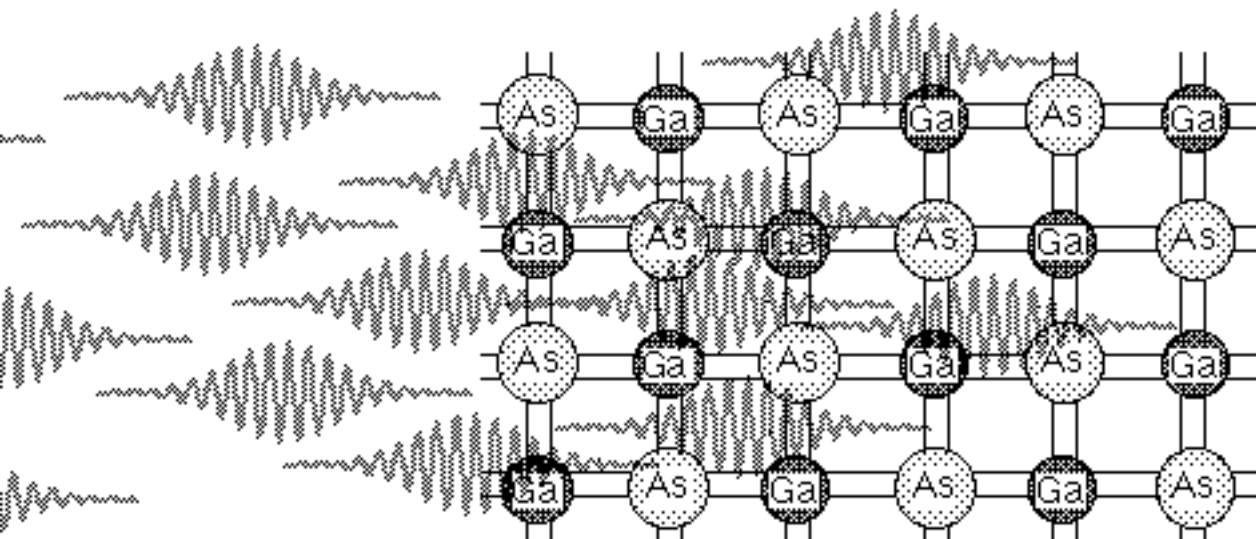
EARLY MODELS



...Or...



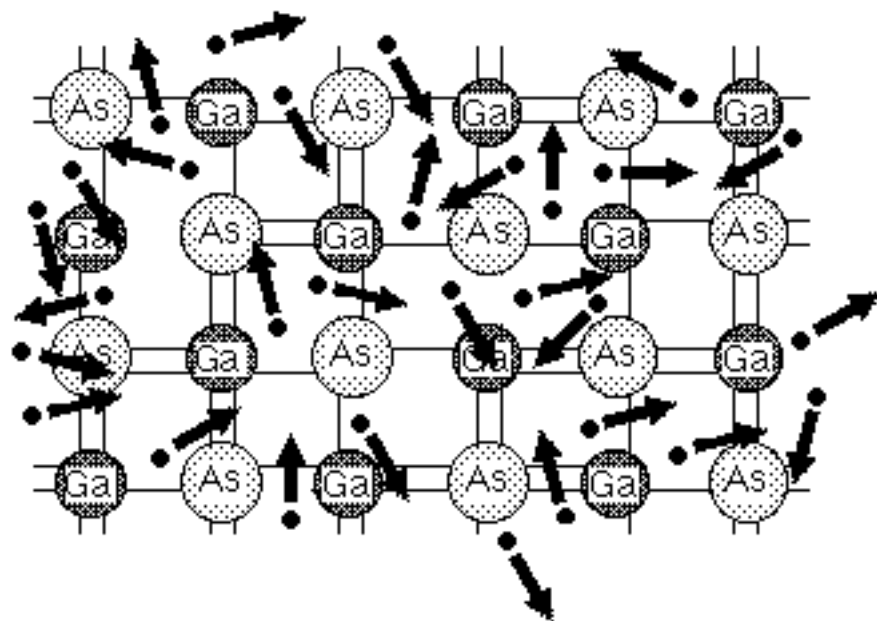
PLASMA ANNEALING



photons excite valence electrons...



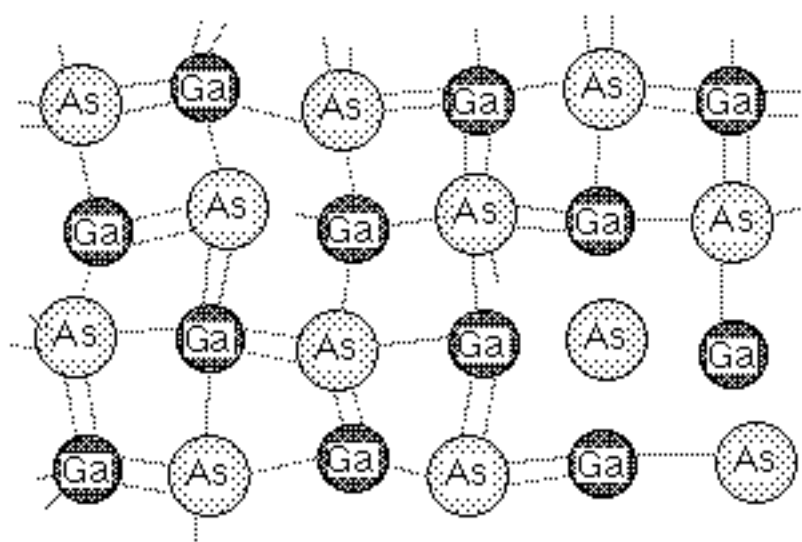
PLASMA ANNEALING



...creating a dense plasma...



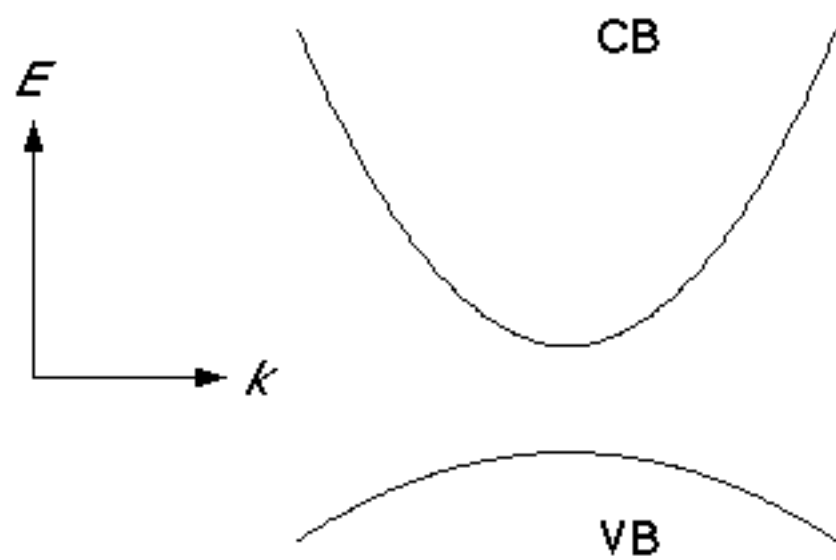
PLASMA ANNEALING



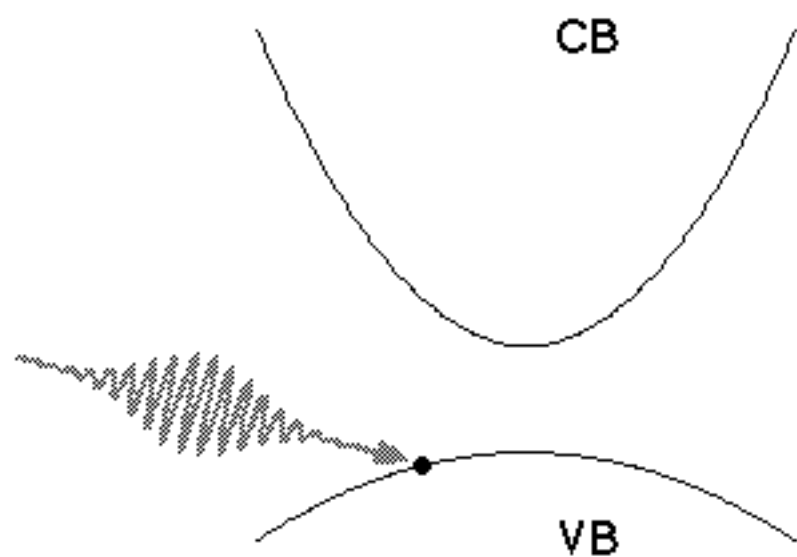
...and breaking bonds



EXCITATION



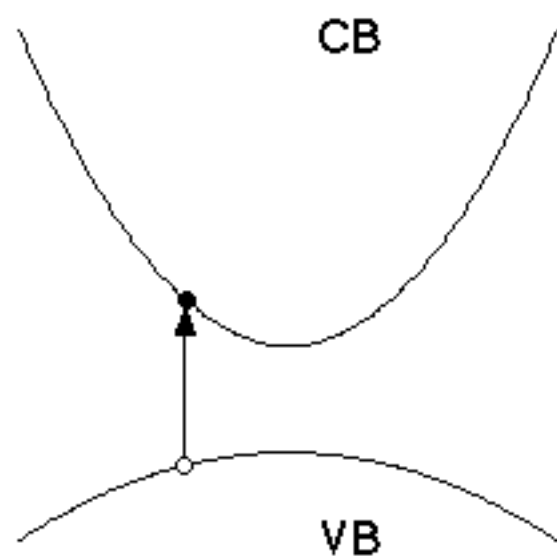
EXCITATION



photon excites valence electron



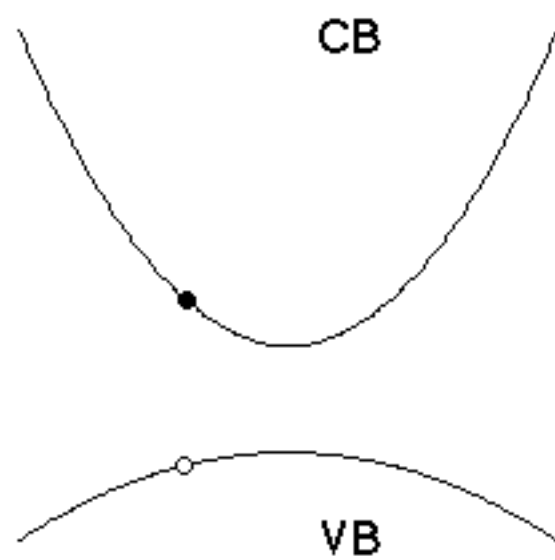
EXCITATION



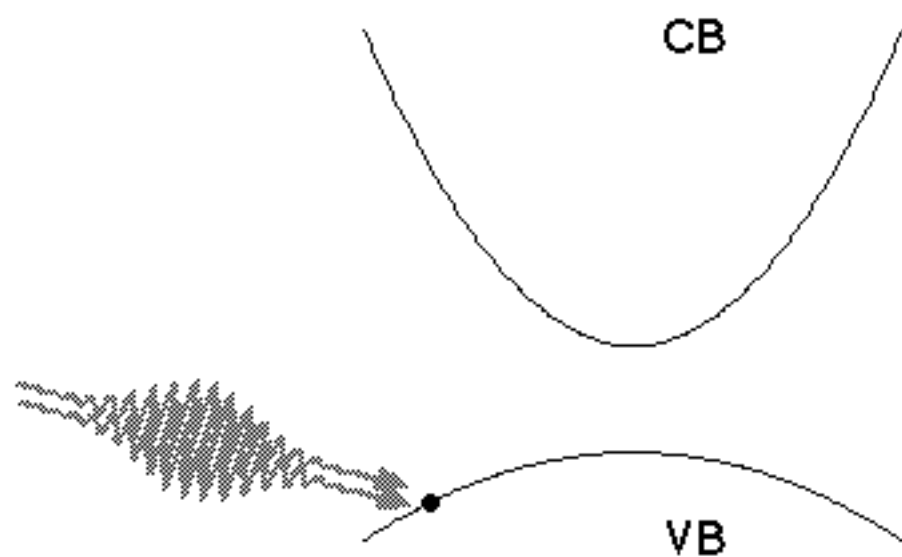
creating an electron-hole pair



EXCITATION



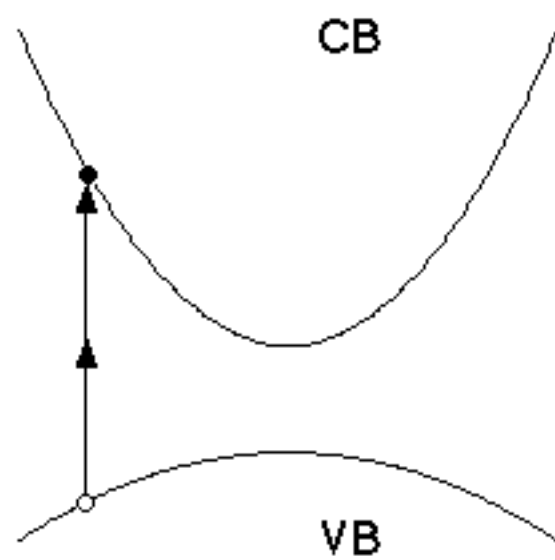
EXCITATION



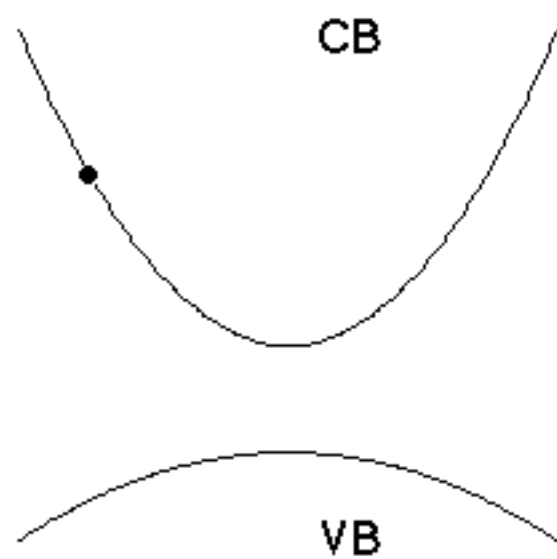
multiphoton excitation



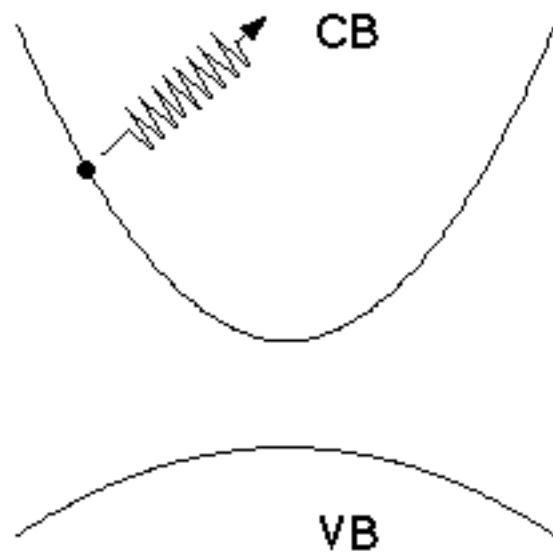
EXCITATION



EXCITATION



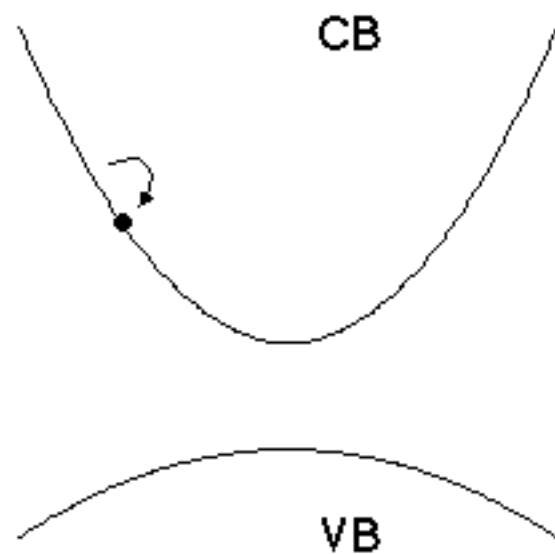
RELAXATION



emission of LO phonons



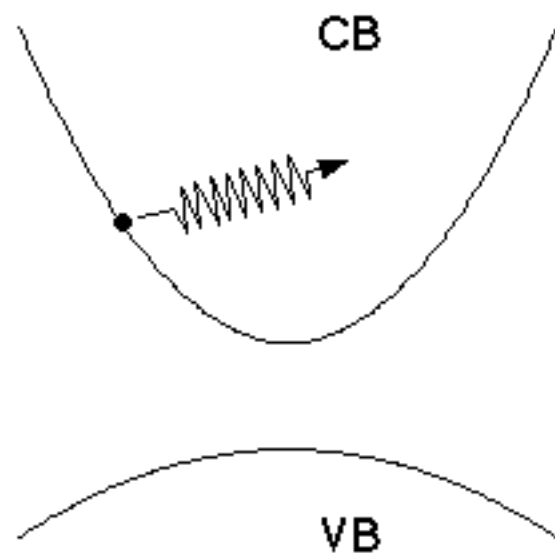
RELAXATION



emission of LO phonons



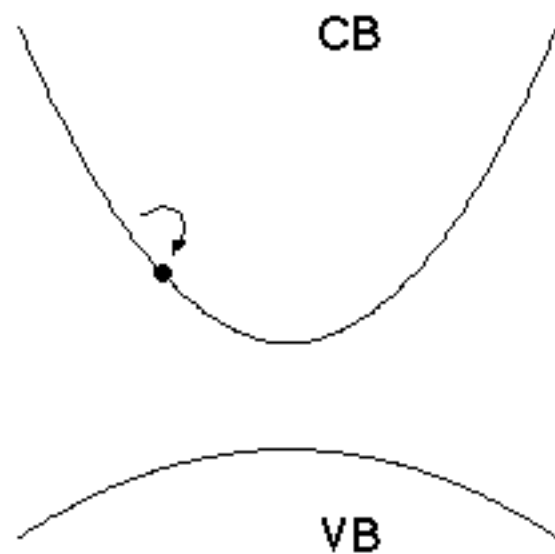
RELAXATION



emission of LO phonons



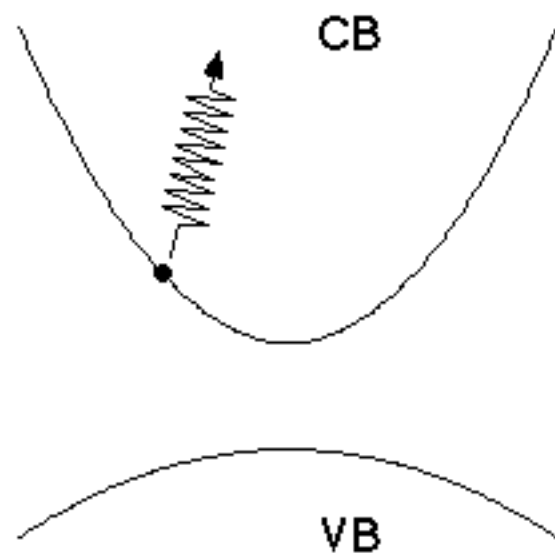
RELAXATION



emission of LO phonons



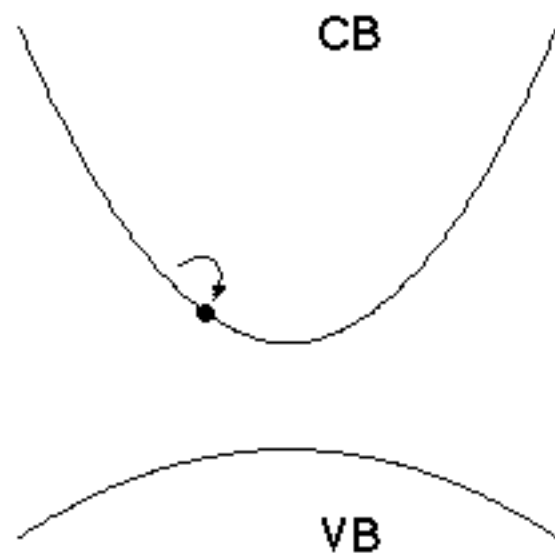
RELAXATION



emission of LO phonons



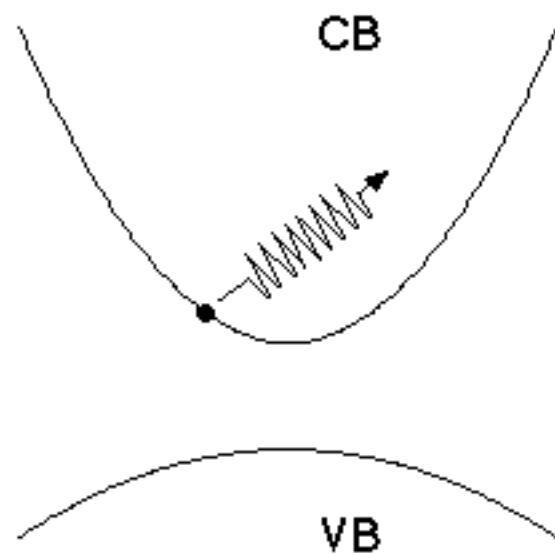
RELAXATION



emission of LO phonons



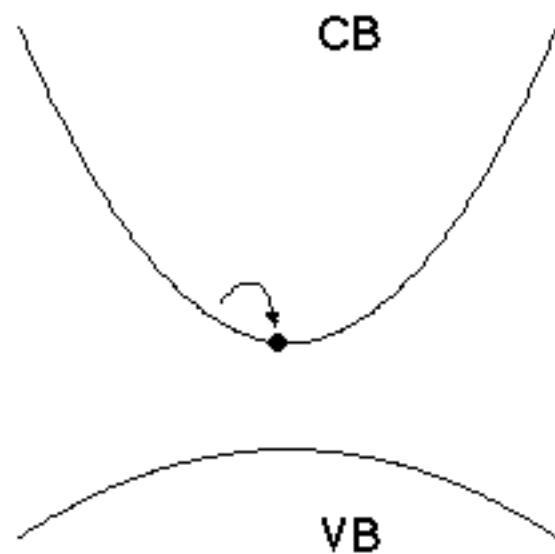
RELAXATION



emission of LO phonons



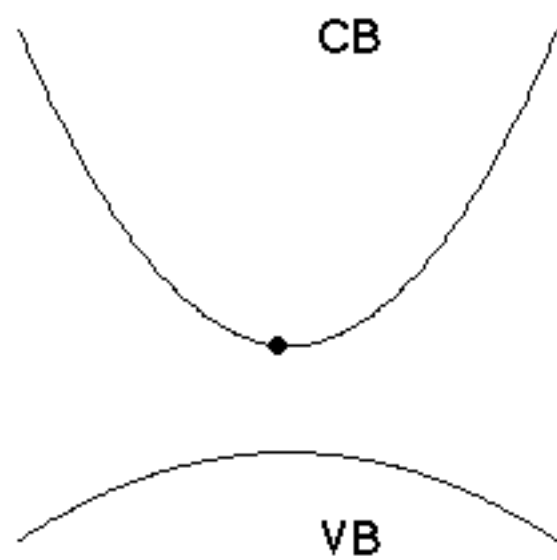
RELAXATION



emission of LO phonons

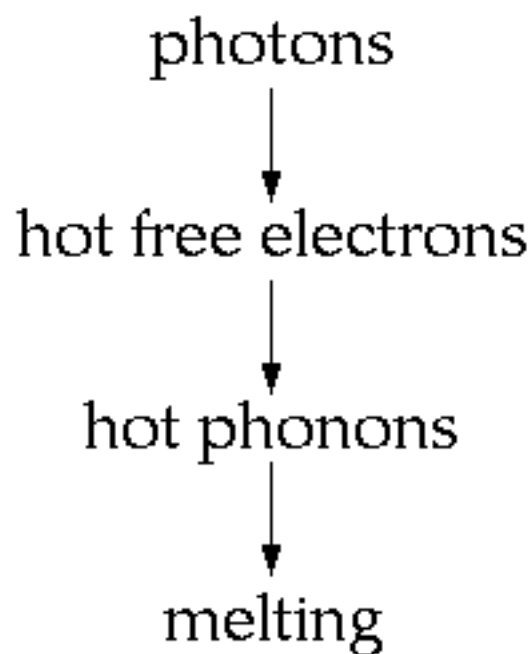


RELAXATION



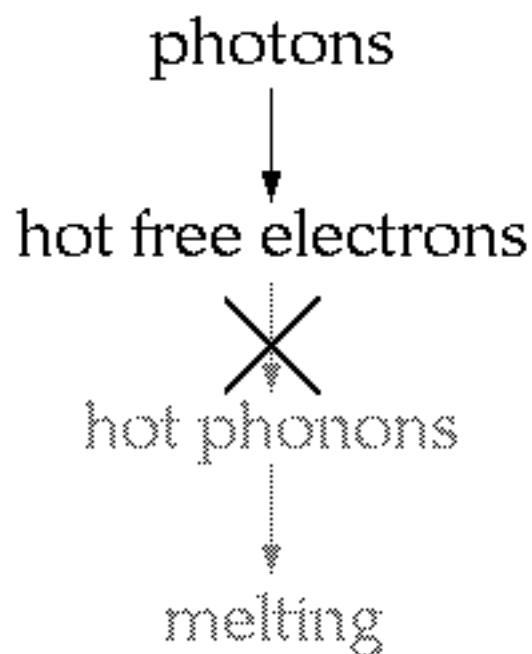
TIME SCALE

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



TIME SCALE

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

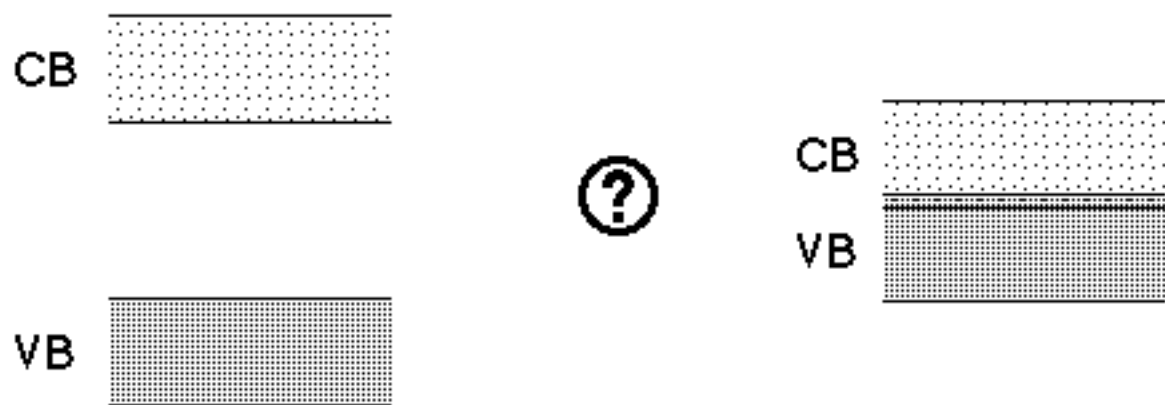


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- ② Single frequency measurements
- ③ Detailed Picture



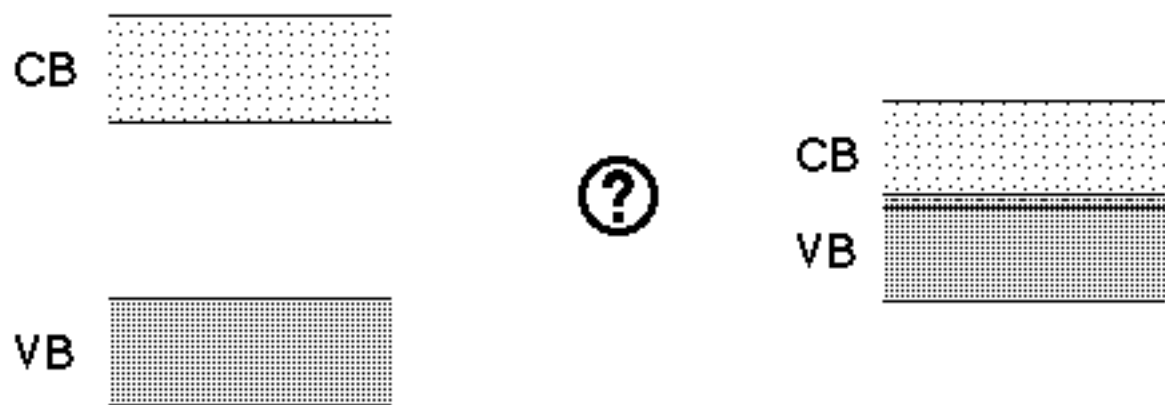
FEMTOSECOND WORK

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



FEMTOSECOND WORK

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FEMTOSECOND WORK

intrinsic properties

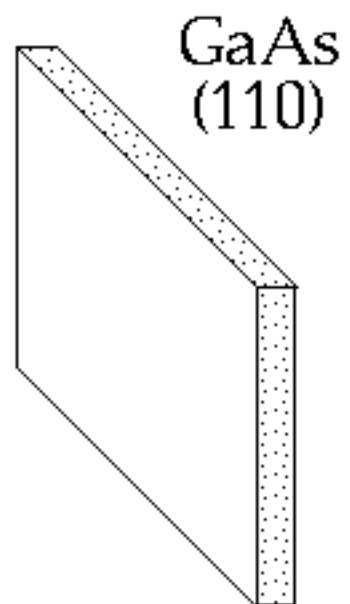
measured properties

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

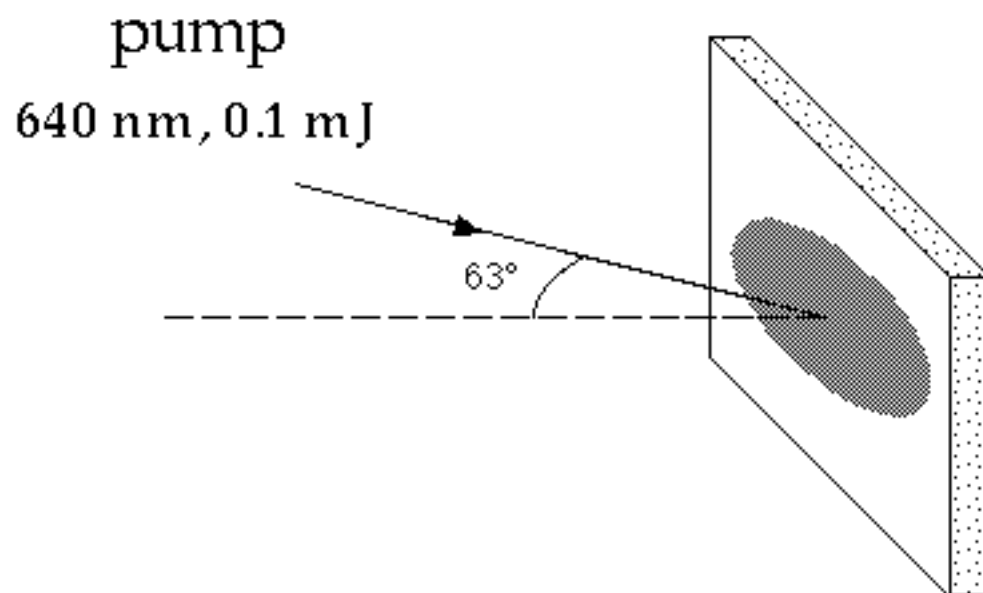
$$\begin{array}{l} \epsilon', \epsilon''(\omega) \\ \epsilon', \epsilon''(2\omega) \\ \chi^{(2)} \end{array} \longrightarrow \text{SHG}$$



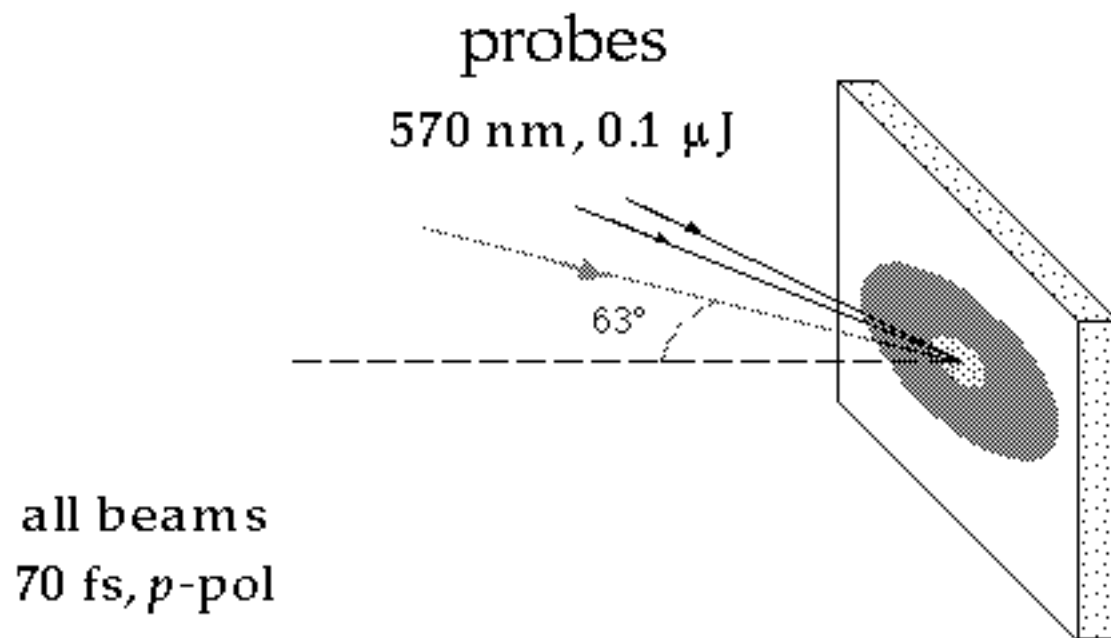
2.2 eV DIELECTRIC CONSTANT



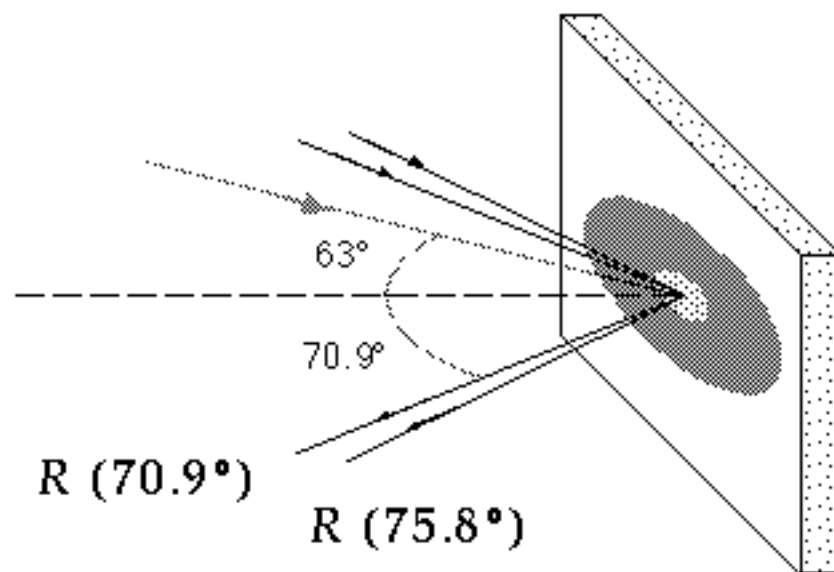
2.2 eV DIELECTRIC CONSTANT



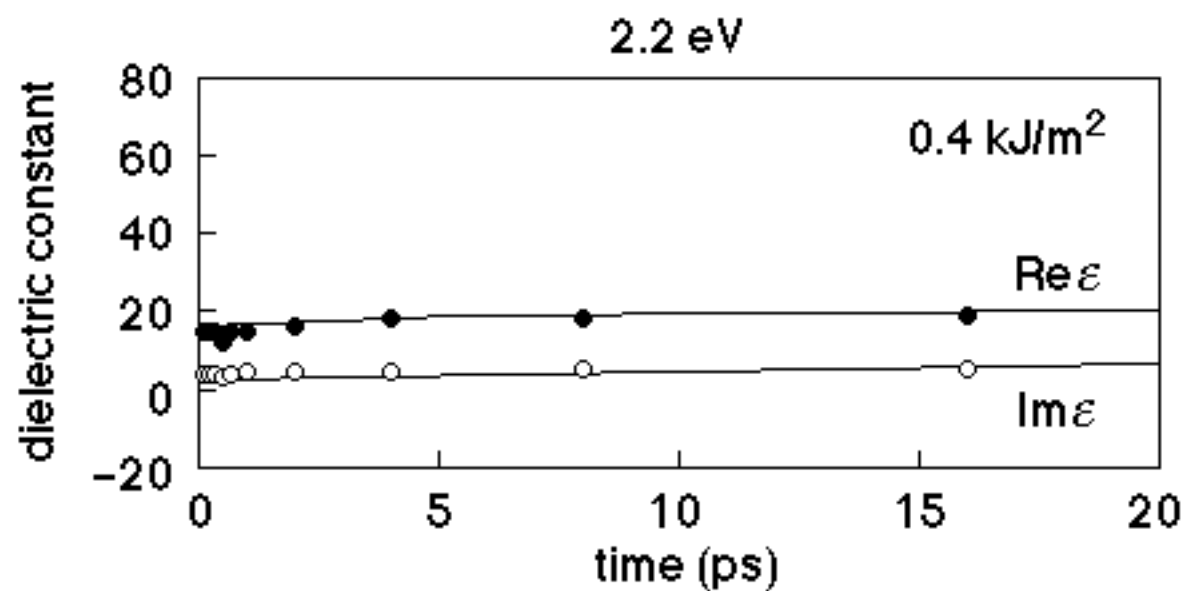
2.2 eV DIELECTRIC CONSTANT



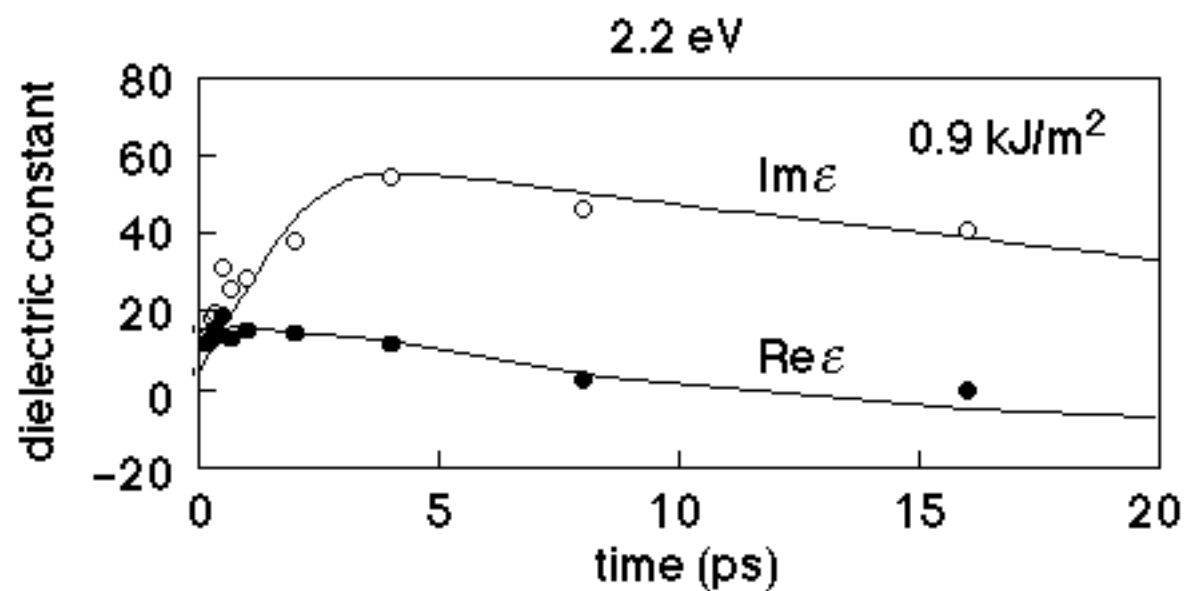
2.2 eV DIELECTRIC CONSTANT



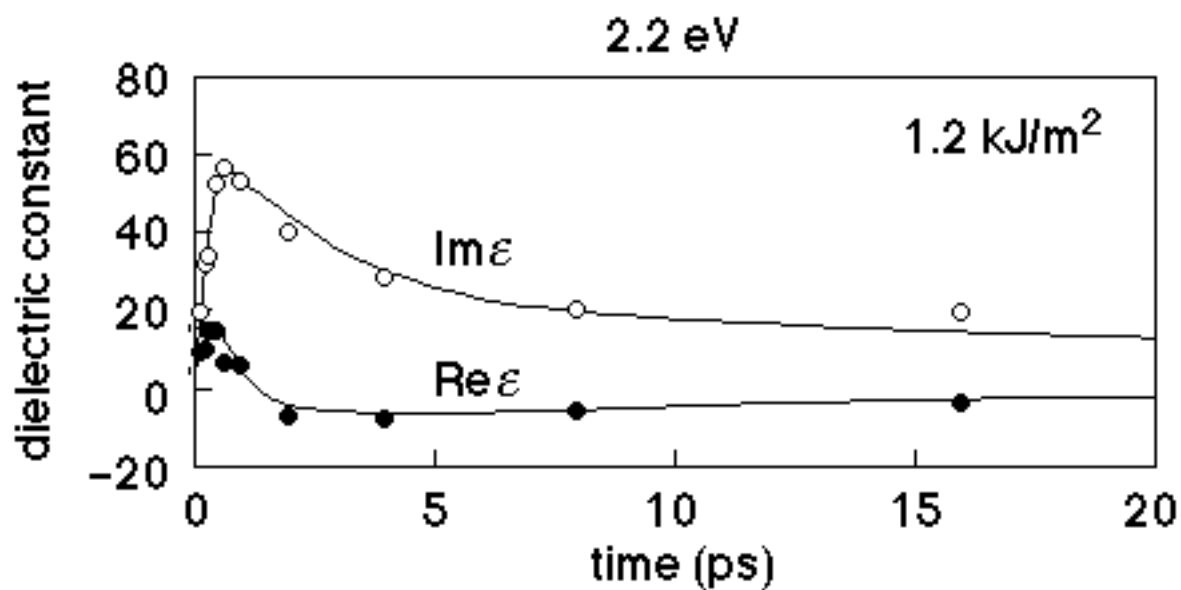
TIME DEPENDENCE



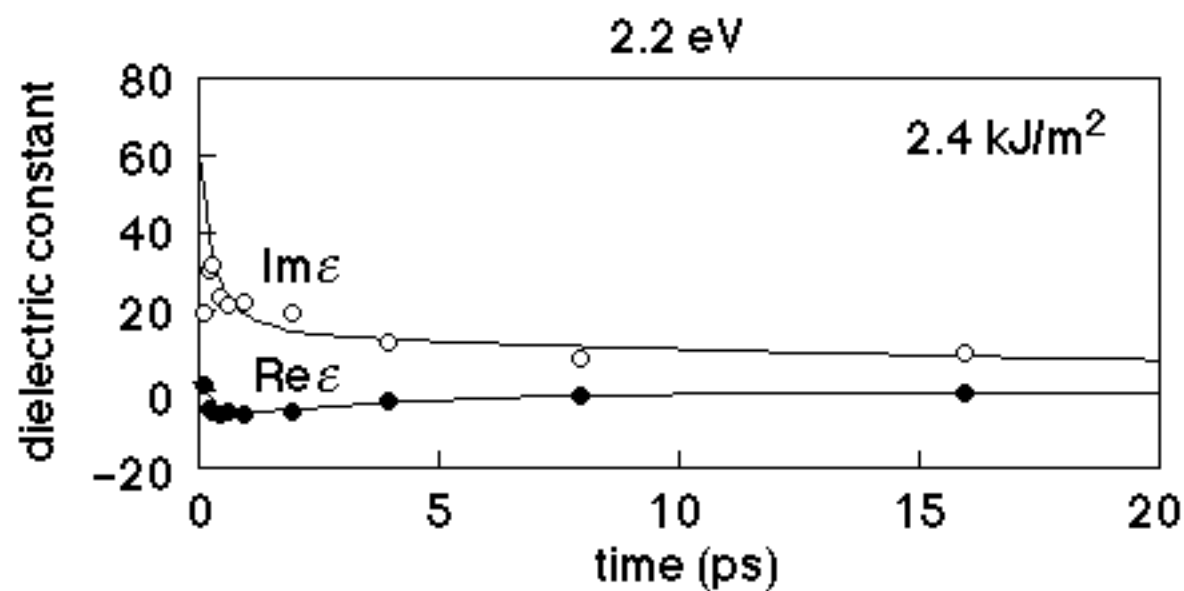
TIME DEPENDENCE



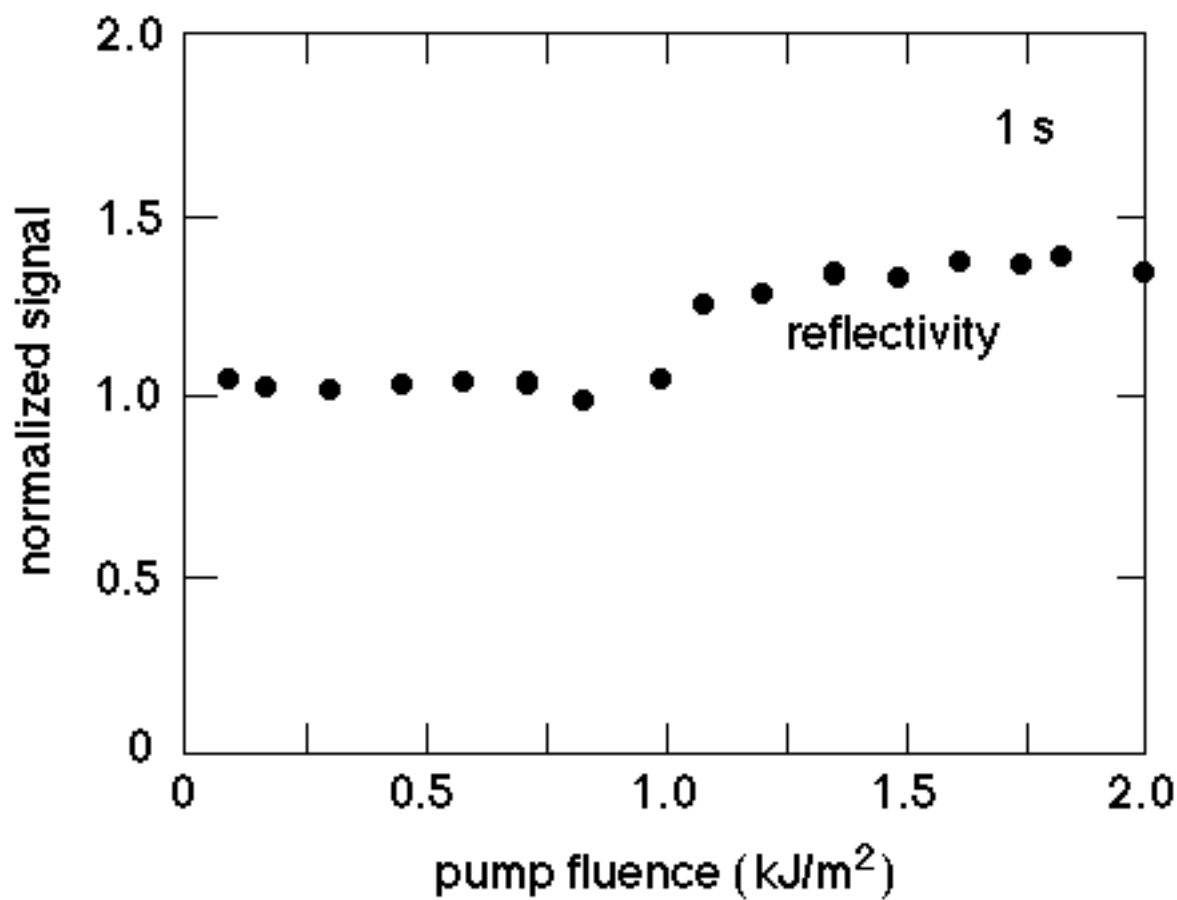
TIME DEPENDENCE



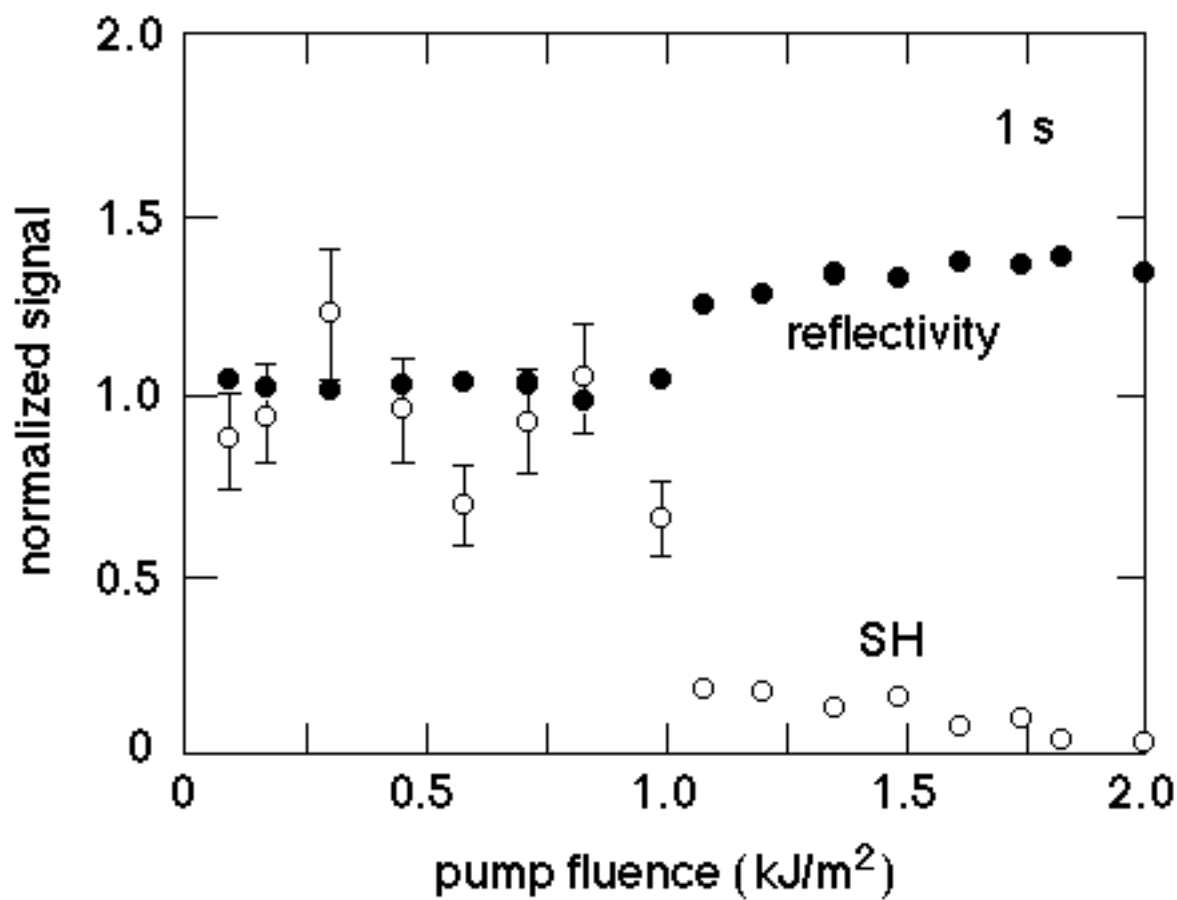
TIME DEPENDENCE



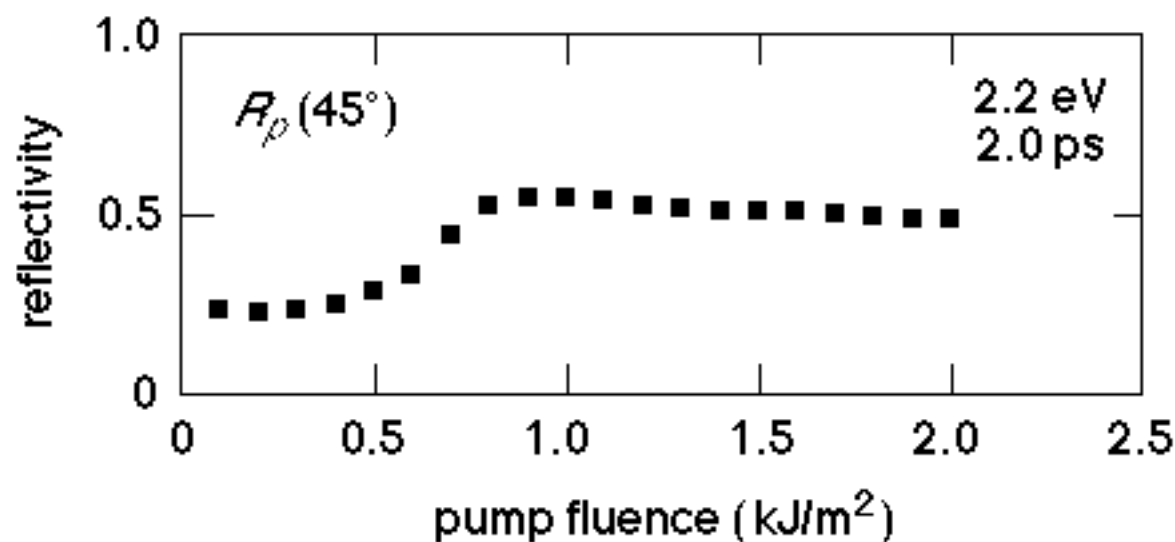
REVERSIBLE vs. IRREVERSIBLE



REVERSIBLE vs. IRREVERSIBLE



SIMPLE MODEL



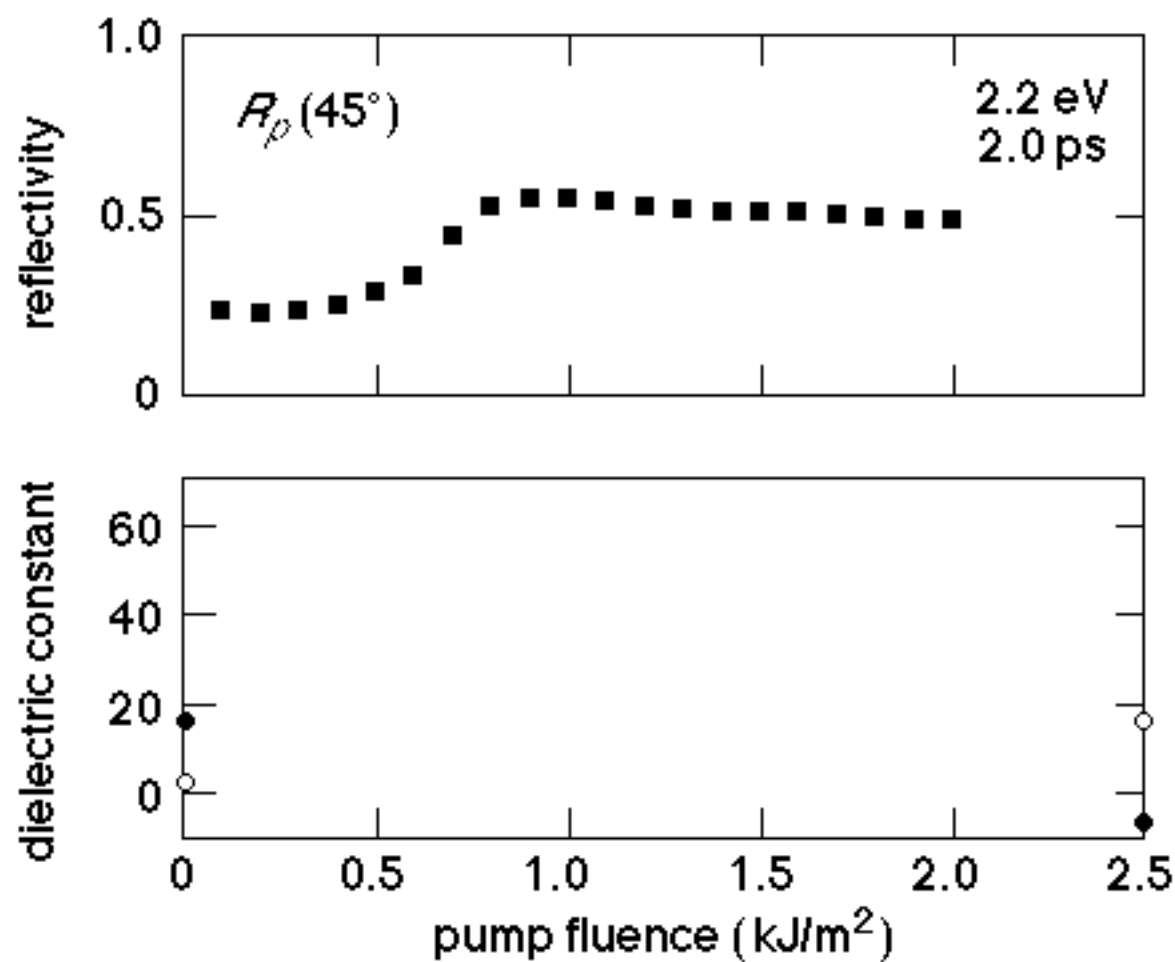
Drude model:

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

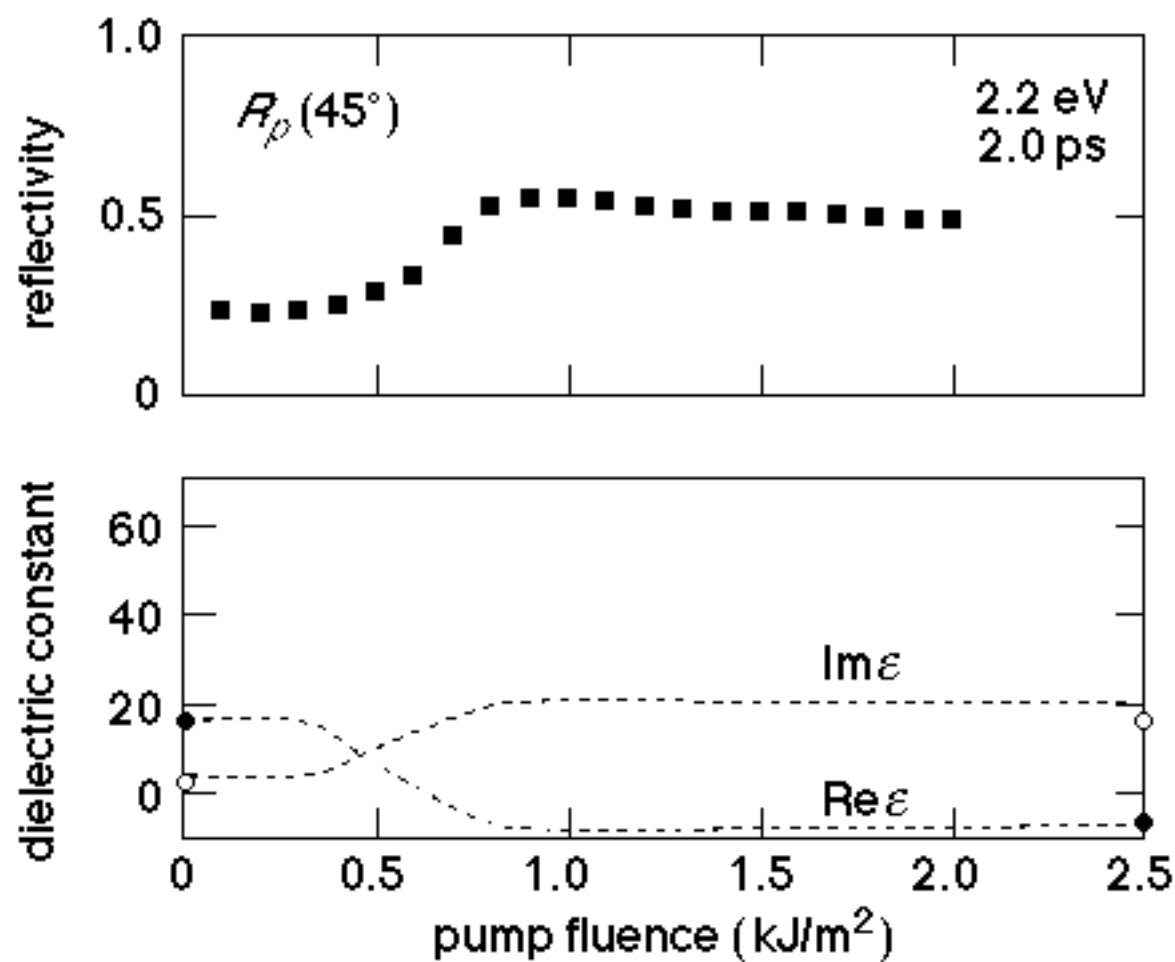
$$\text{Im}(\epsilon) = \frac{Ne^2}{m\epsilon_0} \frac{\gamma\omega}{\omega^4 + \gamma^2\omega^2}$$



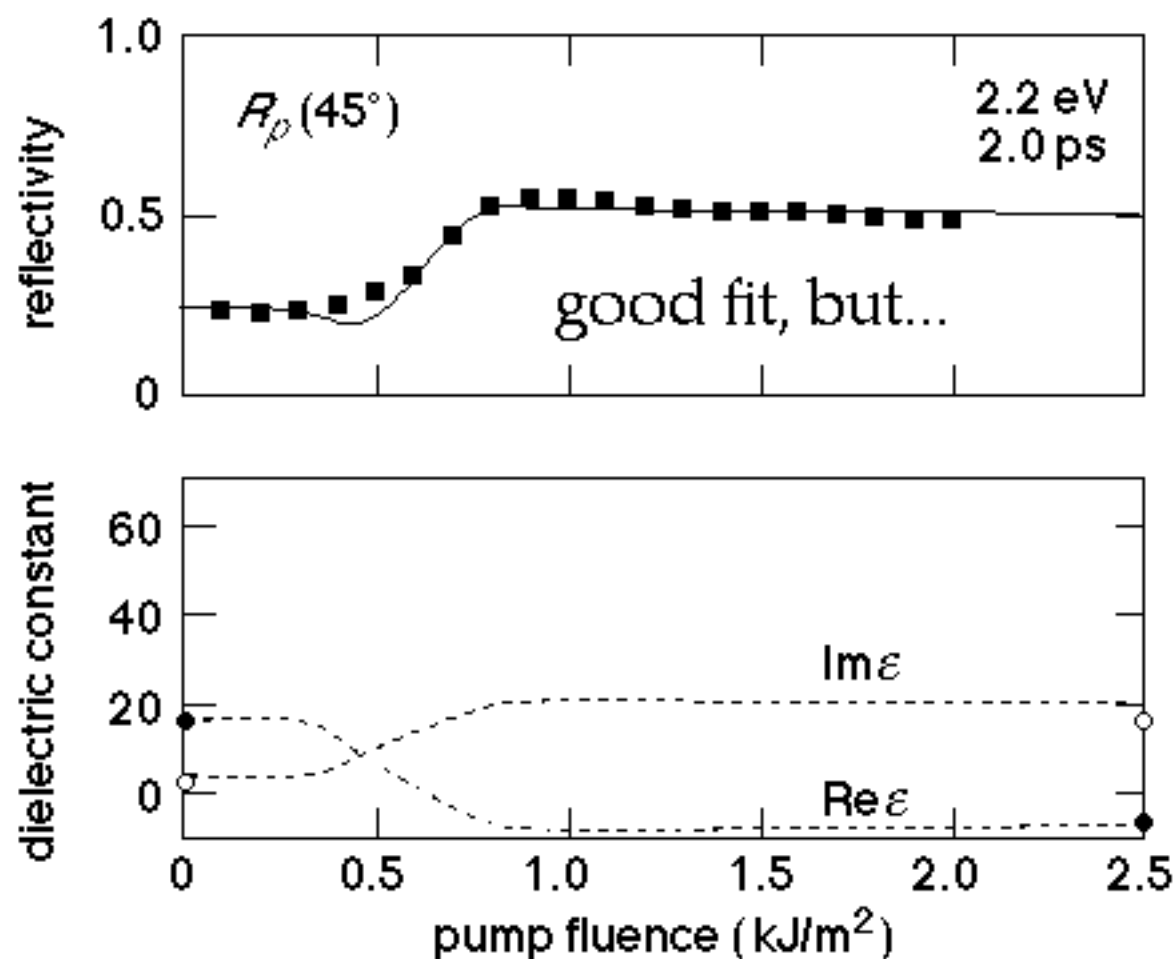
SIMPLE MODEL



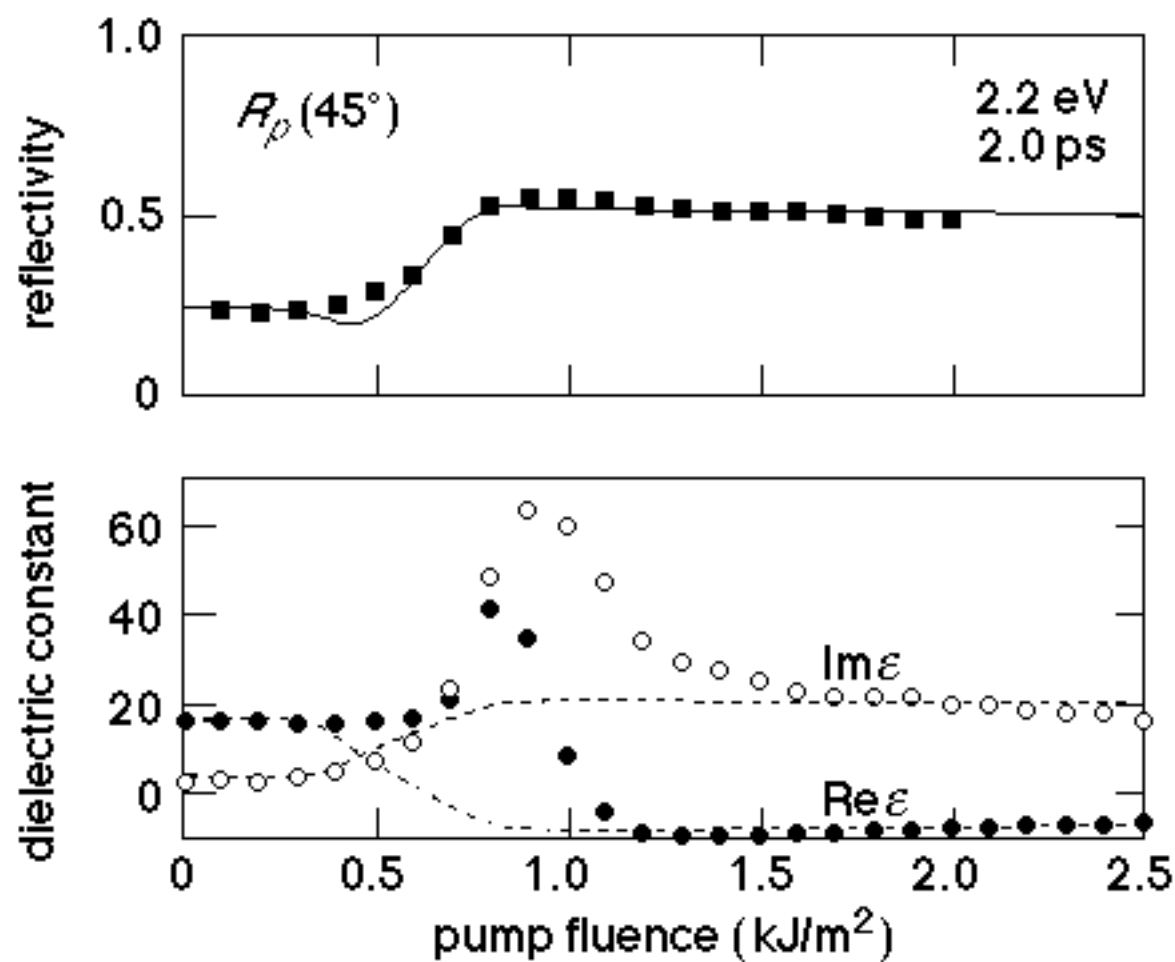
SIMPLE MODEL



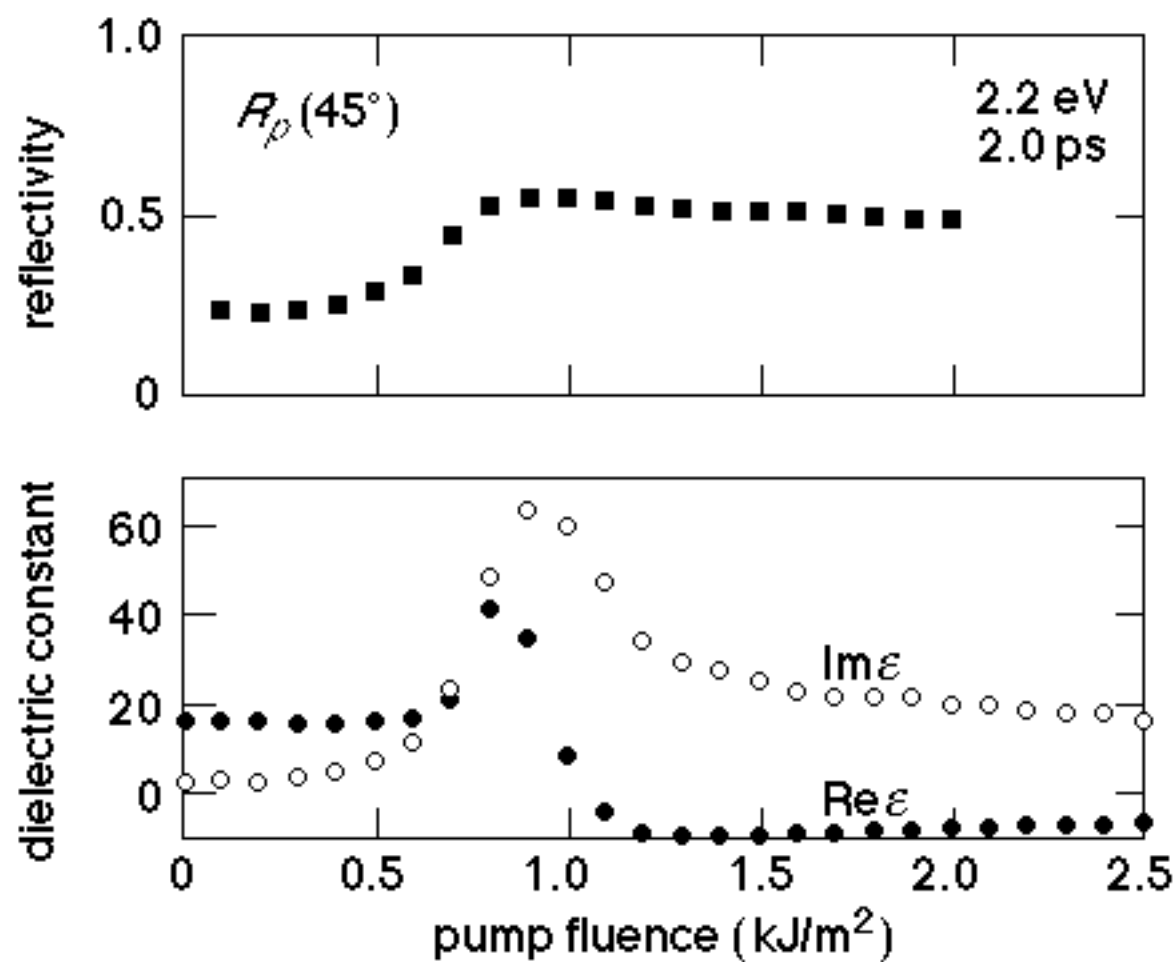
SIMPLE MODEL



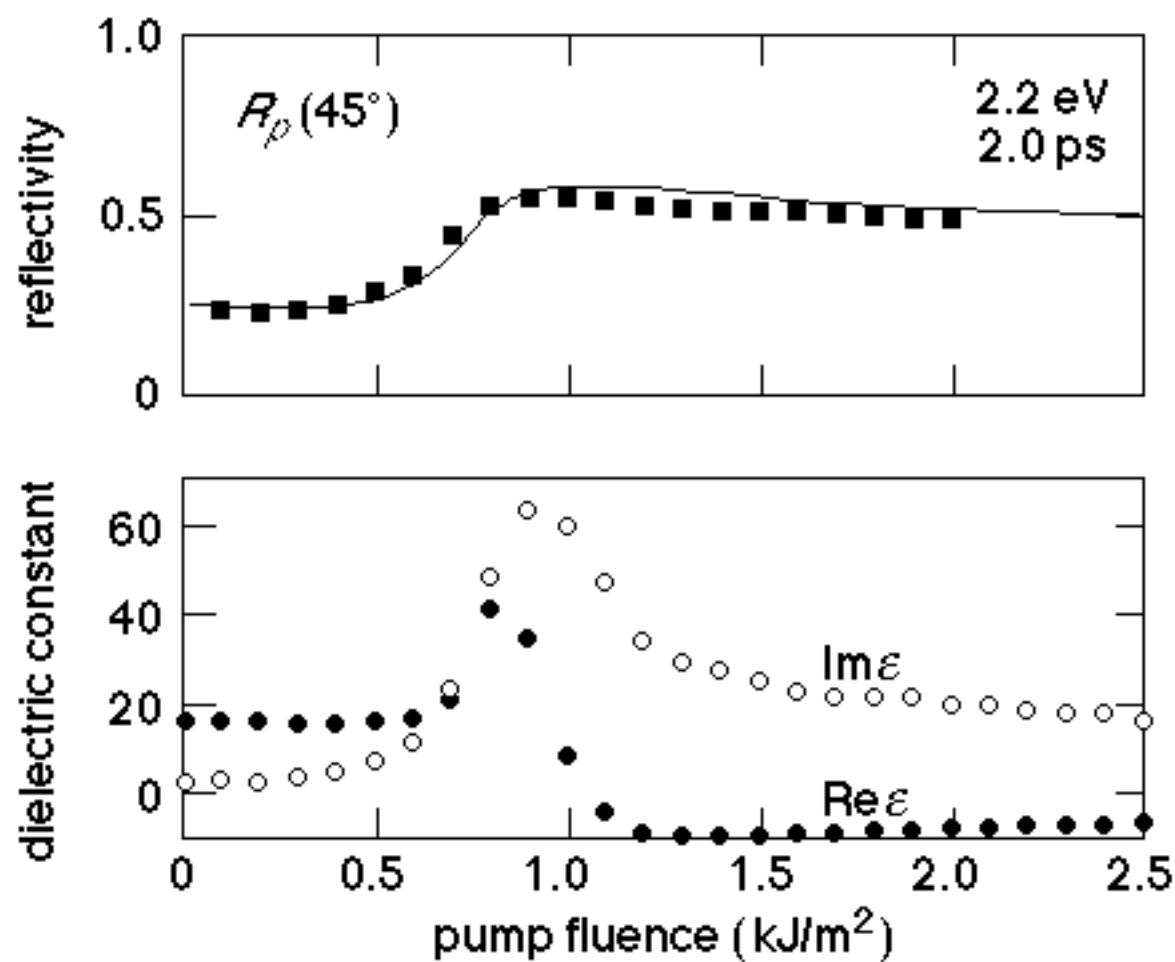
SIMPLE MODEL



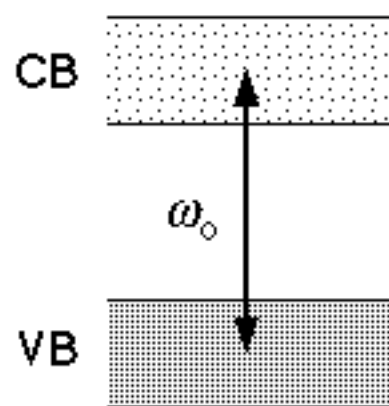
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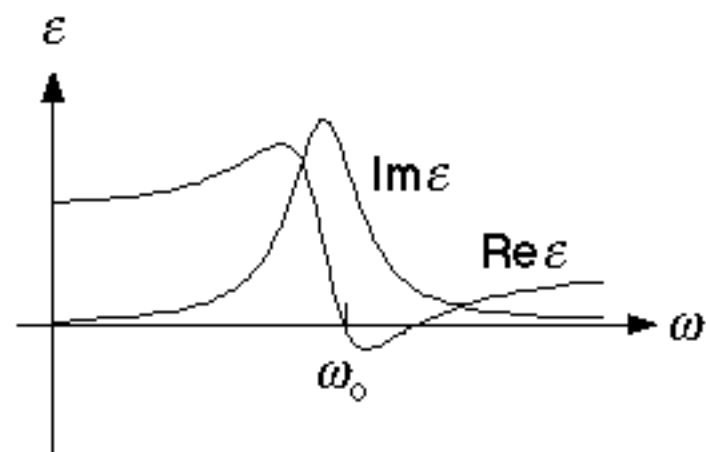
SIMPLE MODEL



WHAT IS HAPPENING?



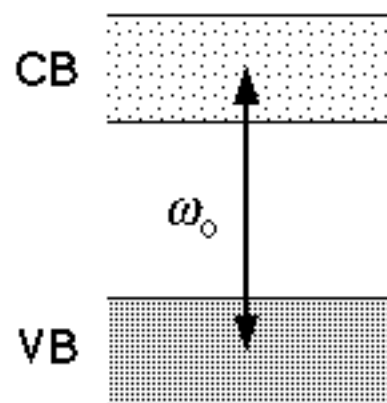
semiconductor



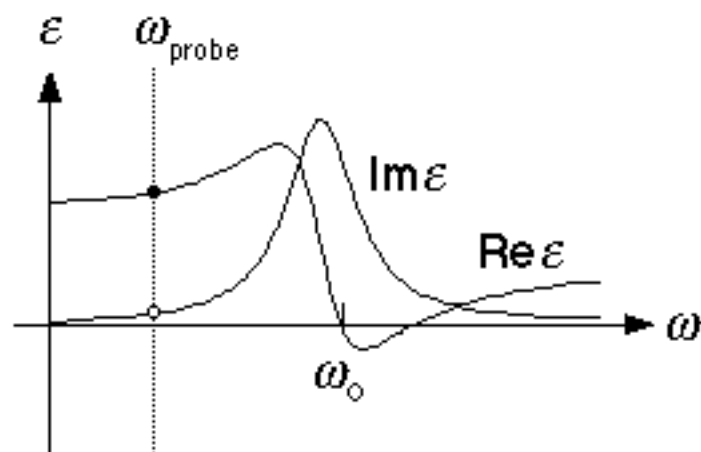
$$\omega_0 \approx 4.5 \text{ eV}$$



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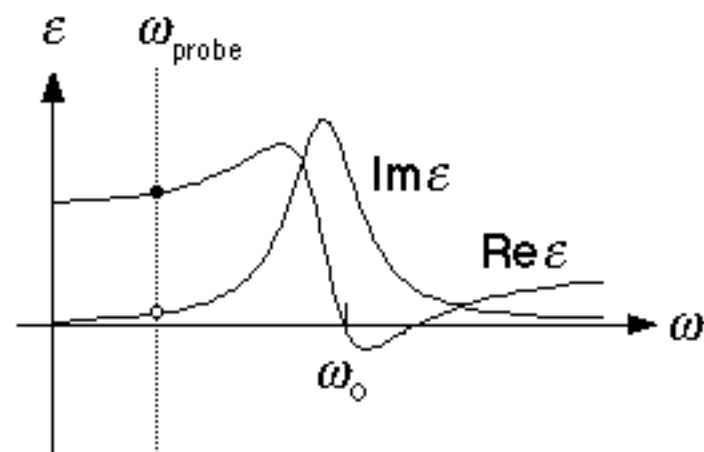
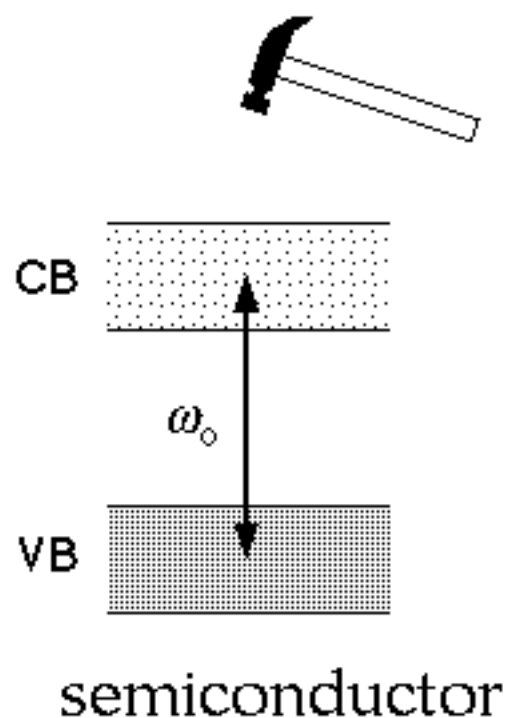
semiconductor



$$\omega_{\text{probe}} = 2.2 \text{ eV}$$



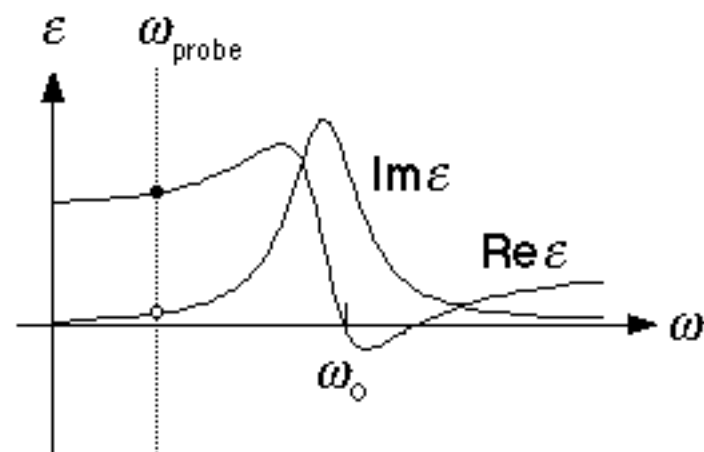
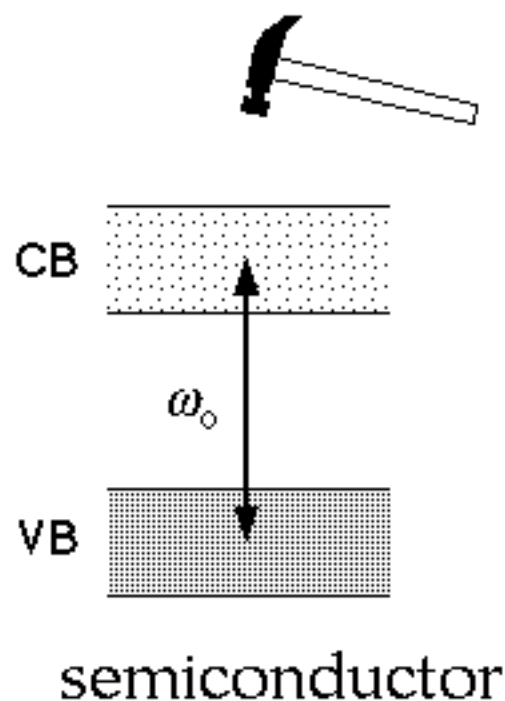
WHAT IS HAPPENING?



pump pulse excites
valence electrons...



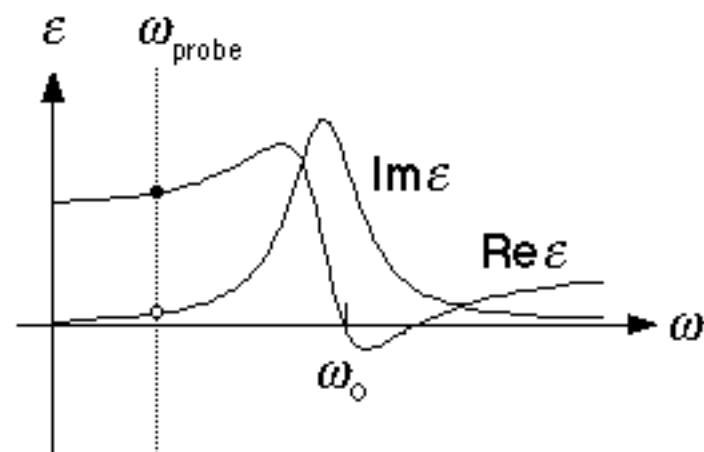
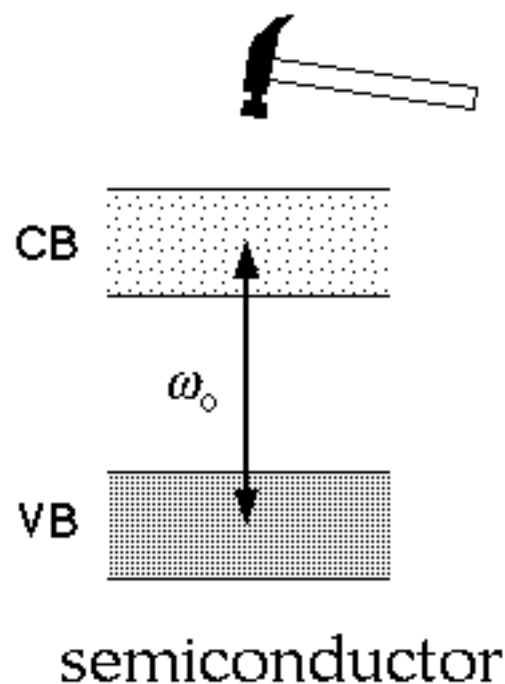
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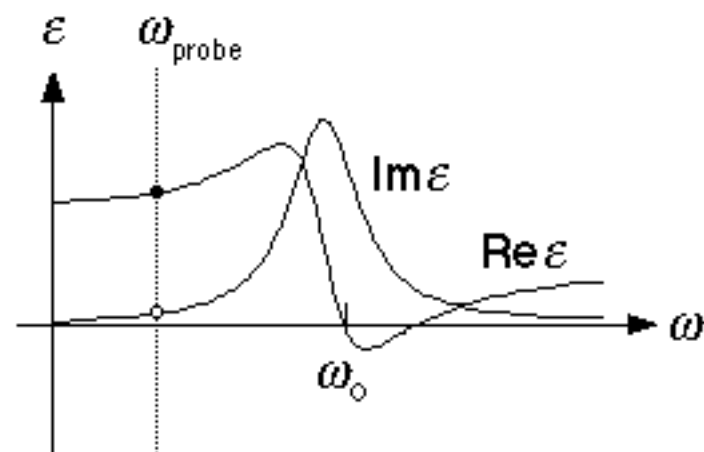
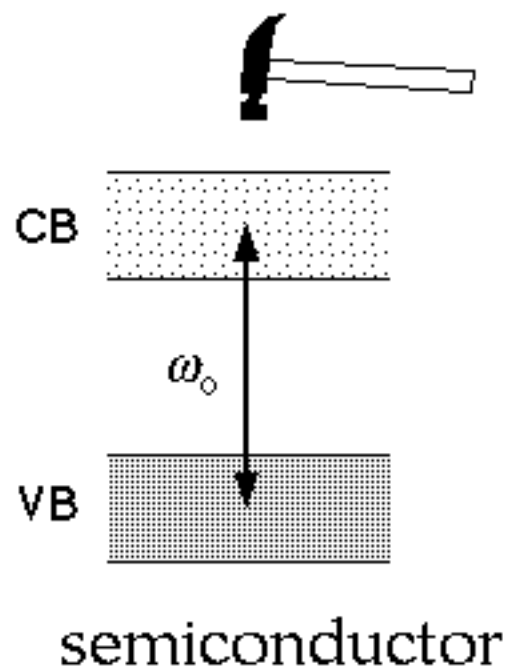
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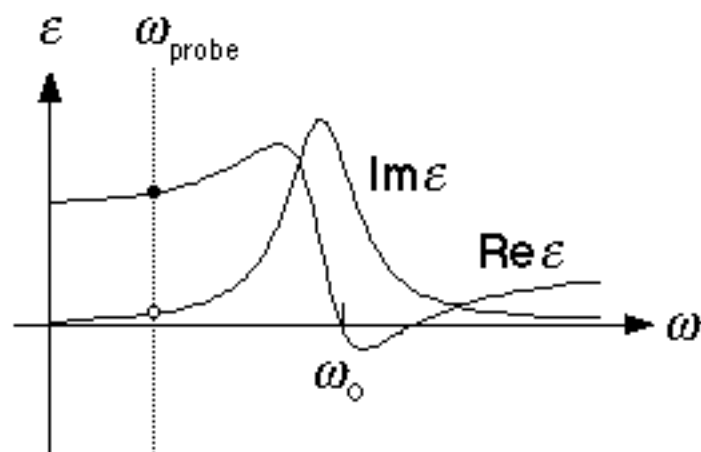
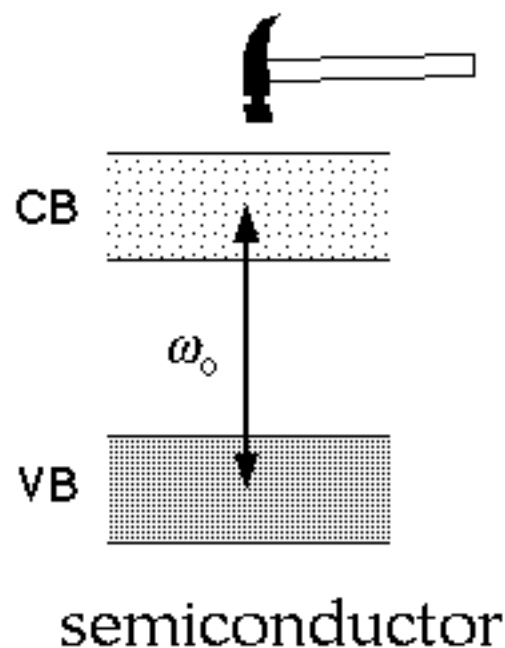
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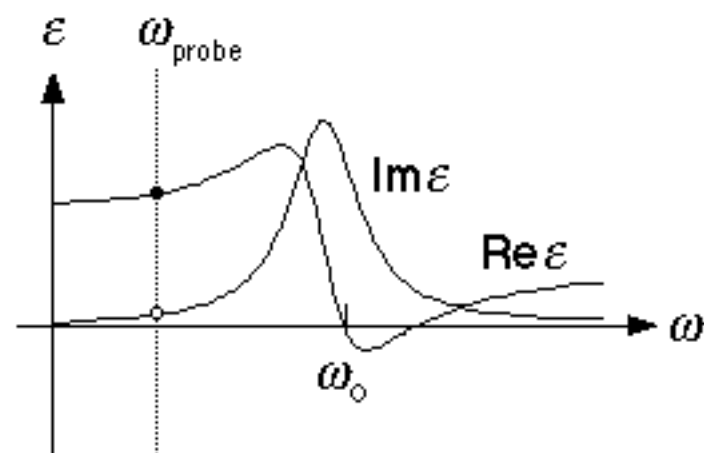
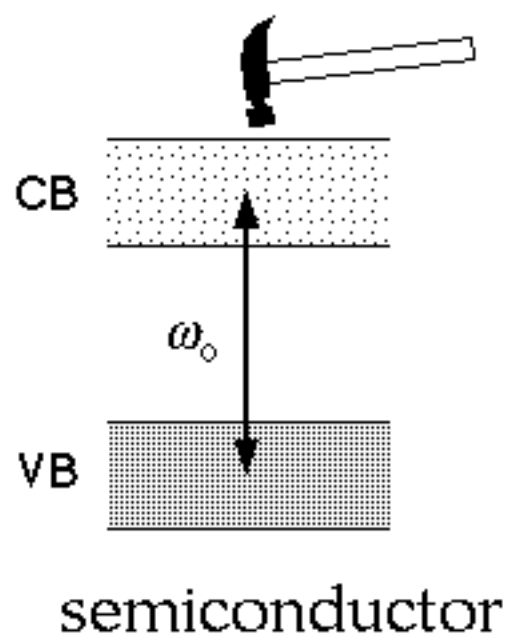
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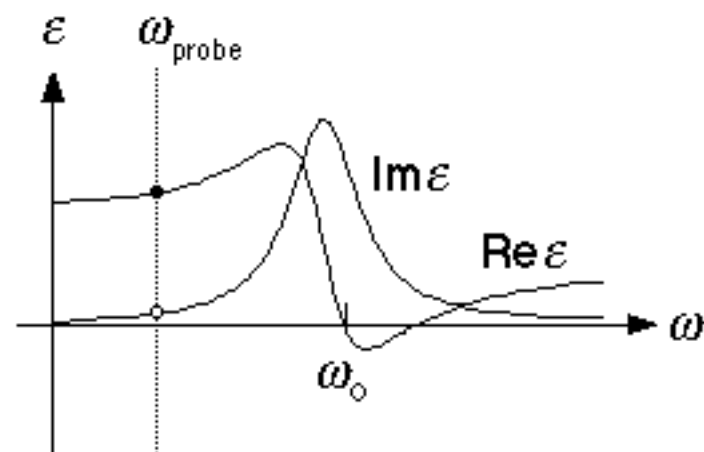
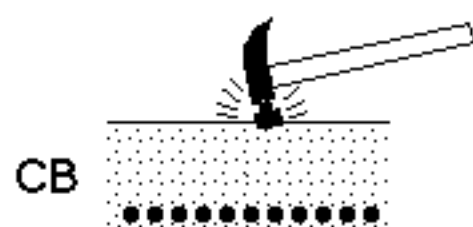
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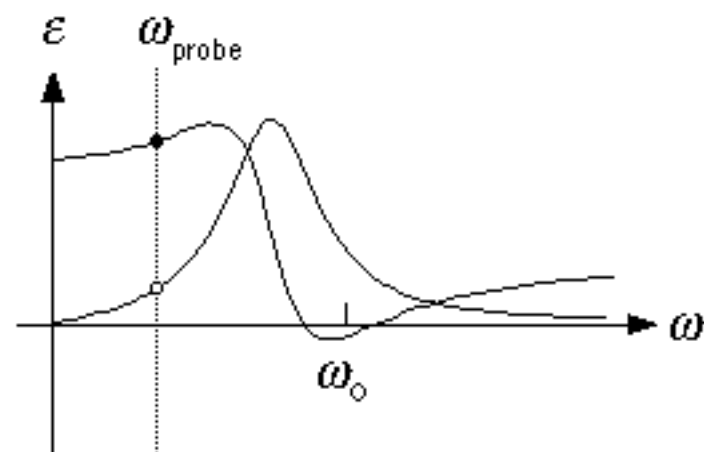
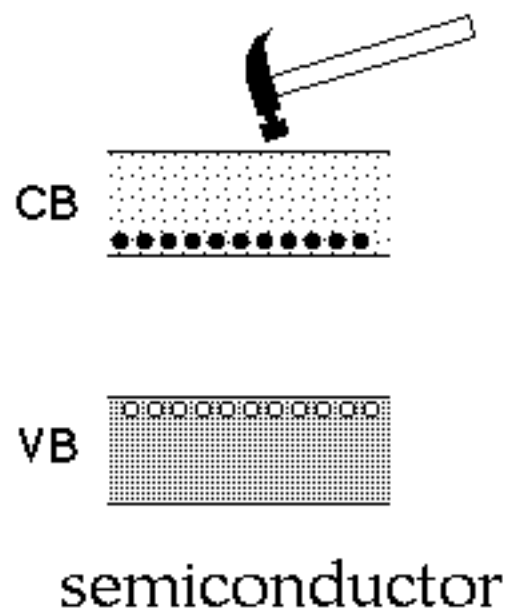


semiconductor

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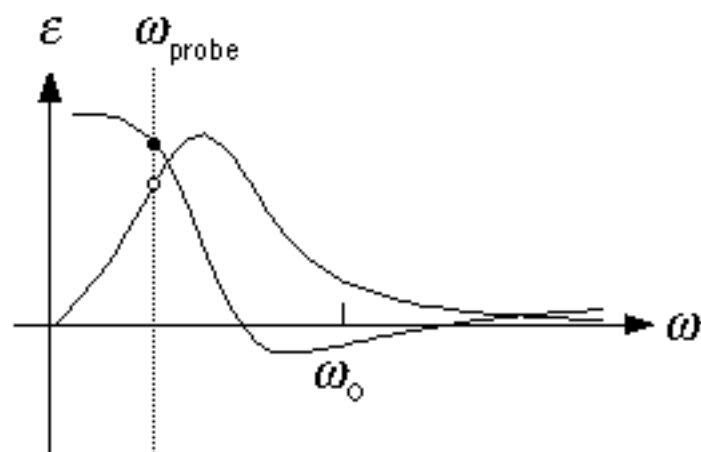
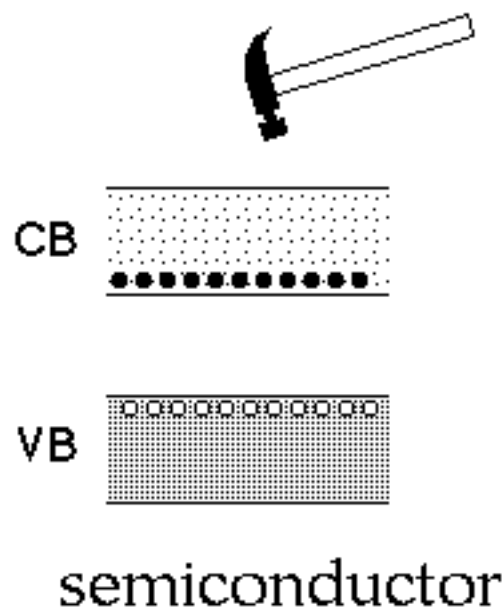
WHAT IS HAPPENING?



...and induces change
in bandstructure



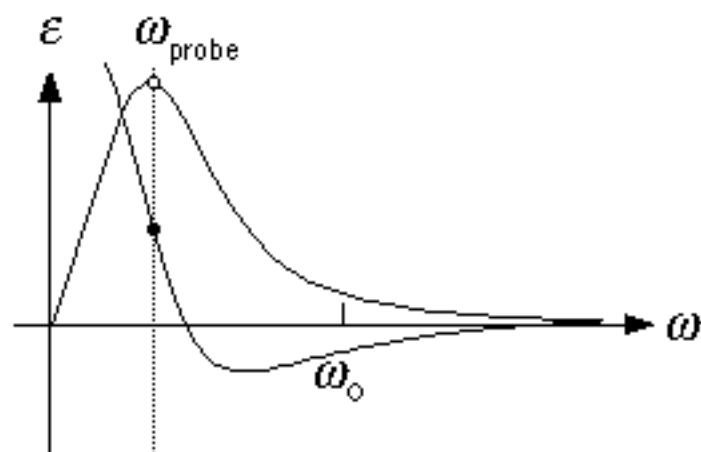
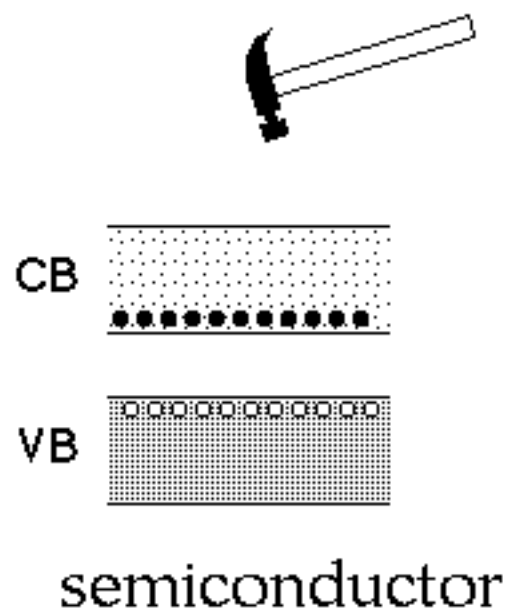
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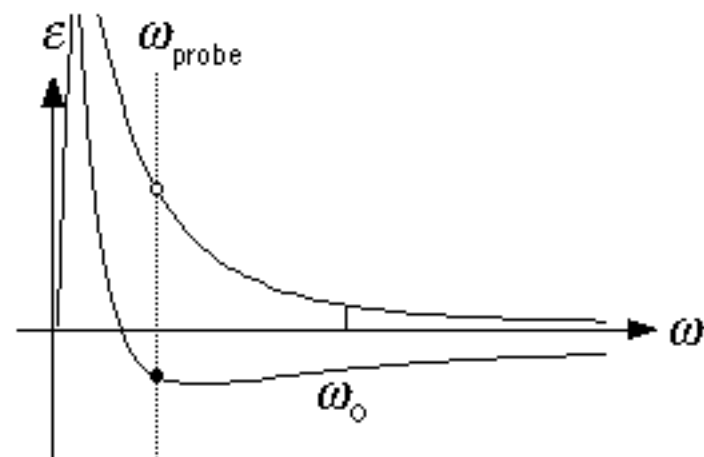
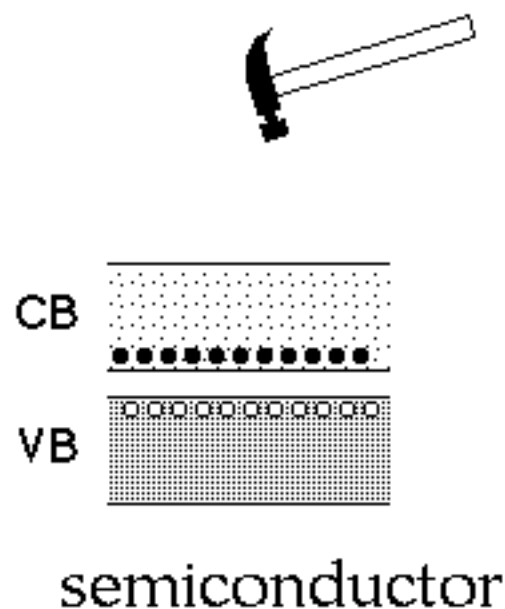
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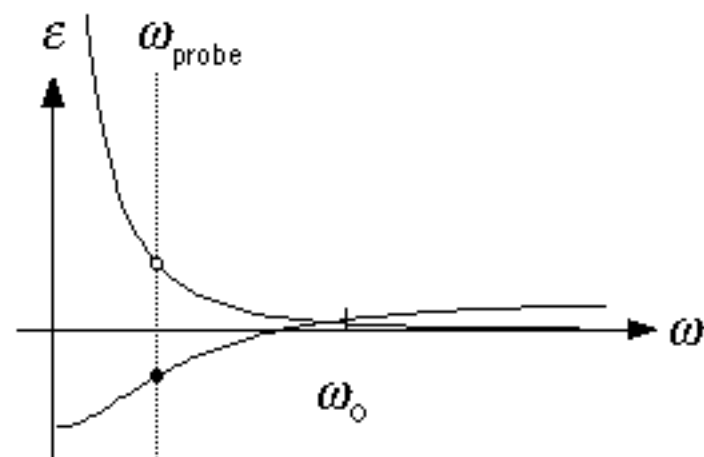
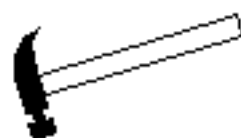
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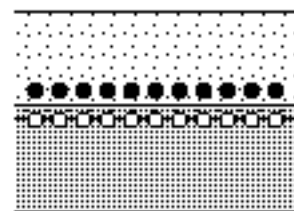


WHAT IS HAPPENING?



CB

VB

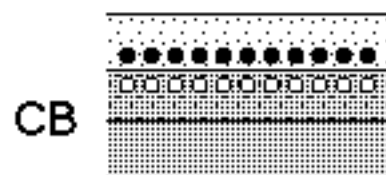
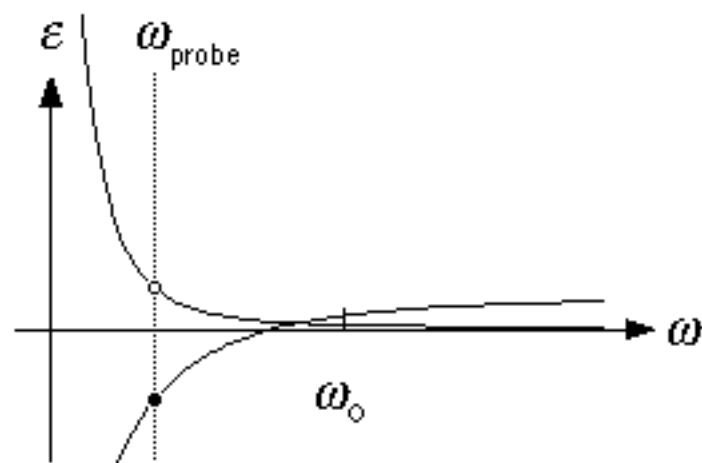
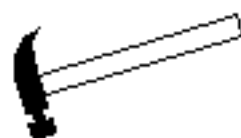


semimetal

...and induces change
in bandstructure



WHAT IS HAPPENING?

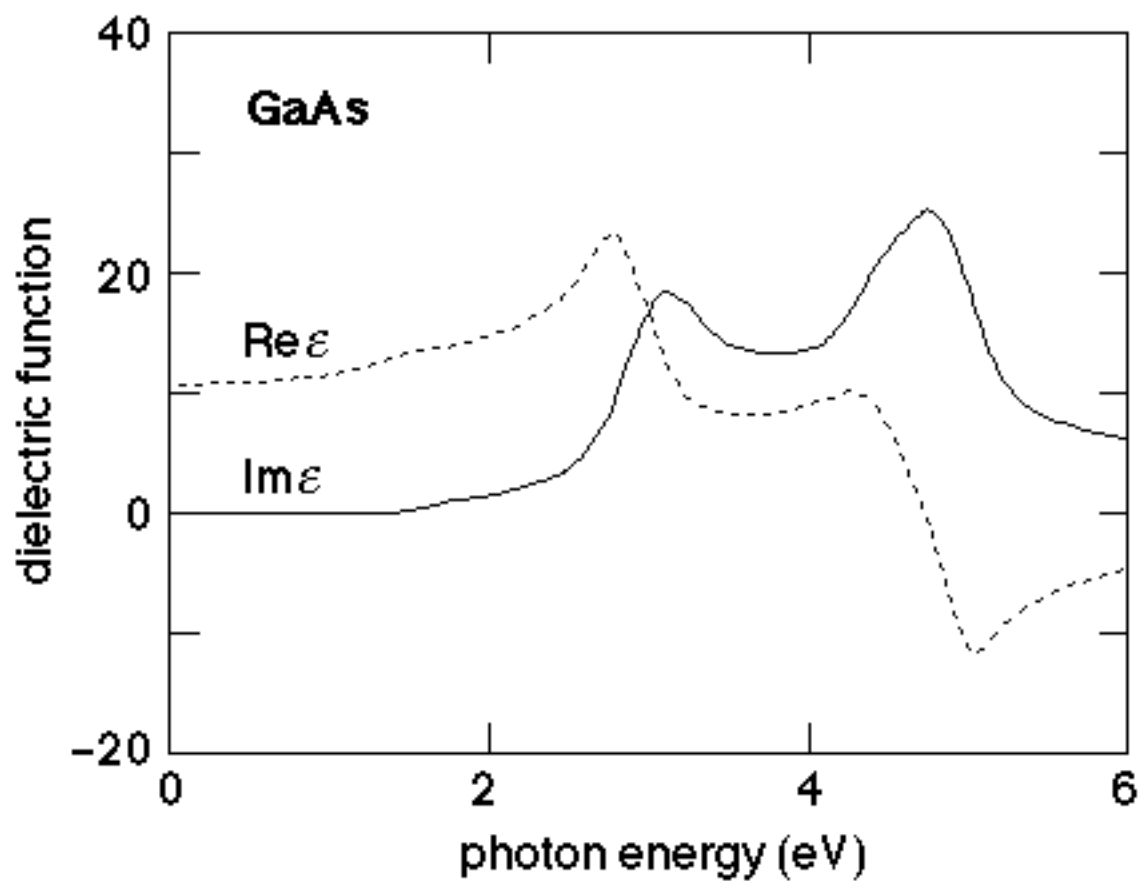


metal

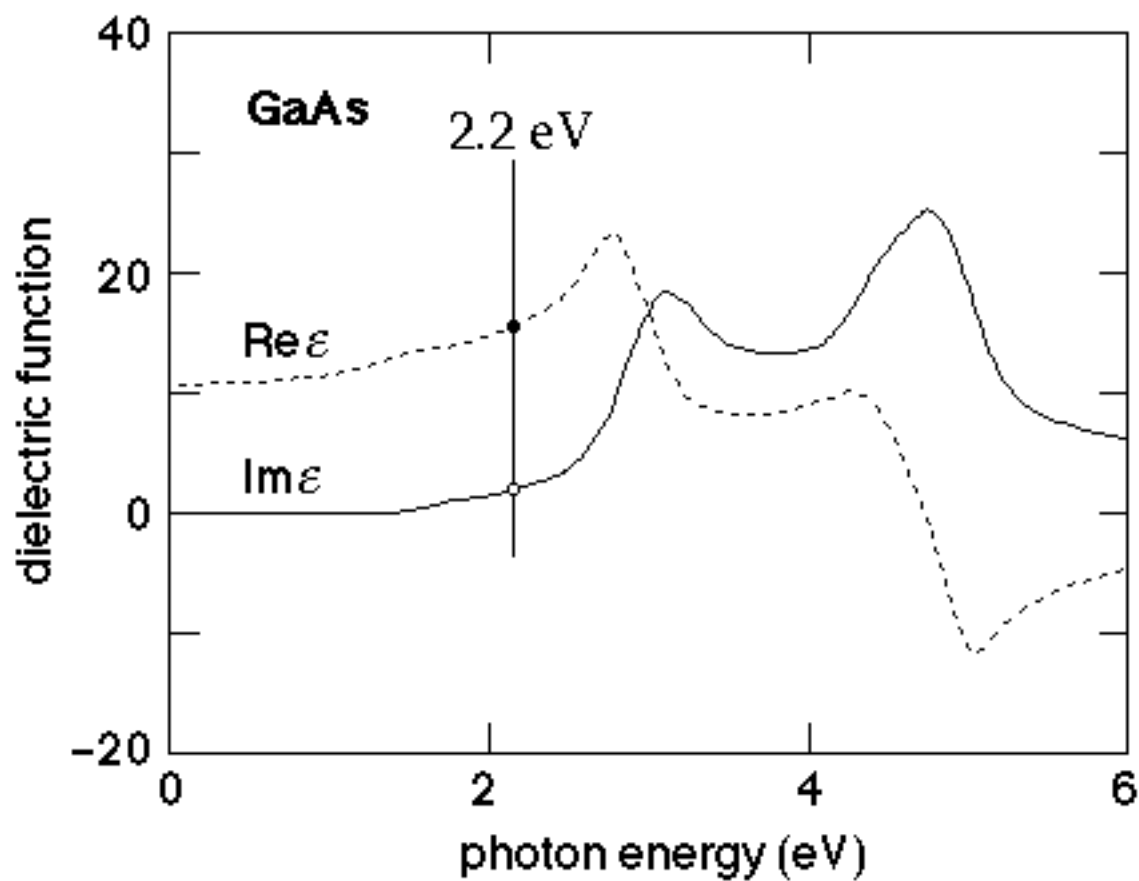
...and induces change
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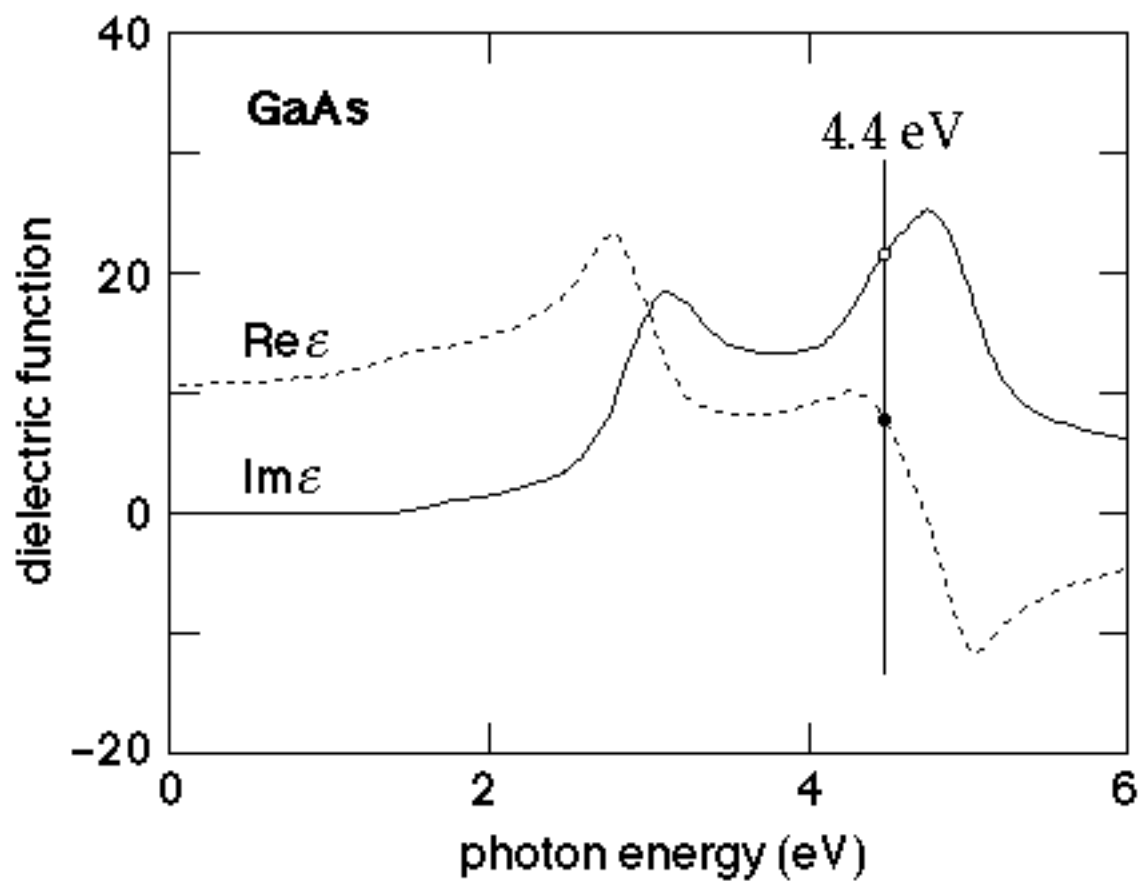
GaAs DIELECTRIC FUNCTION



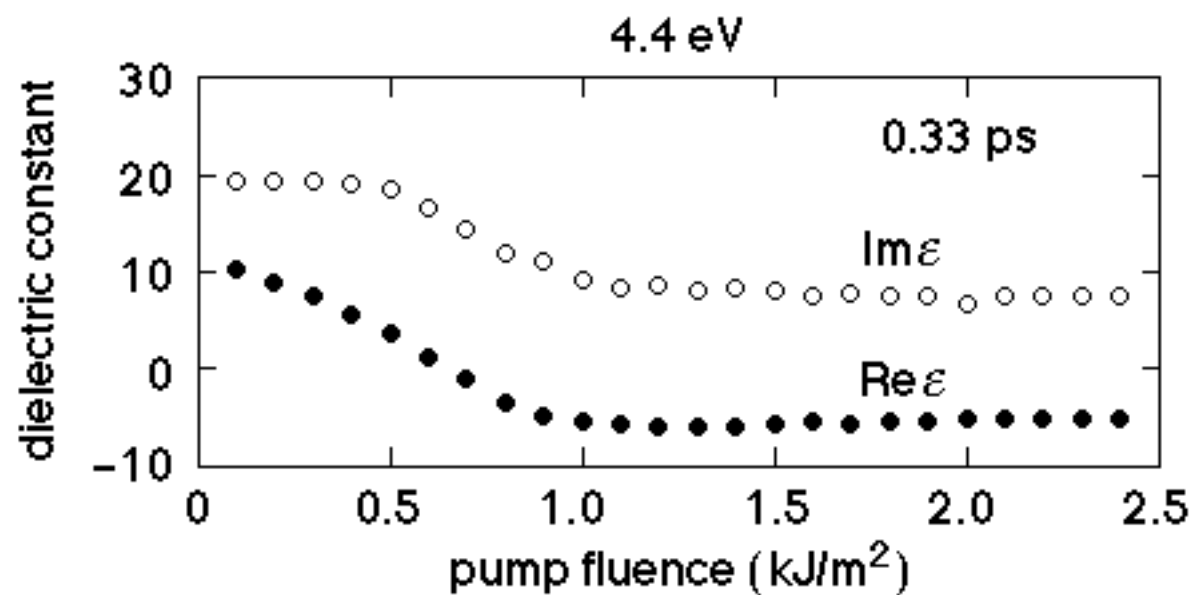
GaAs DIELECTRIC FUNCTION



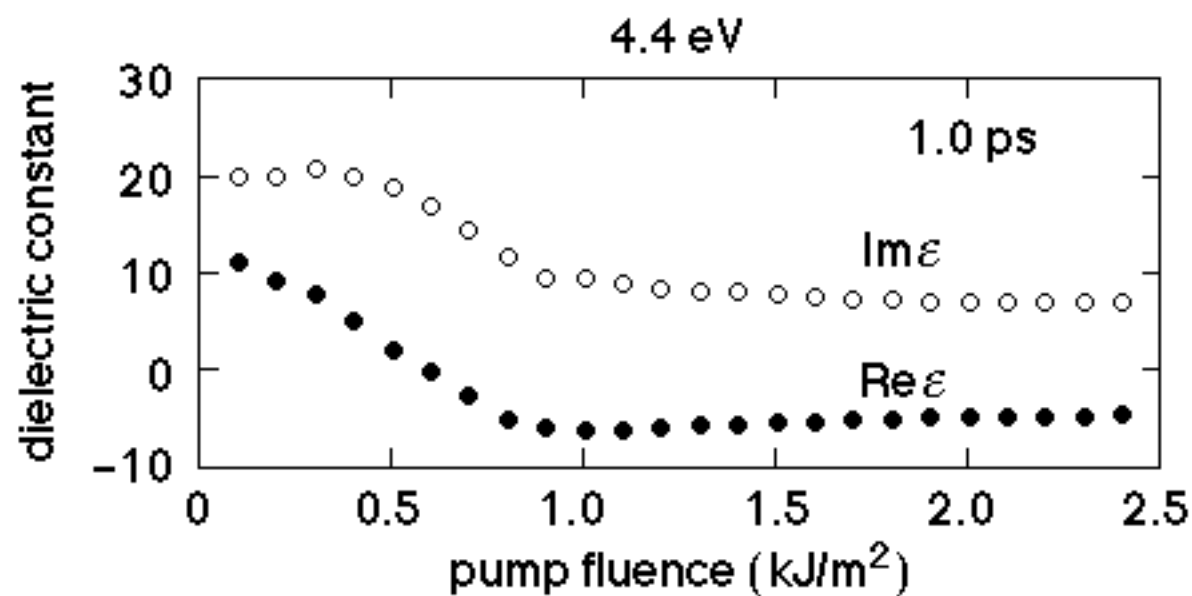
GaAs DIELECTRIC FUNCTION



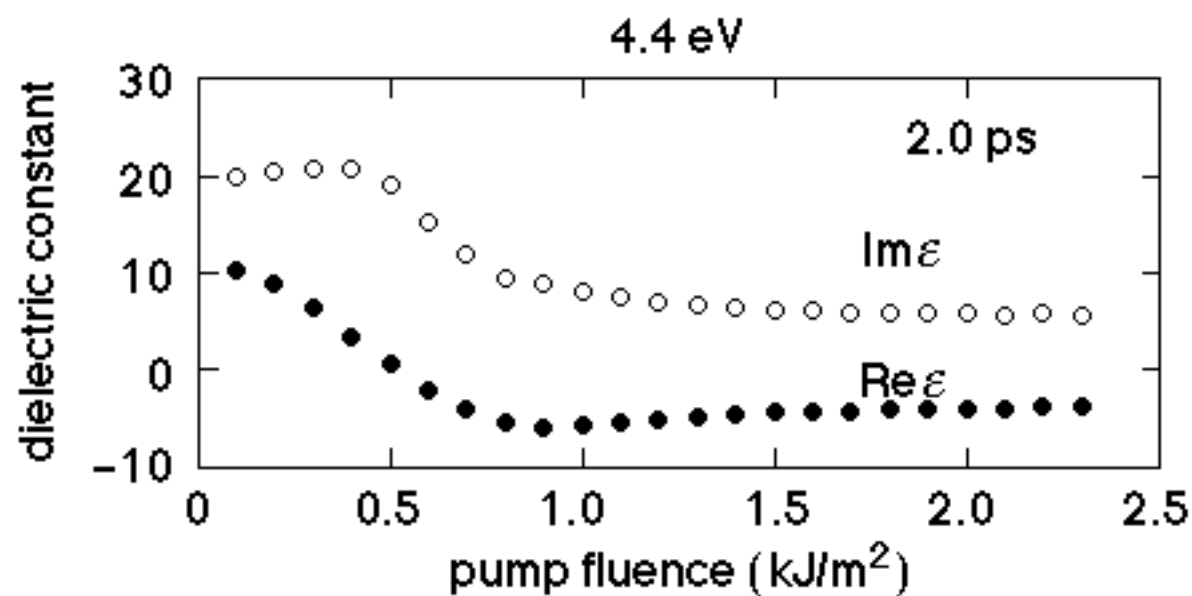
FLUENCE DEPENDENCE



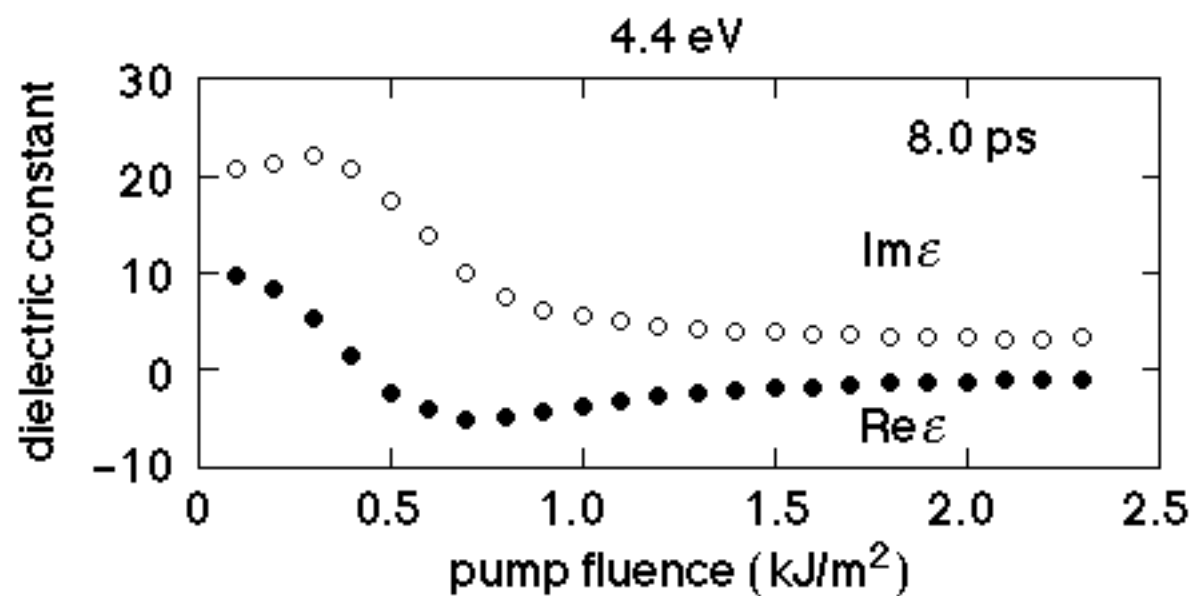
FLUENCE DEPENDENCE



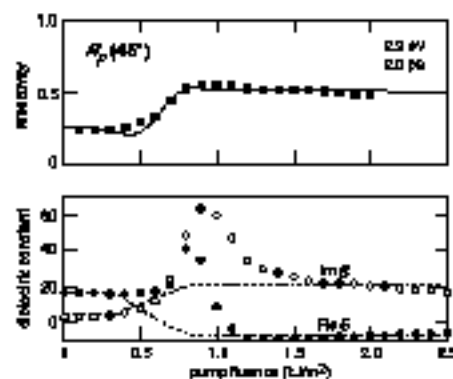
FLUENCE DEPENDENCE



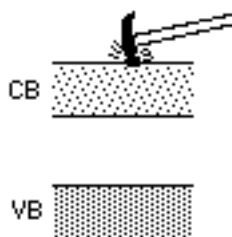
FLUENCE DEPENDENCE



SUMMARY



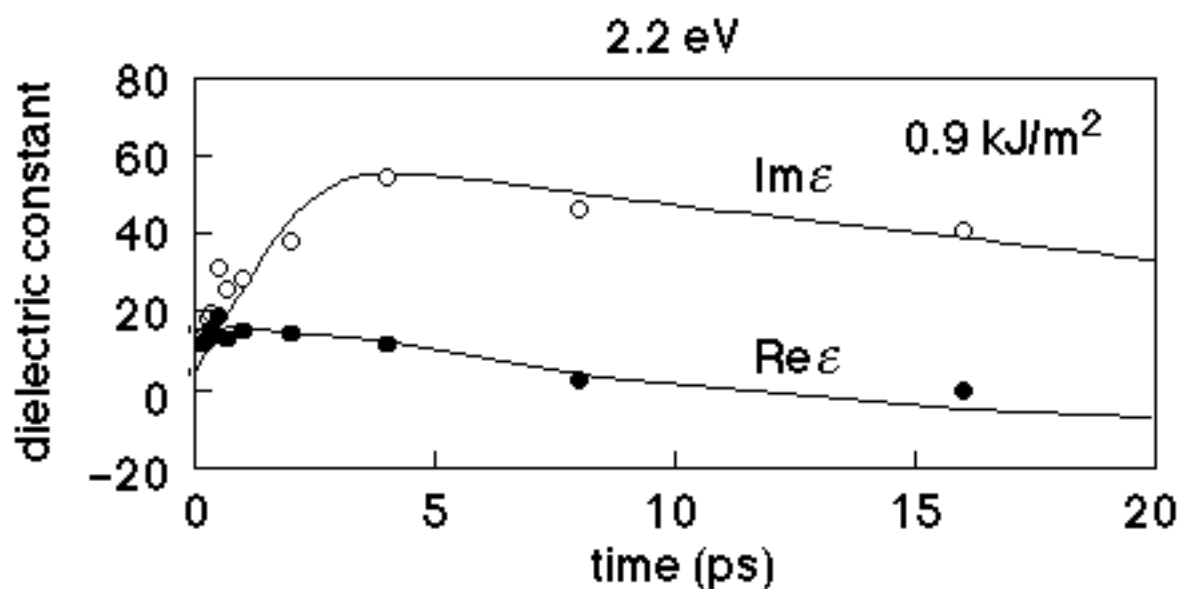
single-angle reflectivity not a good indicator of what happens



laser pulse causes bandgap collapse



CAUSE OF COLLAPSE



slow rise time → collapse cannot be caused by electronic effects alone

structural change?



NONLINEAR OPTICS

Field strength at second harmonic:

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



NONLINEAR OPTICS

Field strength at second harmonic:

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where:

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

so:



NONLINEAR OPTICS

Field strength at second harmonic:

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where:

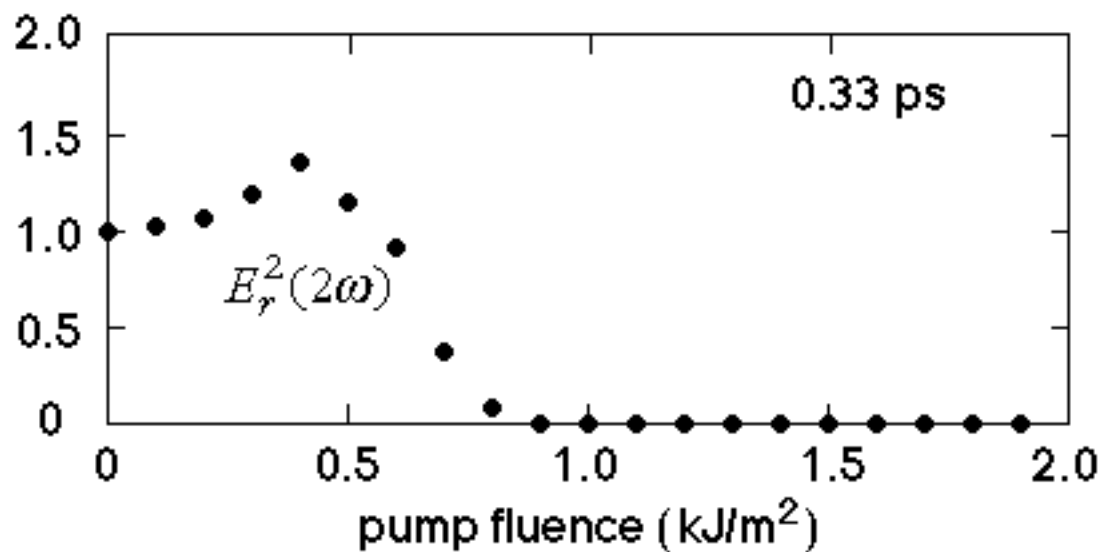
$$P^{(2)}(2\omega) = 2\chi^{(2)} E_i^2(\omega) f[\epsilon(\omega), \theta_i]$$

so:

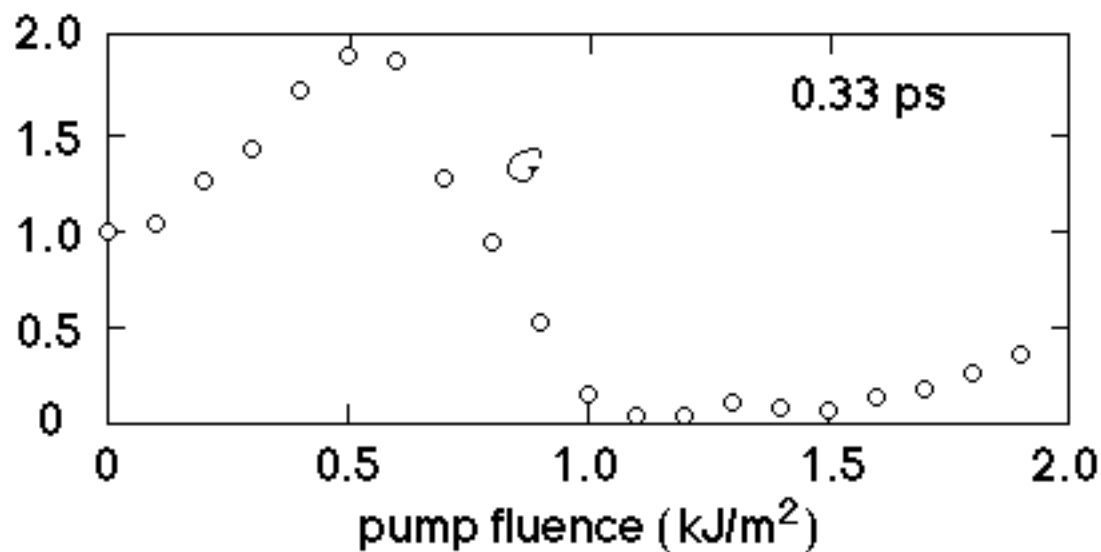
$$|\chi^{(2)}|^2 = \frac{E_r^2(2\omega)}{E_i^2(\omega) G[\epsilon(\omega), \epsilon(2\omega), \theta_i]}$$



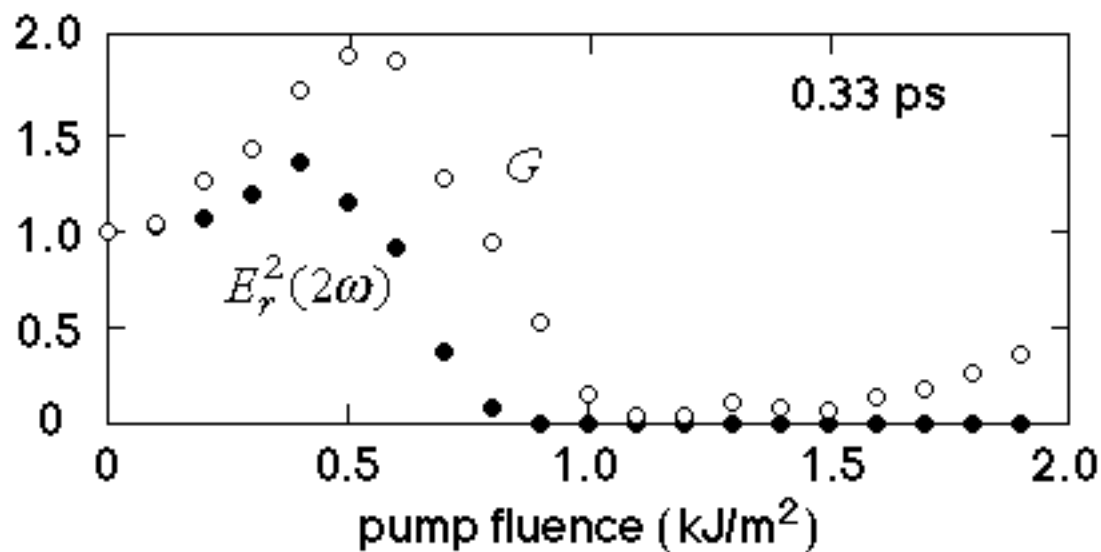
SECOND HARMONIC DATA



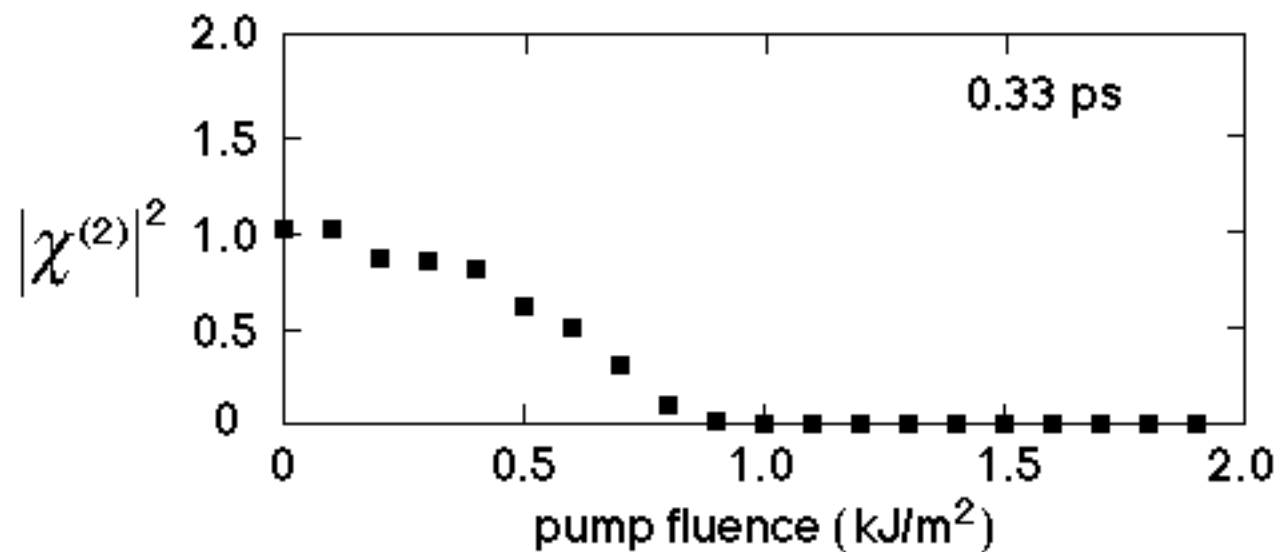
SECOND HARMONIC DATA



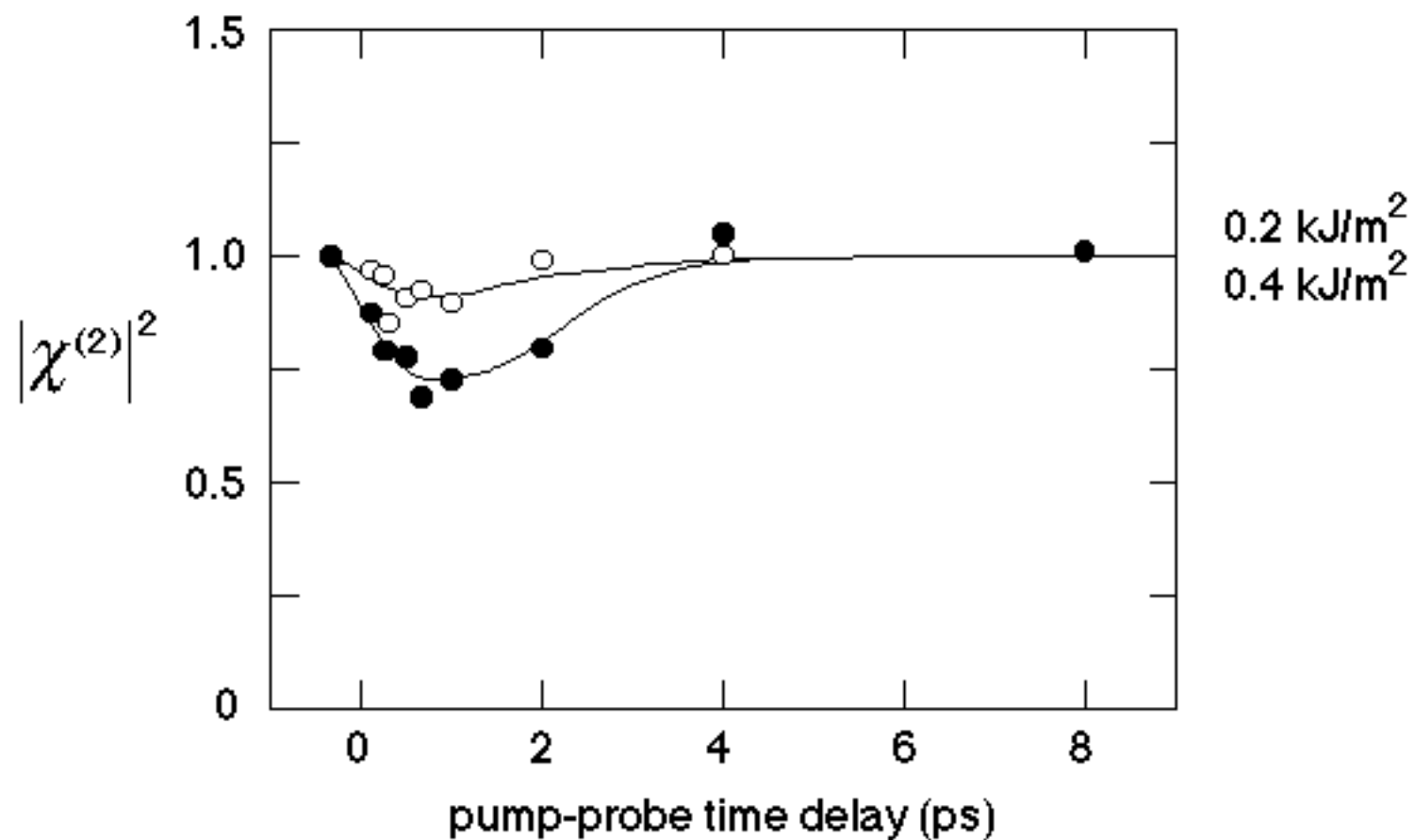
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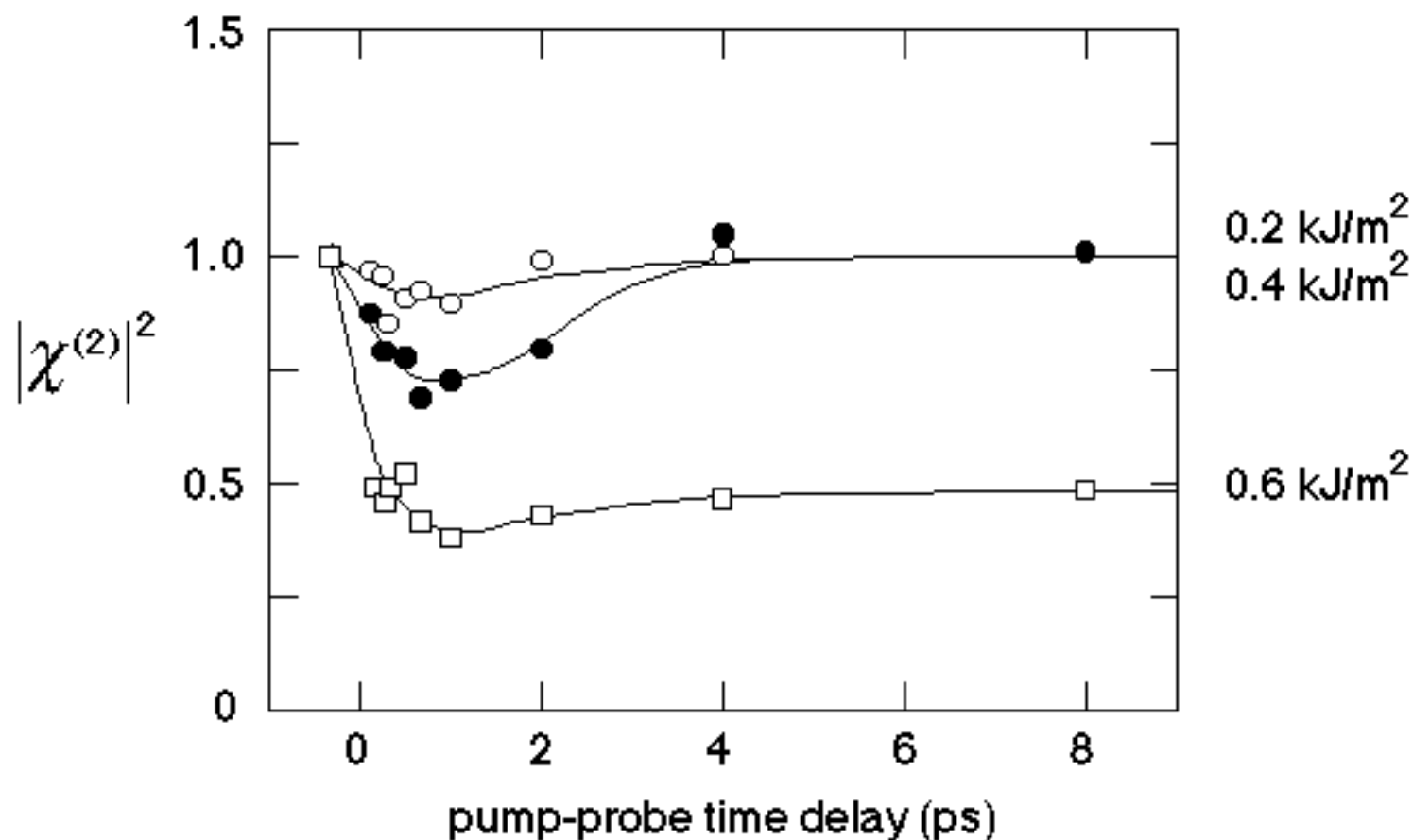
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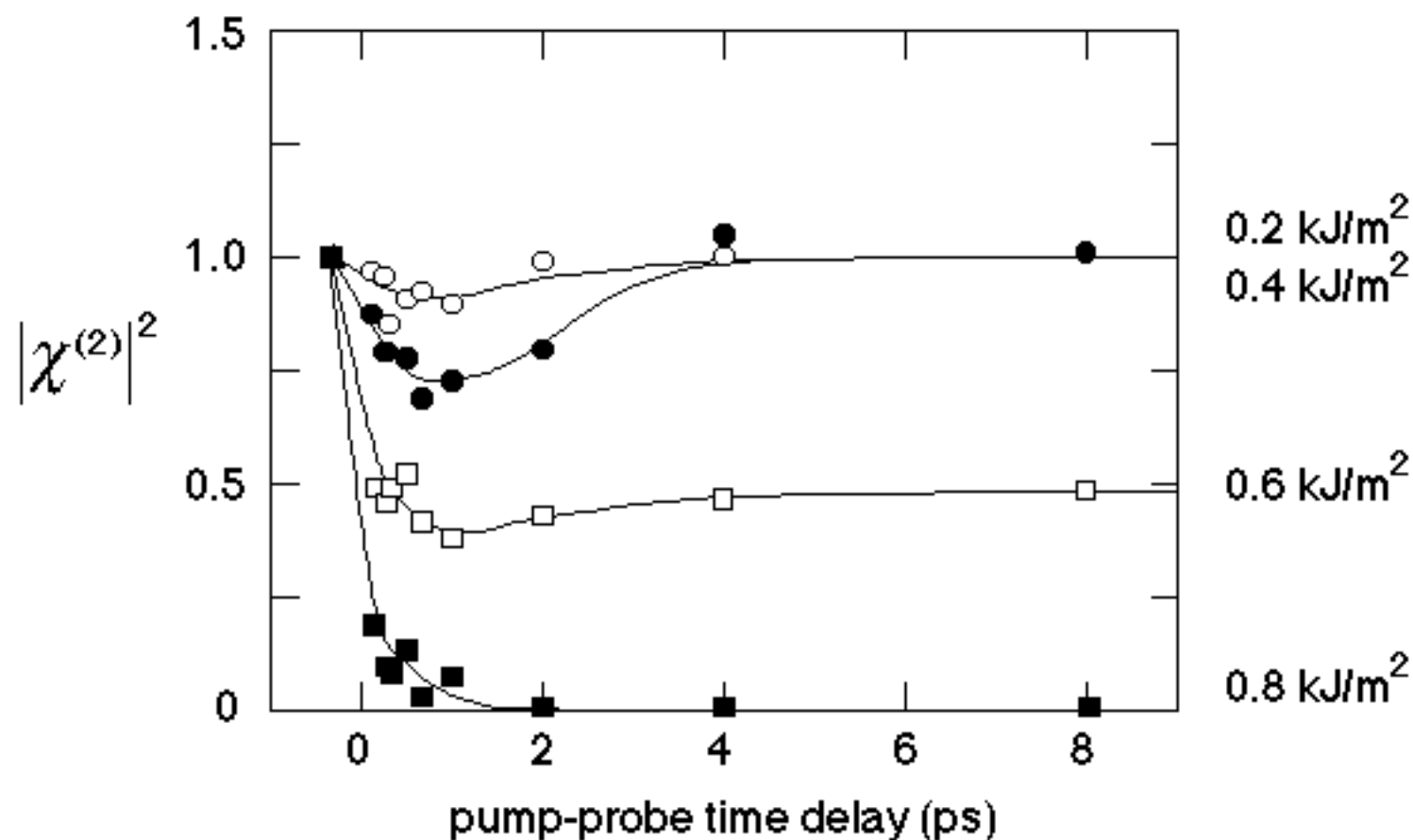
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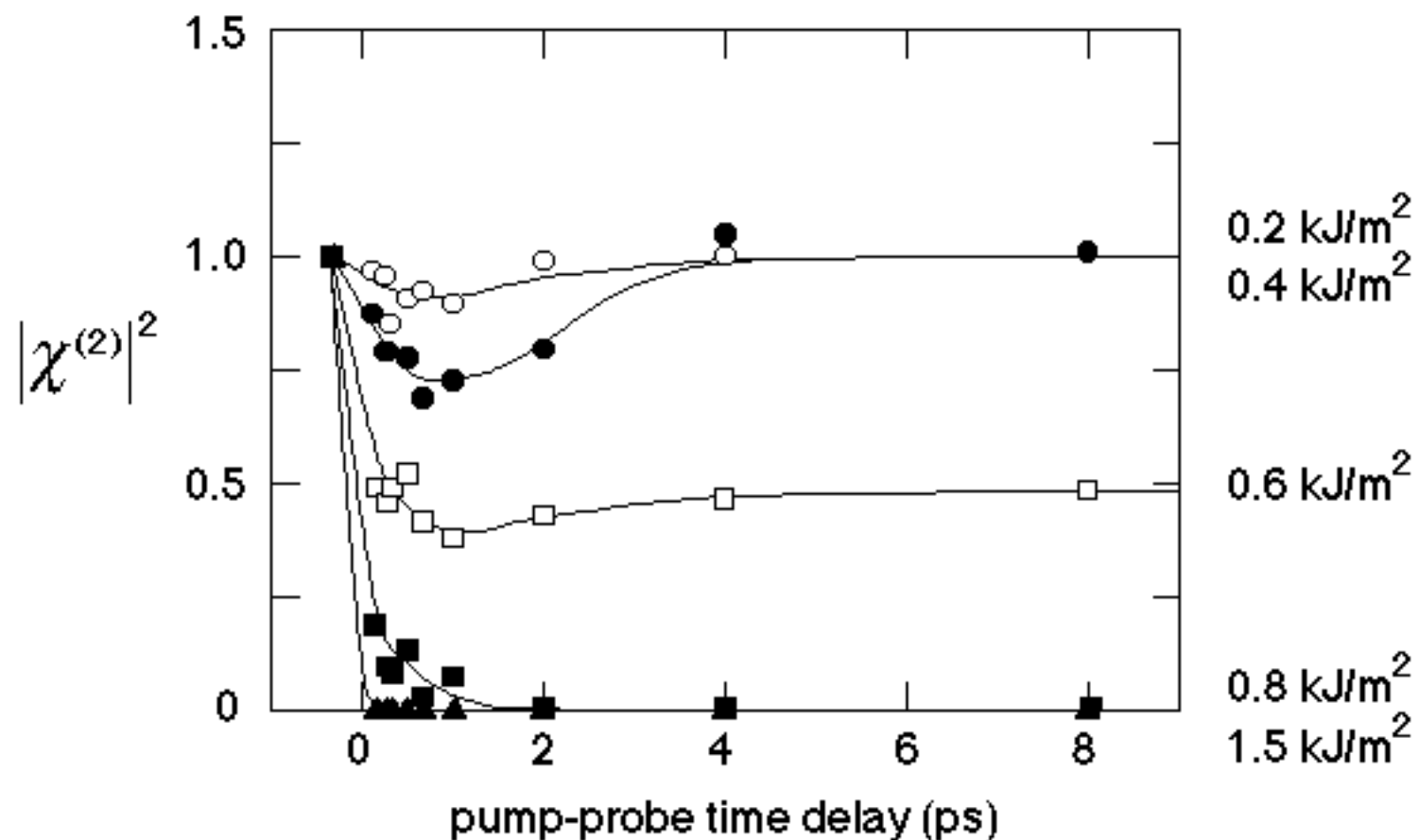
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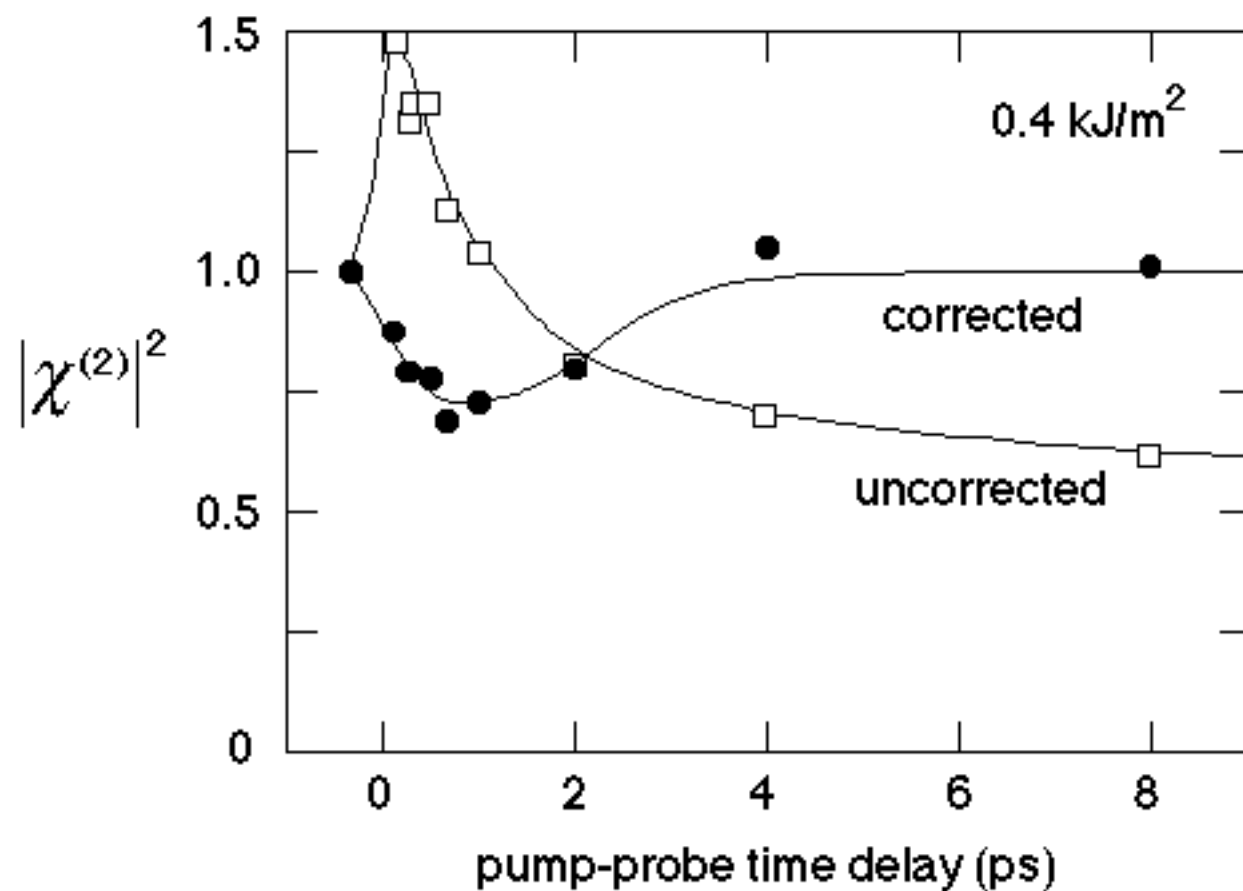
SECOND HARMONIC DATA



SECOND HARMONIC DATA



EFFECT OF CHANGES IN ϵ

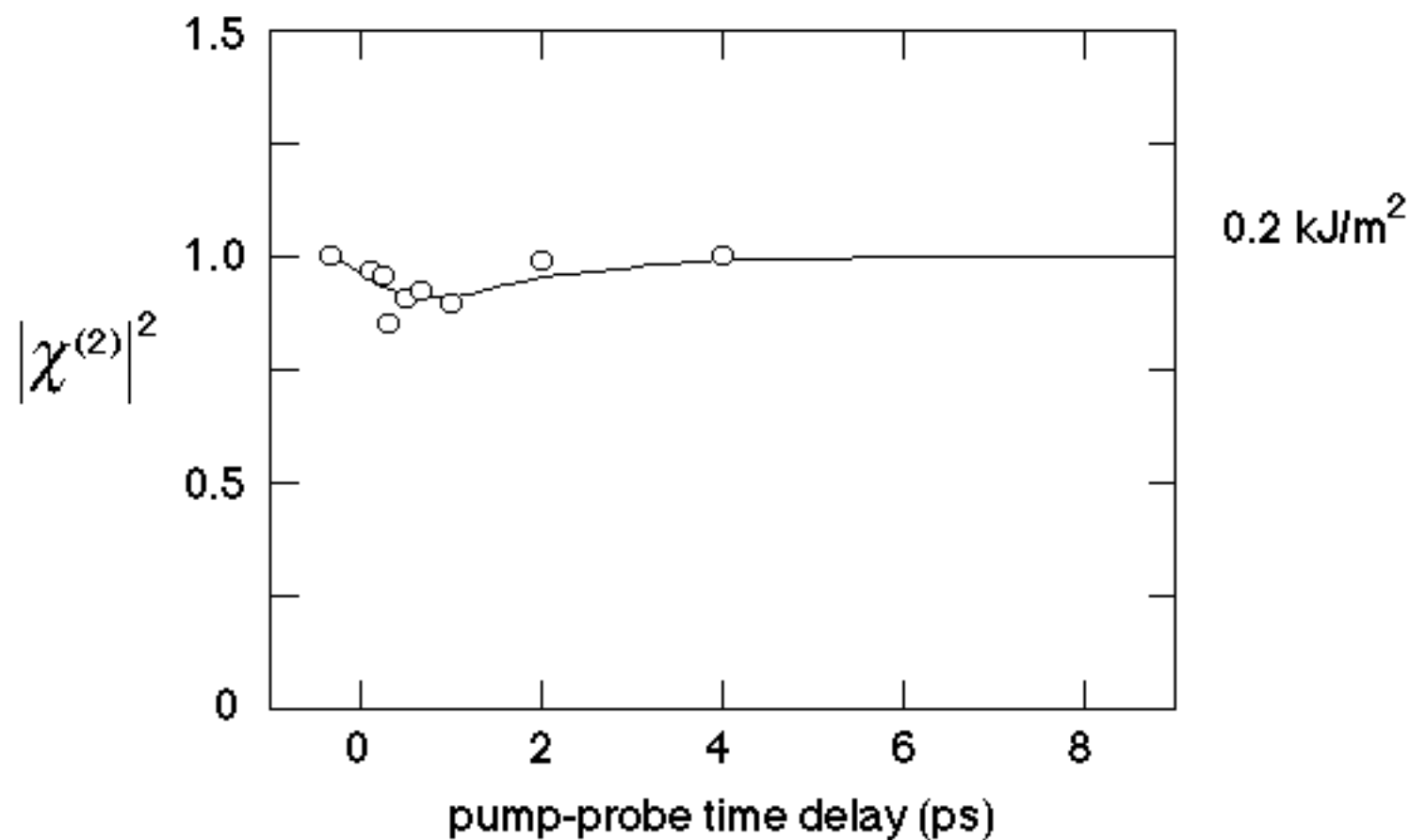


FEMTOSECOND WORK

- $\chi^{(2)}$ sensitive to loss of long-range order
- long-range order disappears even below threshold
- structural change due to destabilization of covalent bonds
- time-scale for structural change reasonable (10% of bond length in 1 ps requires 25 m/s)



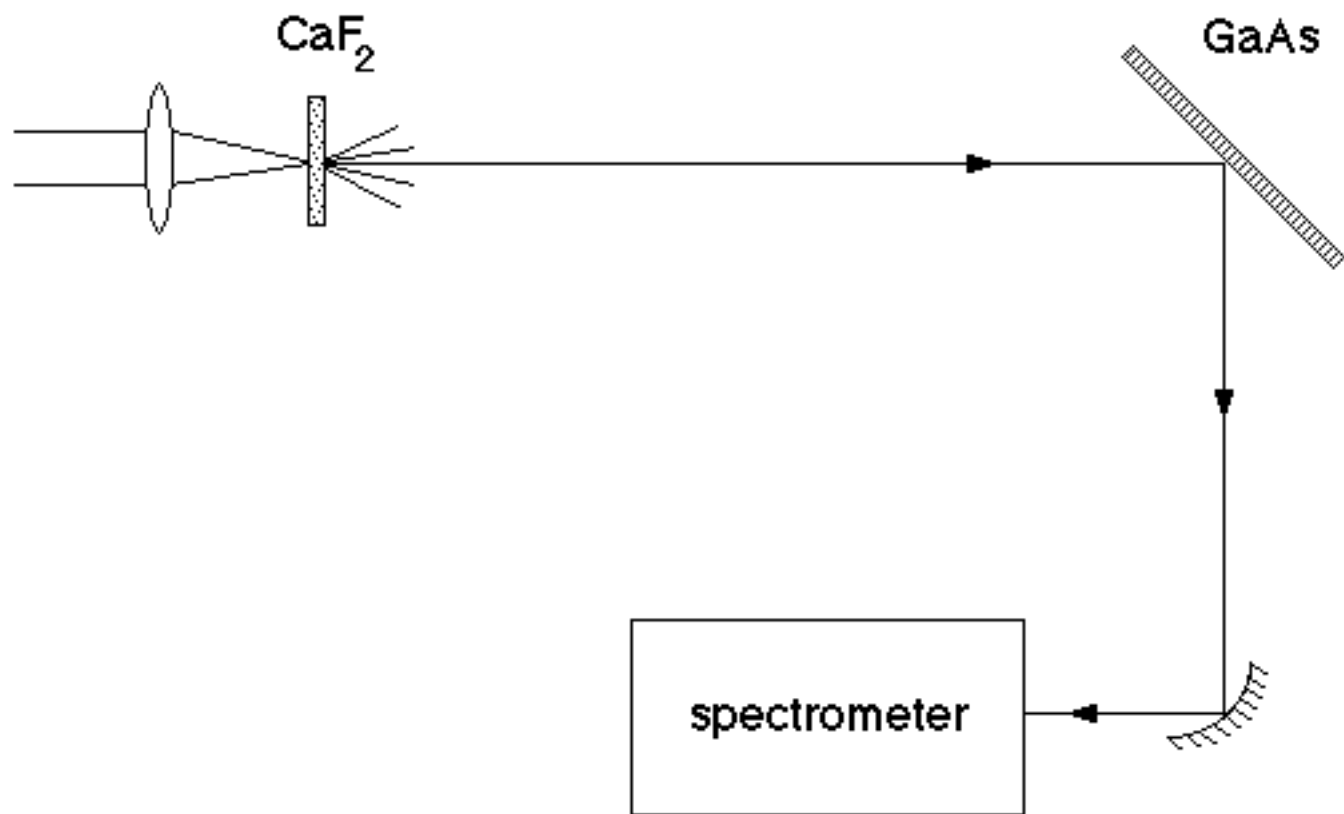
SECOND HARMONIC DATA



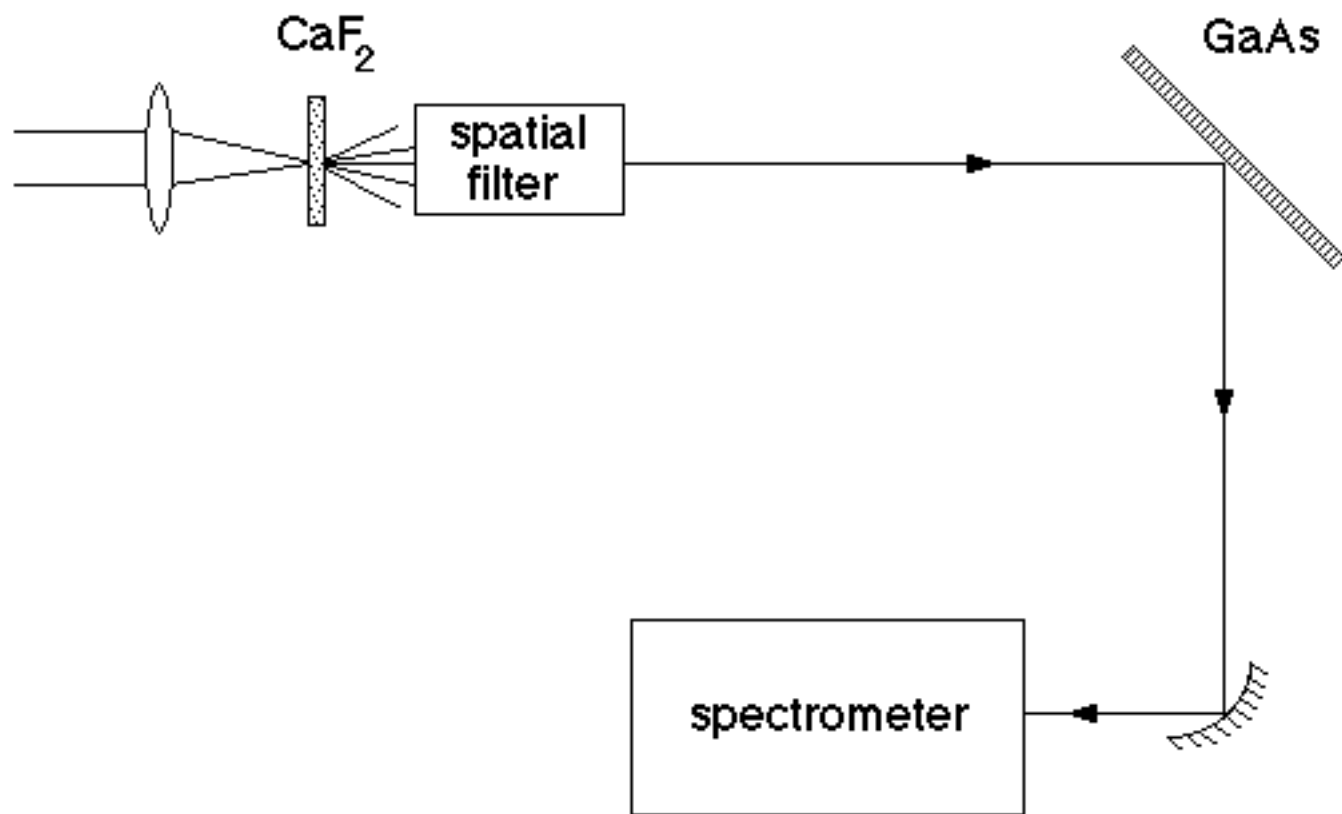
- ① Background
- ② Single frequency measurements
- ③ Detailed Picture



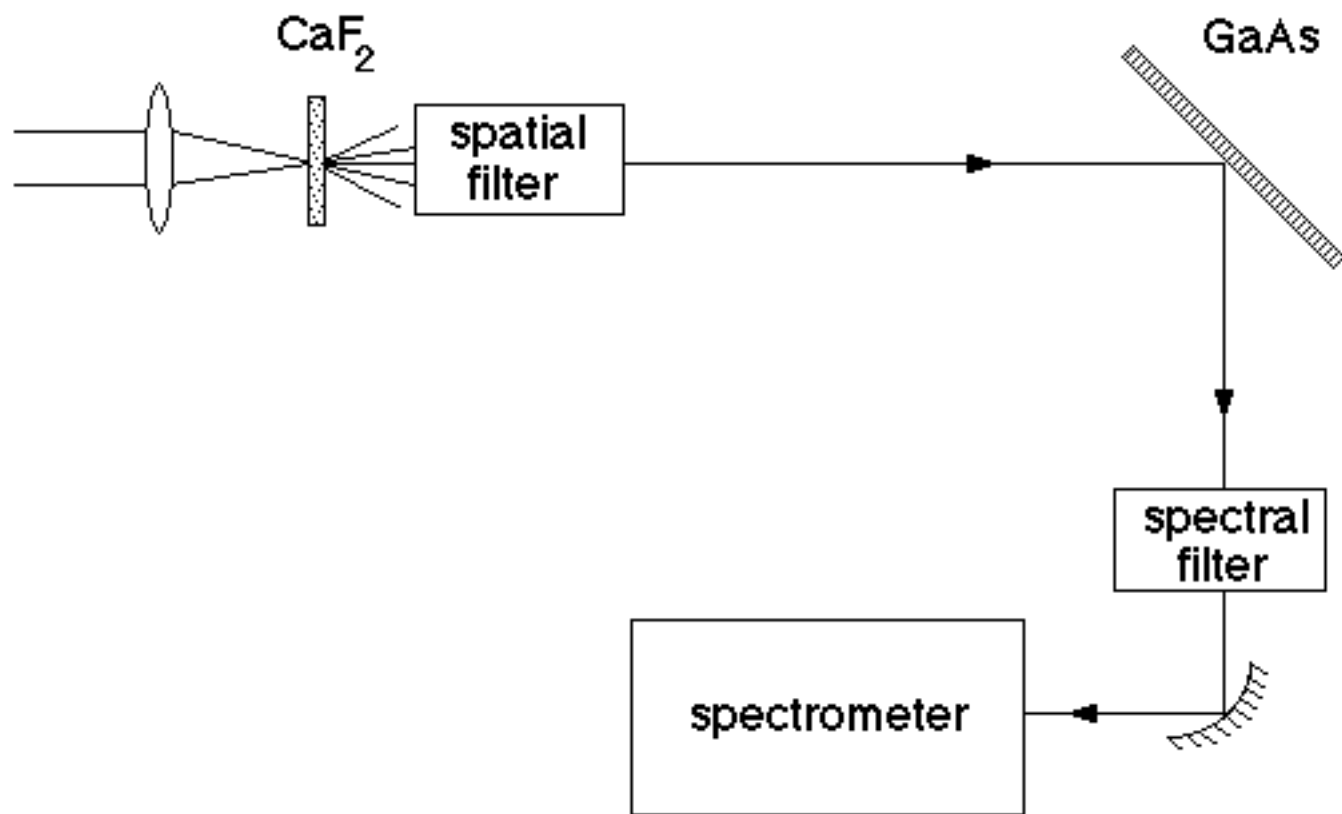
BROADBAND PROBE



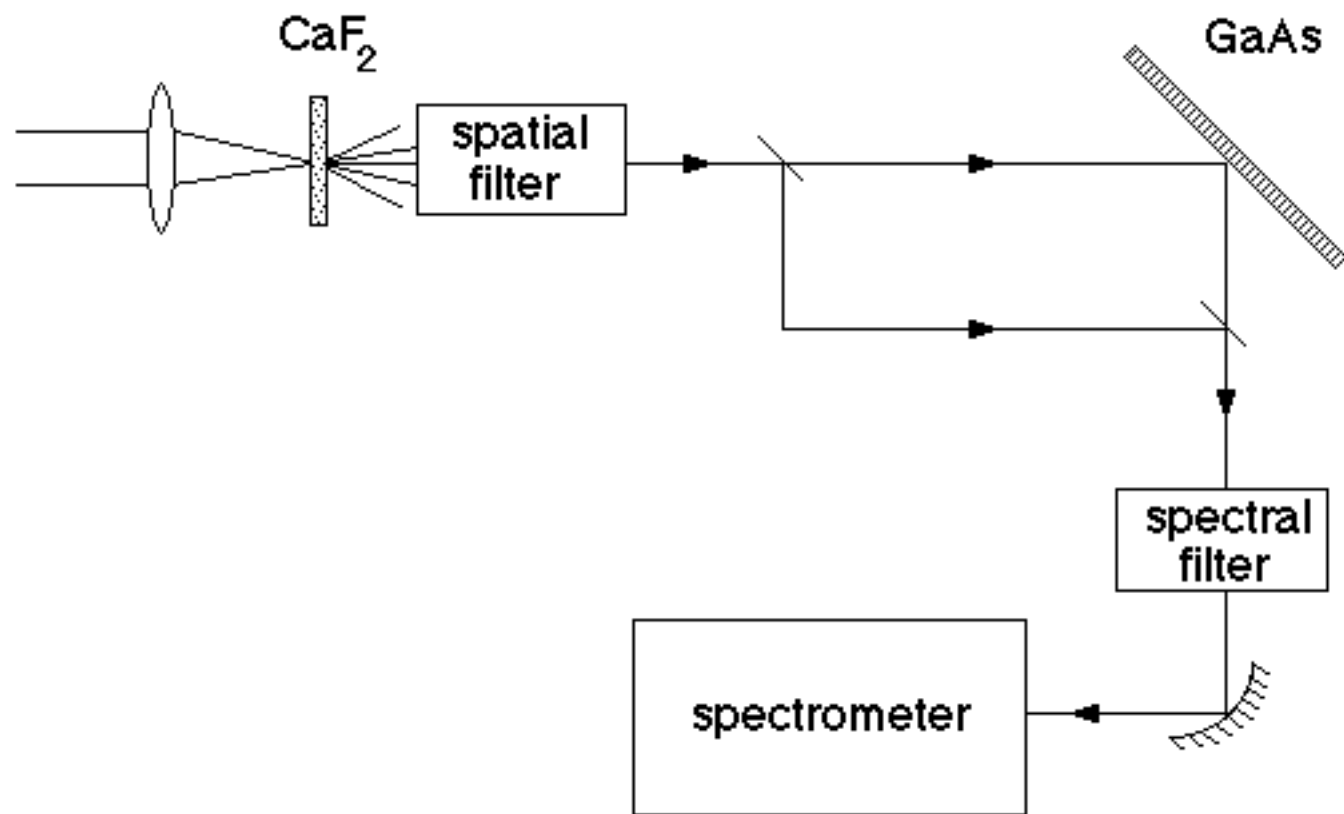
BROADBAND PROBE



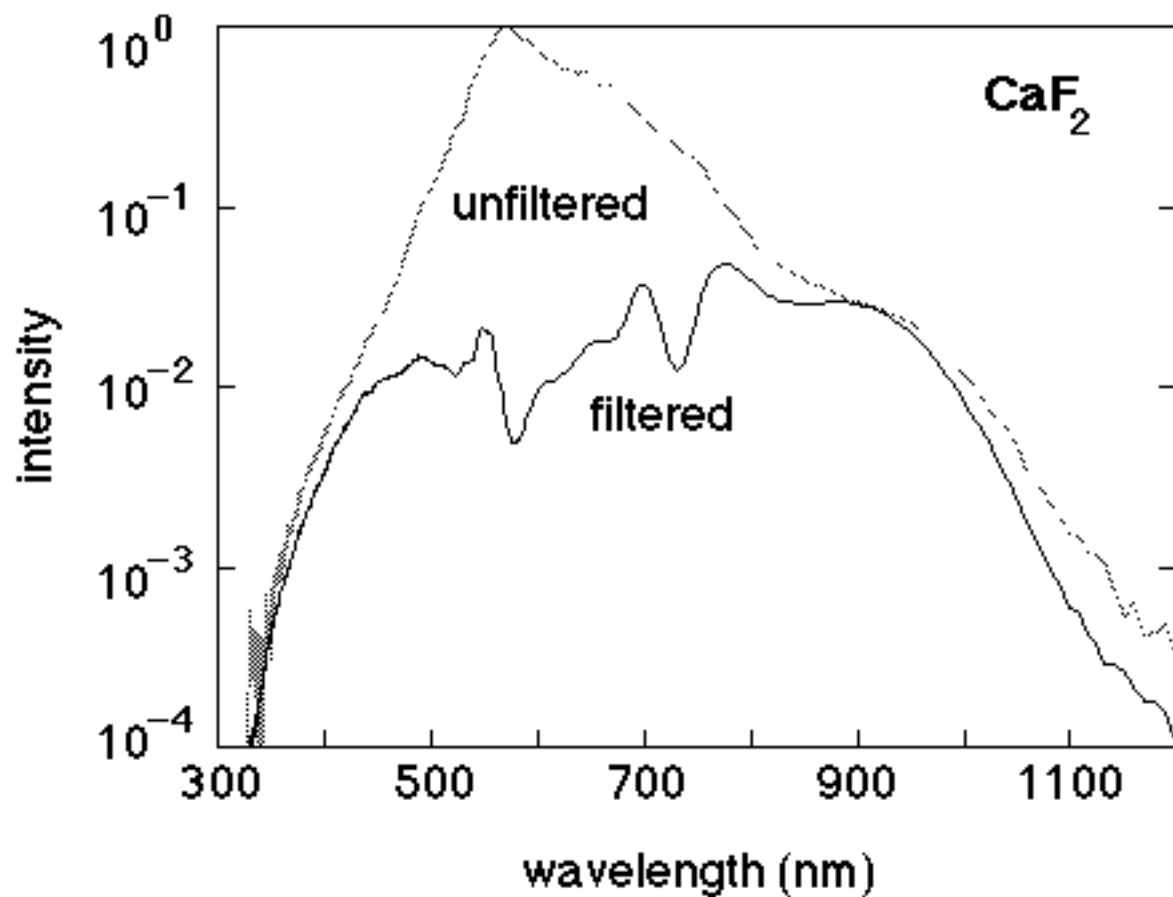
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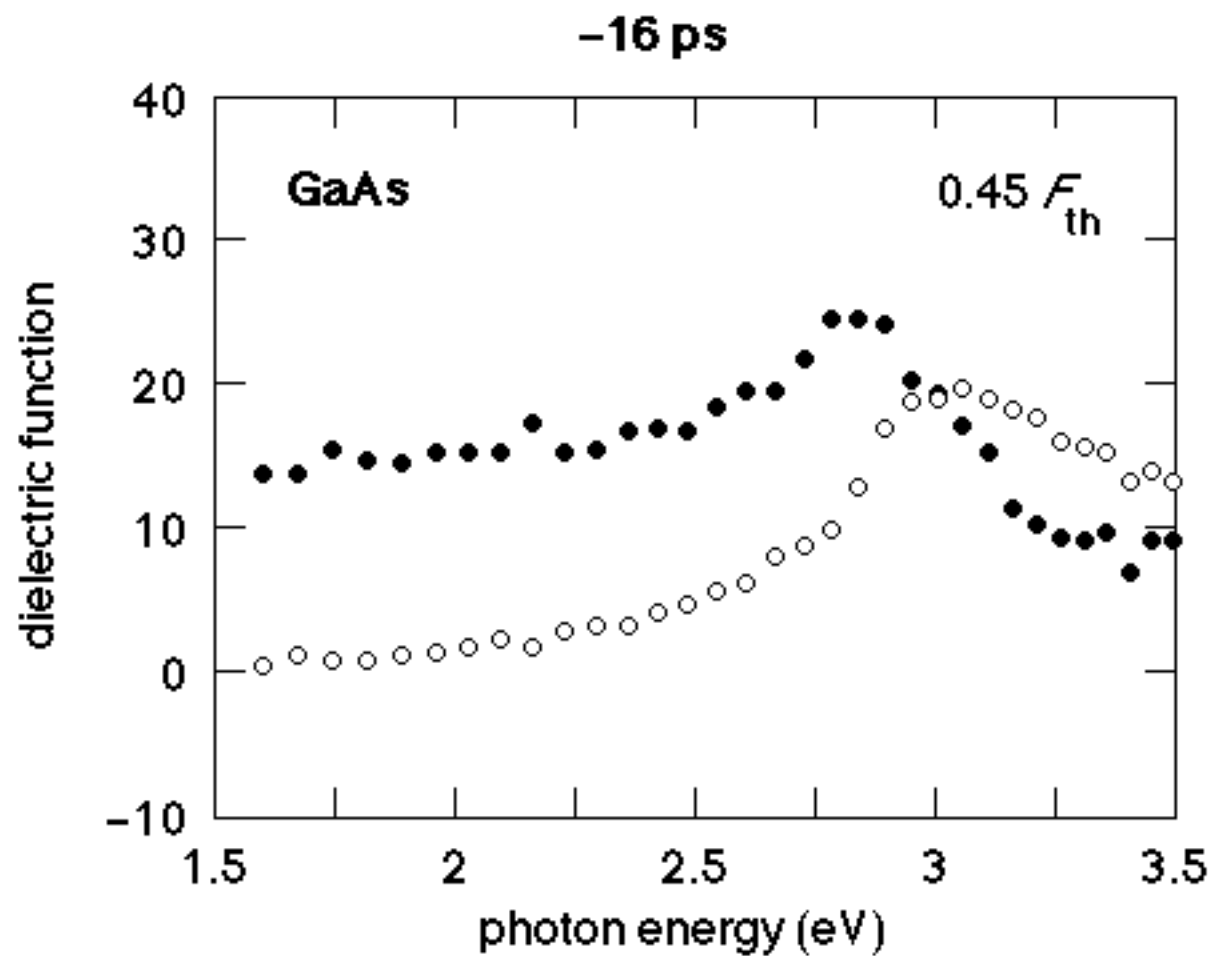
BROADBAND PROBE



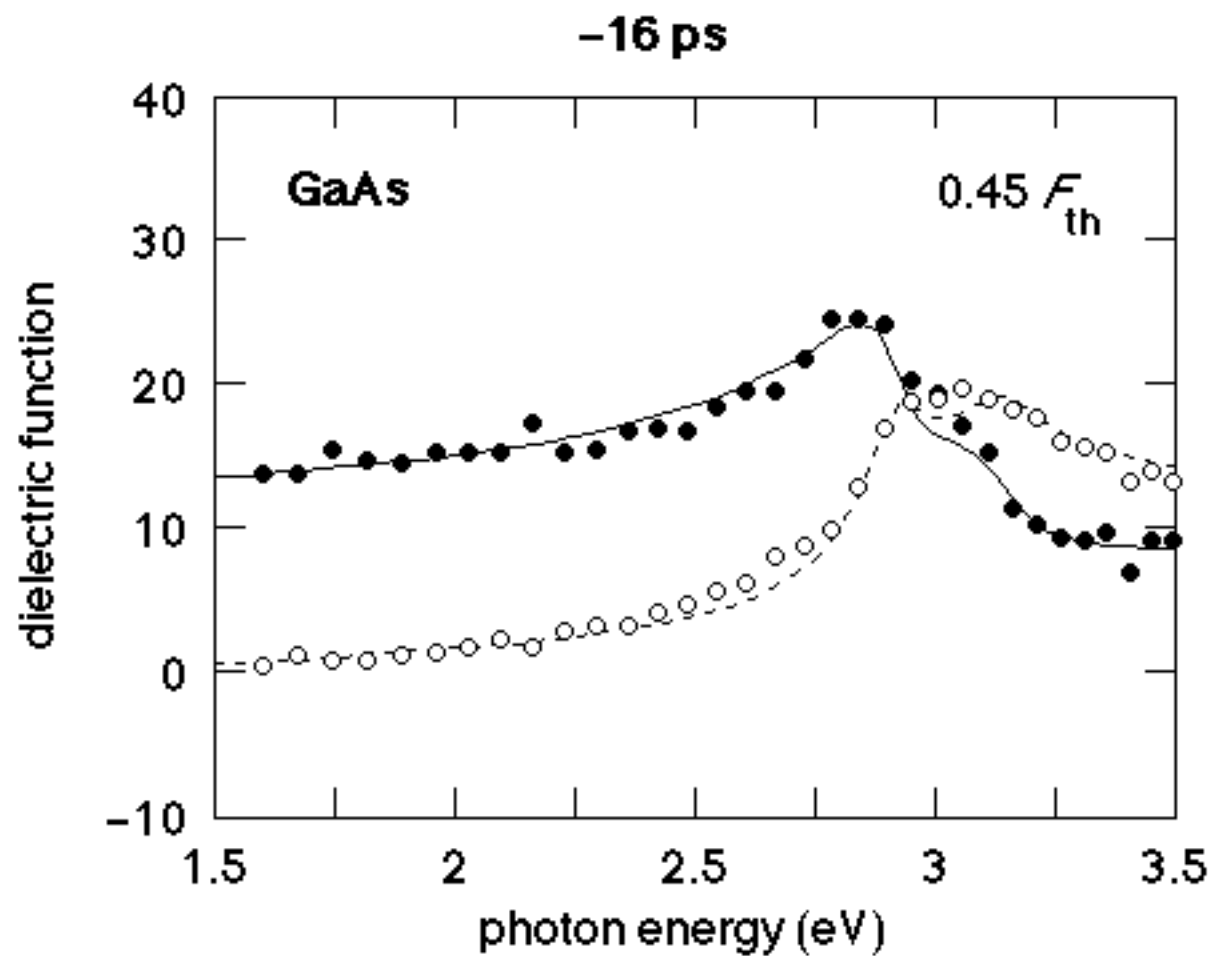
WHITE LIGHT SPECTRUM



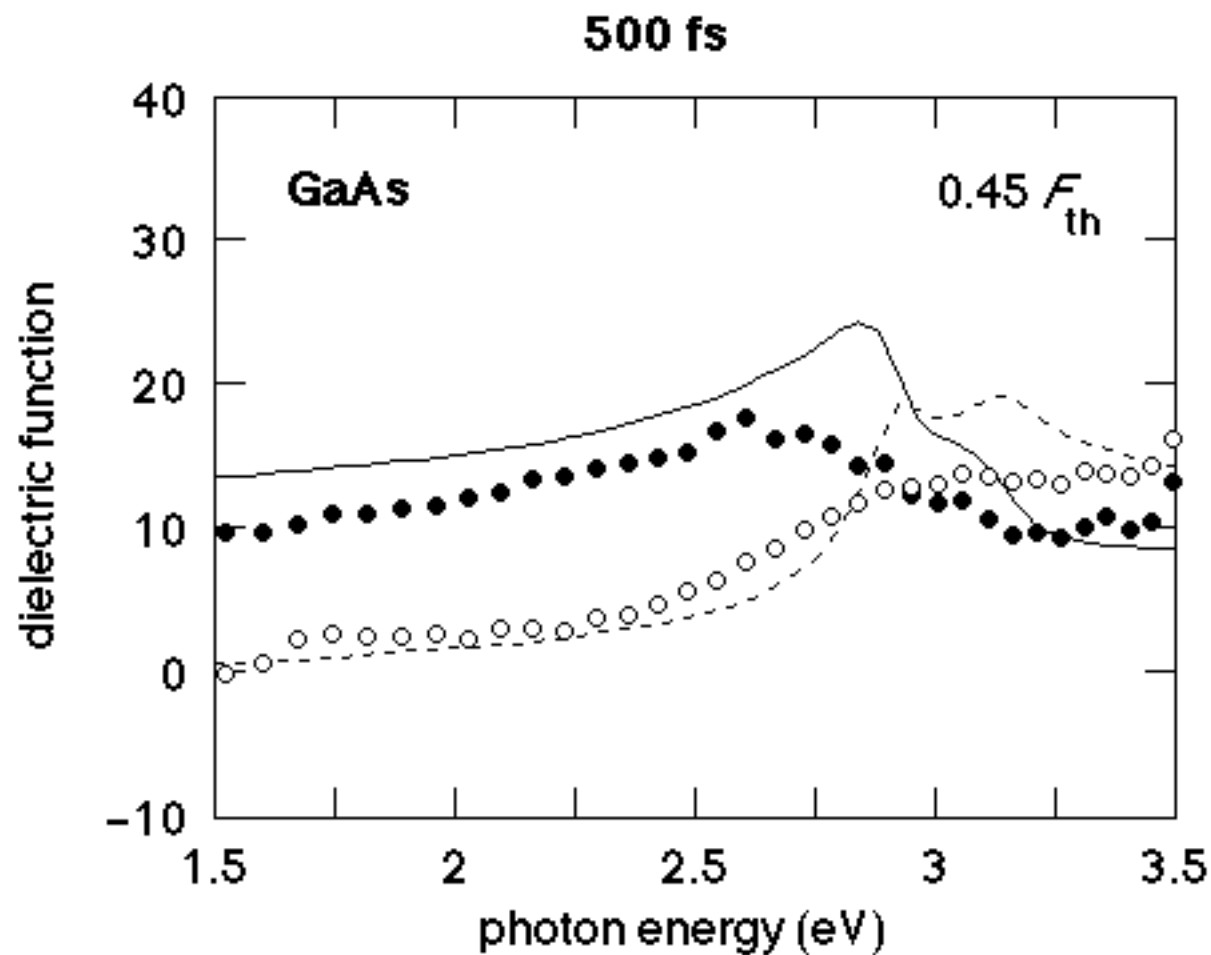
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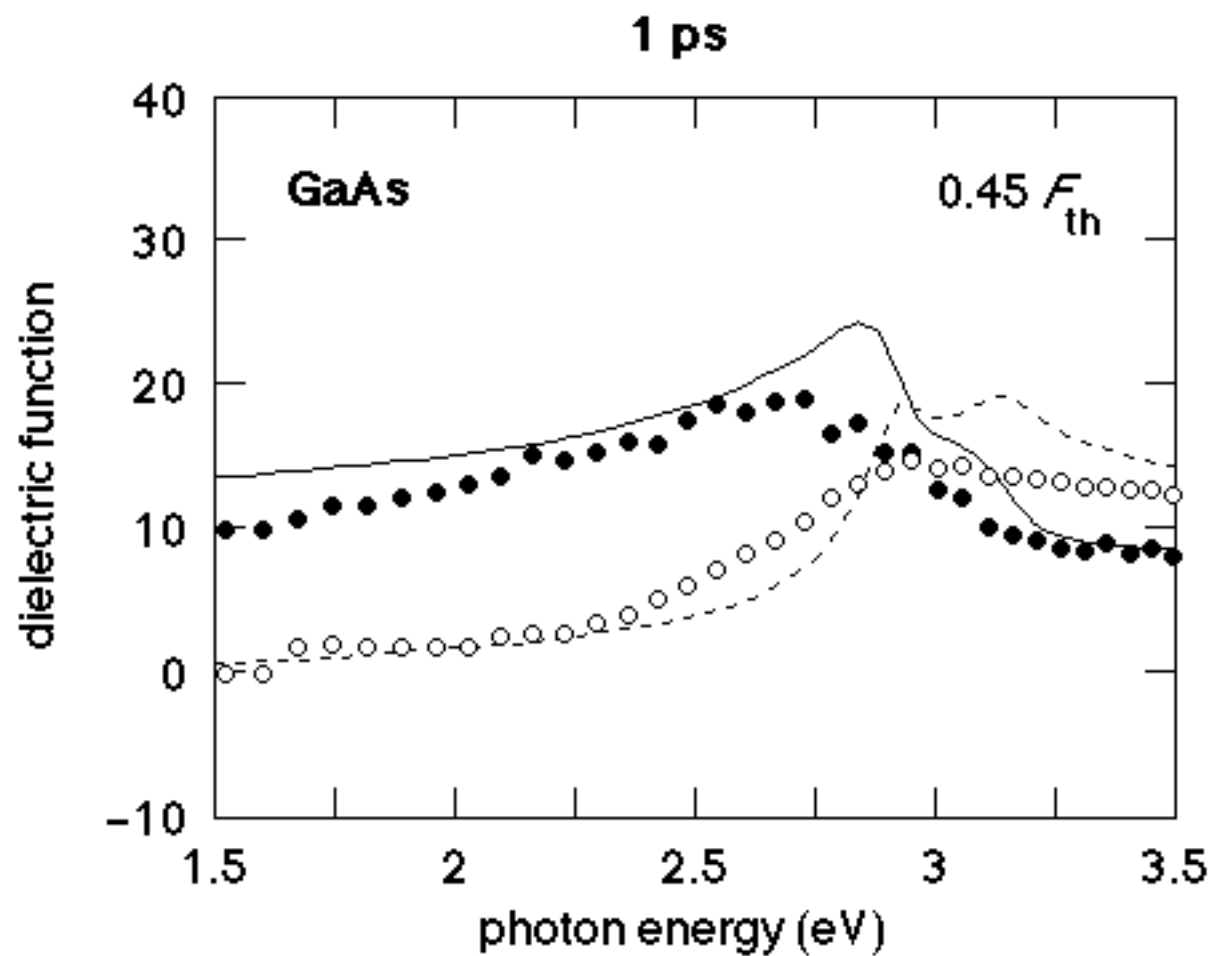
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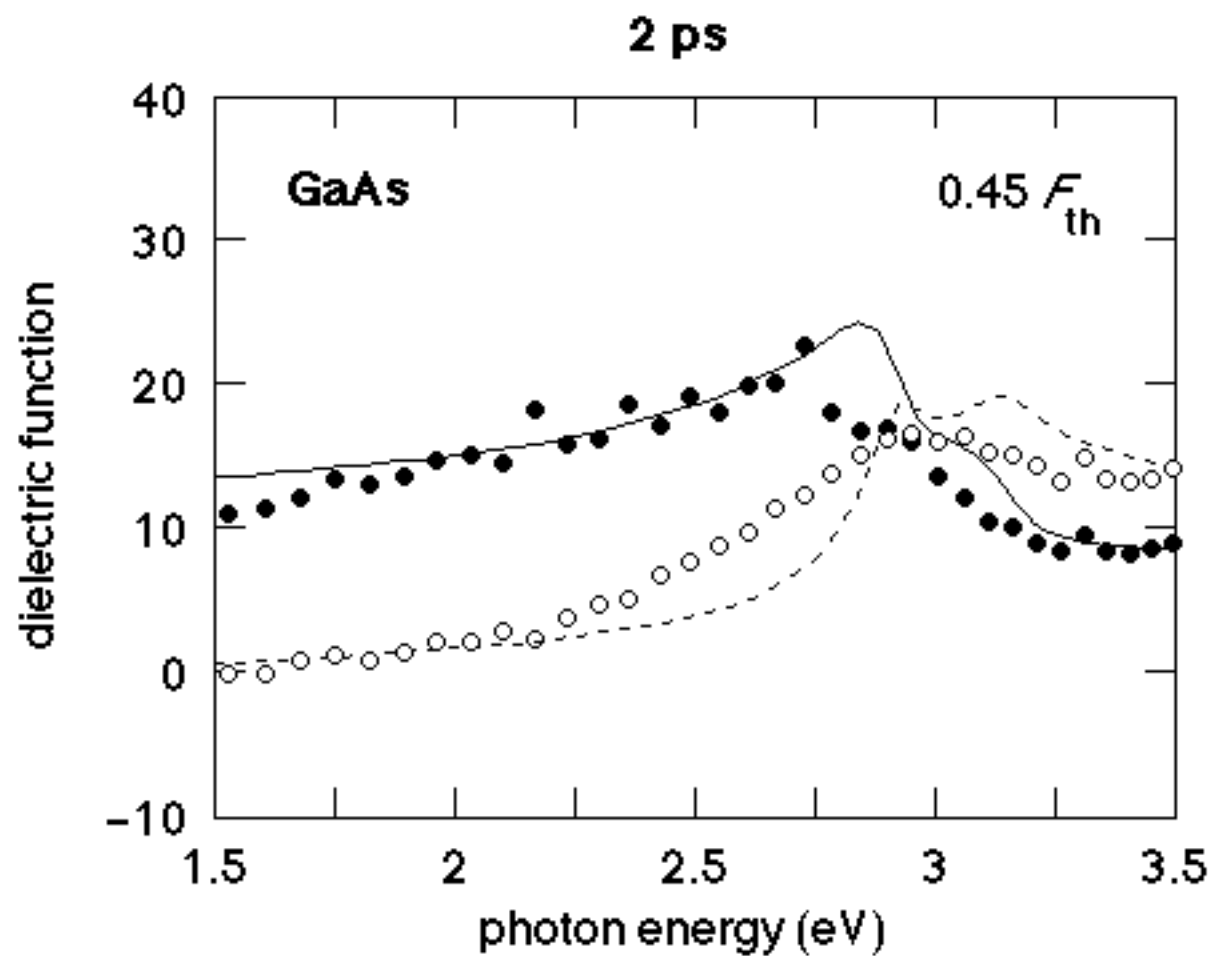
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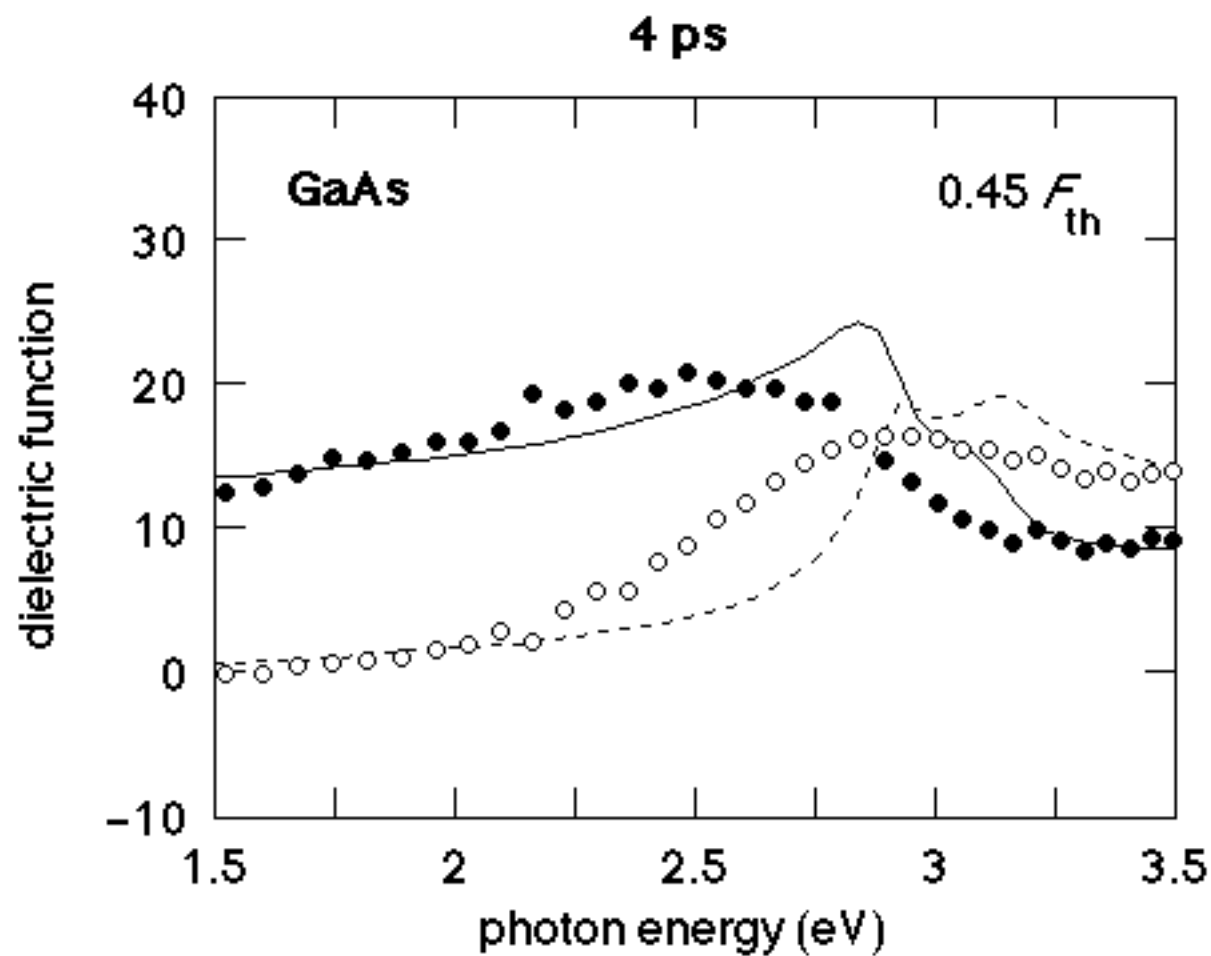
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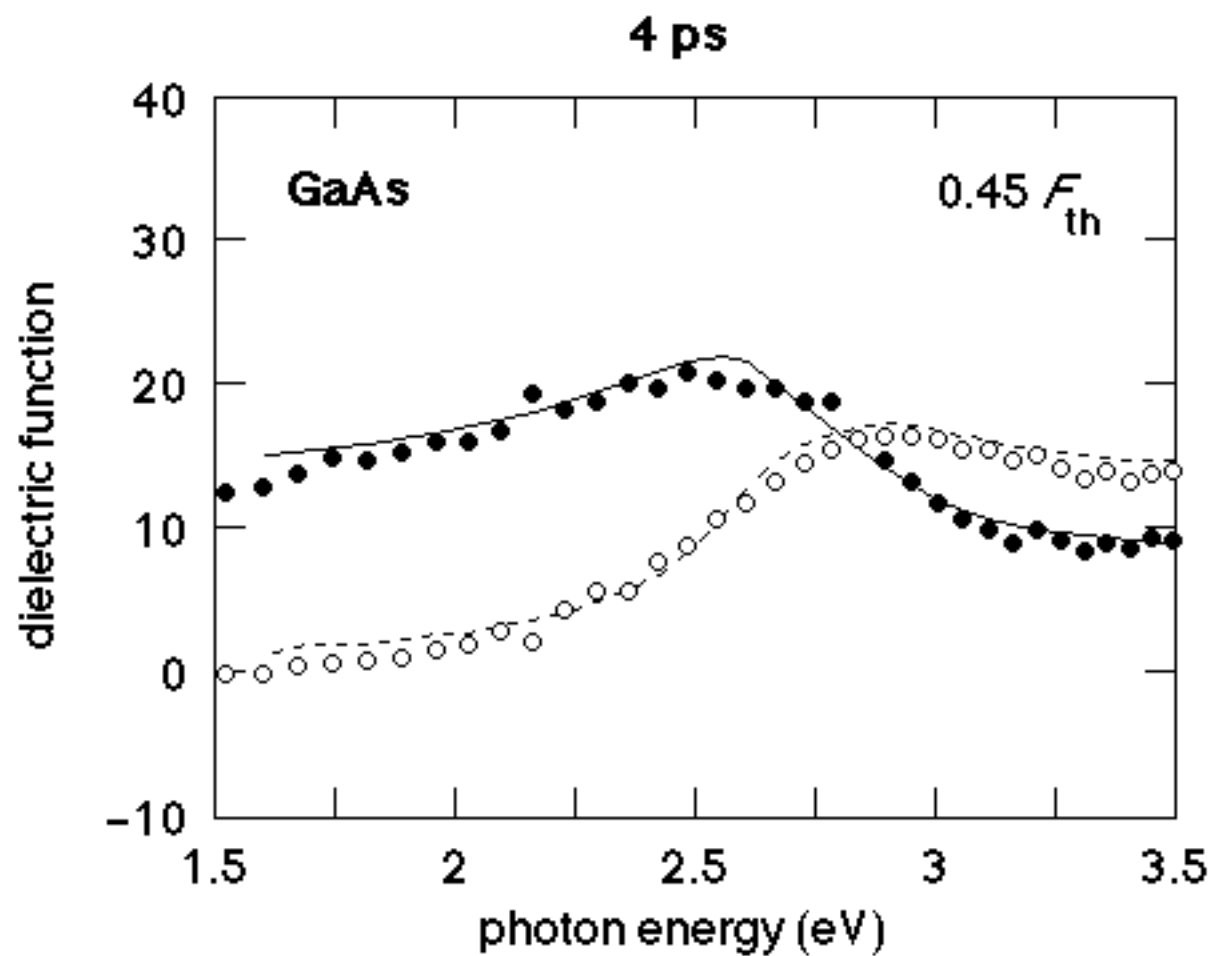
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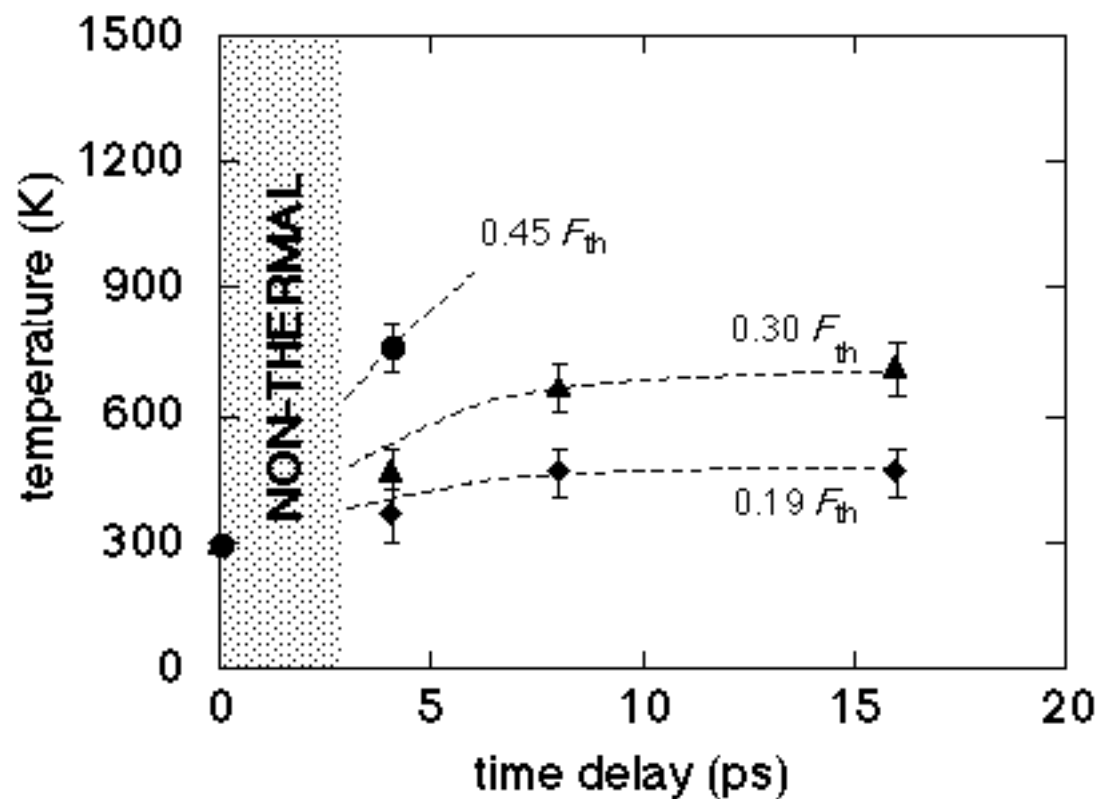
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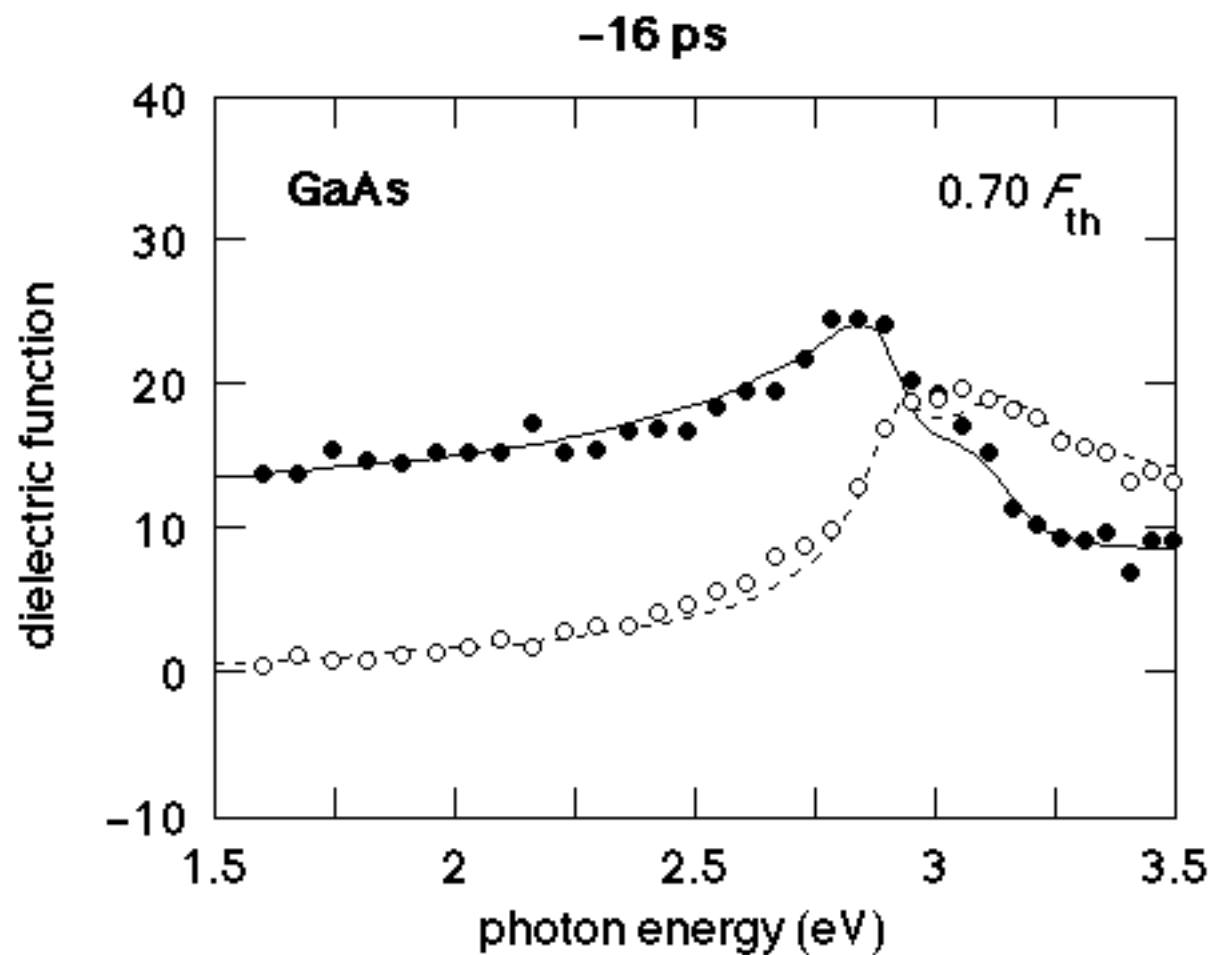
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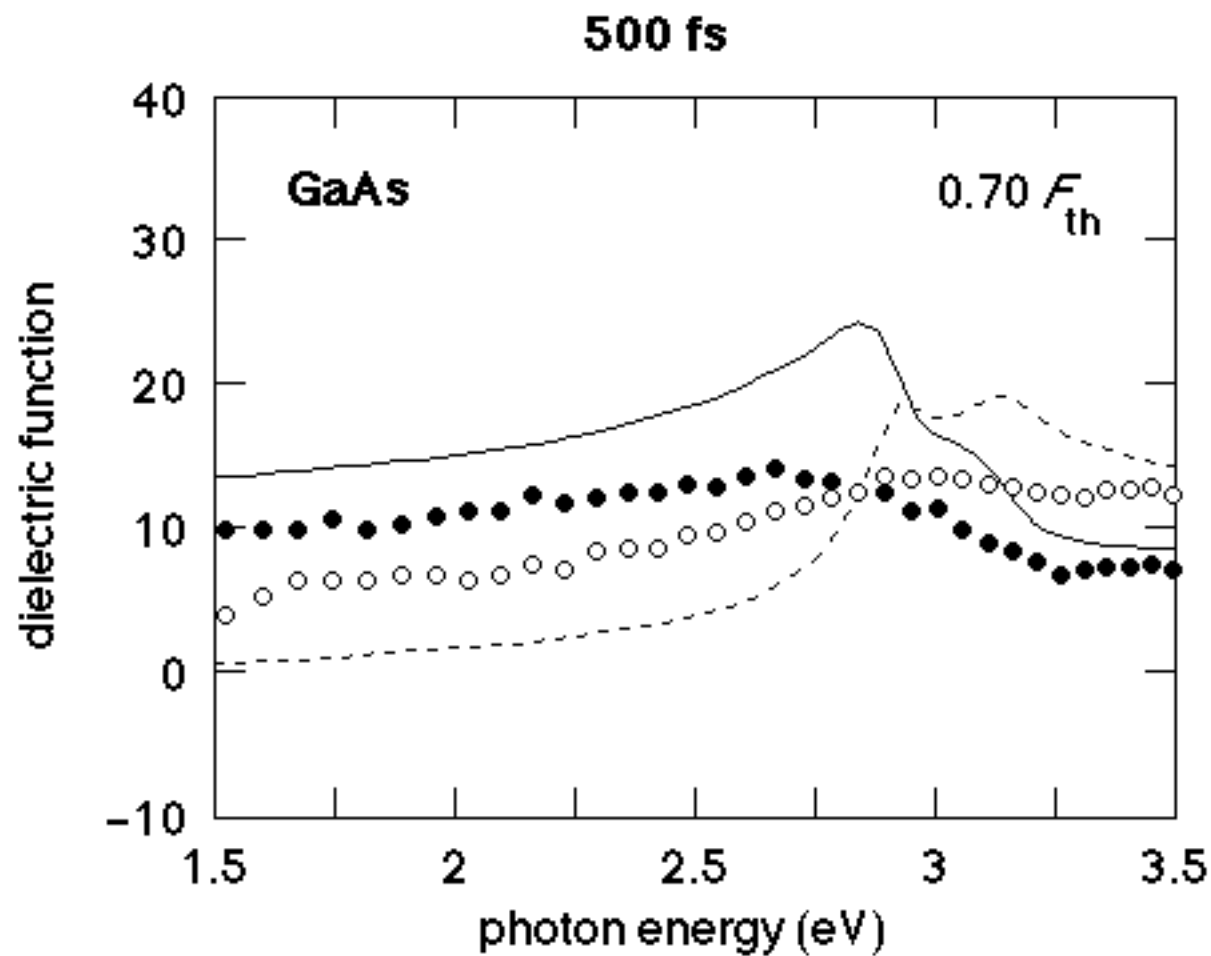
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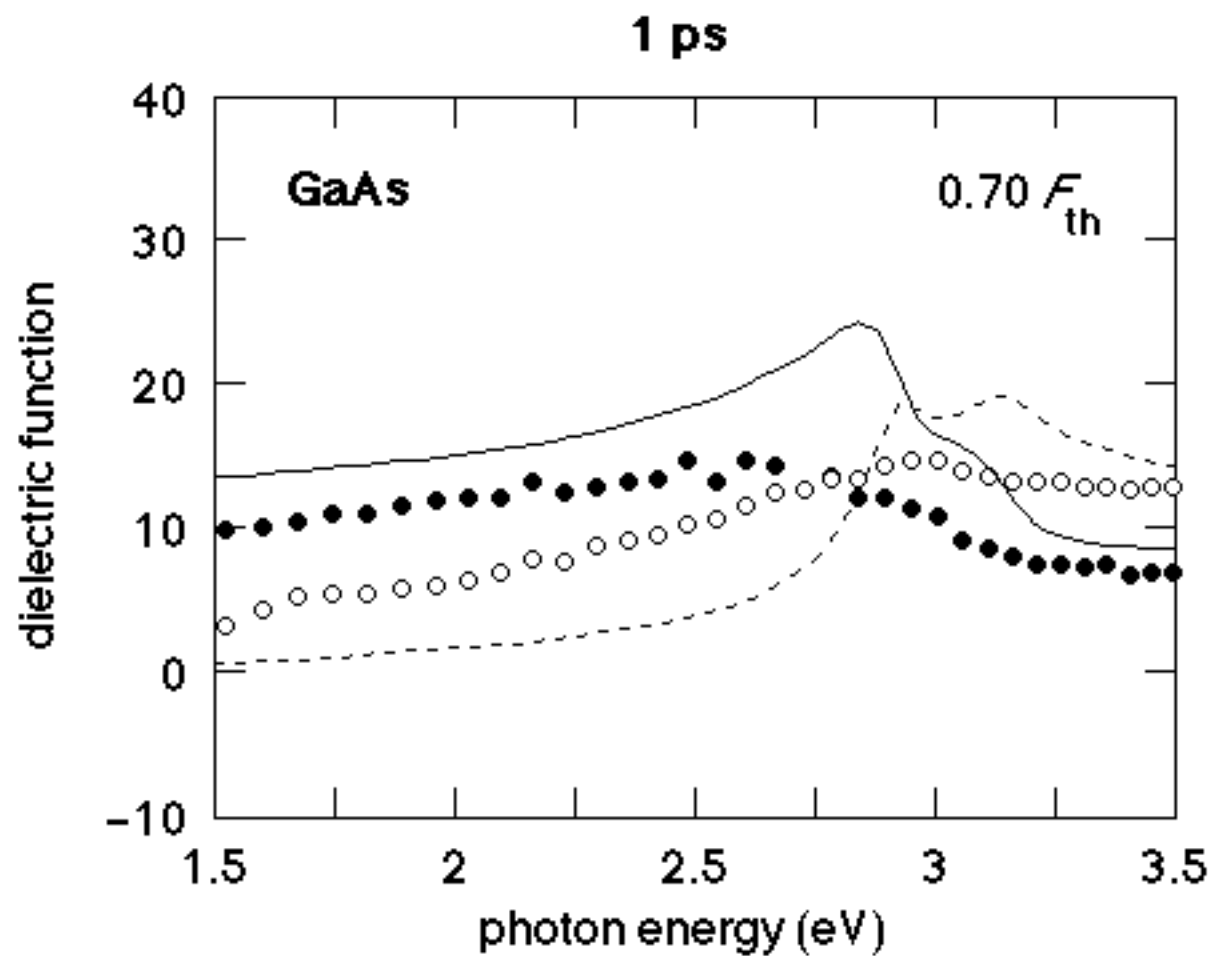
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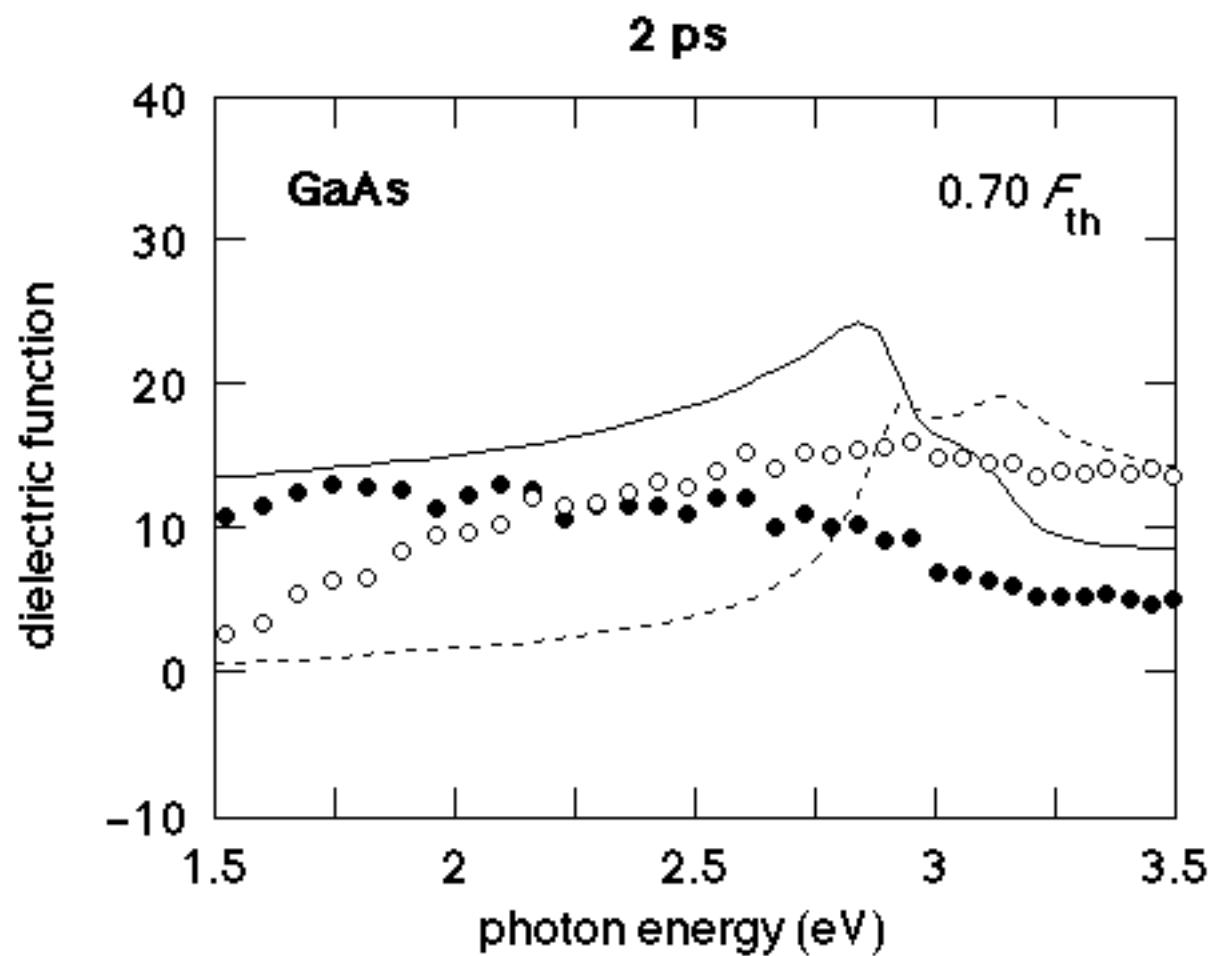
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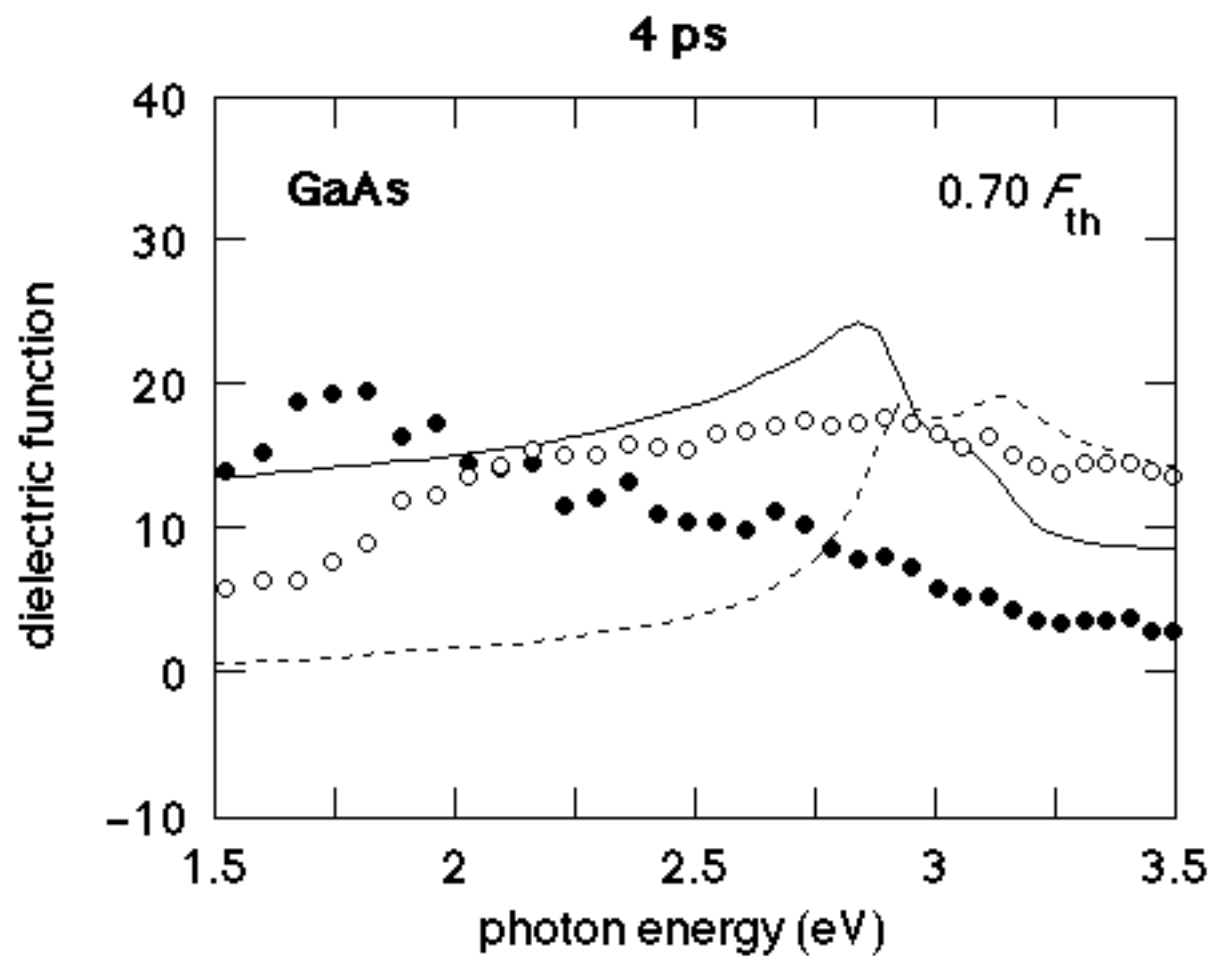
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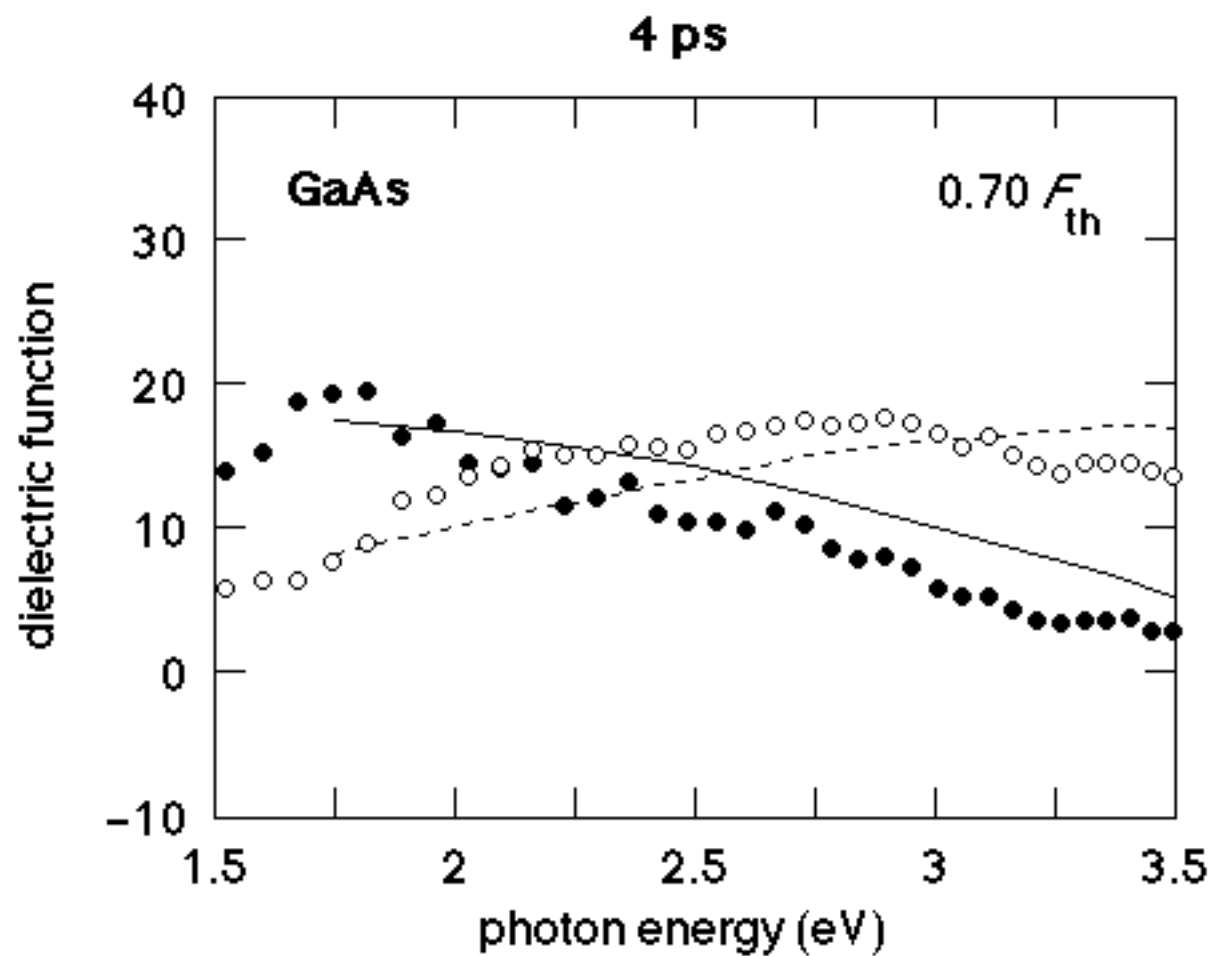
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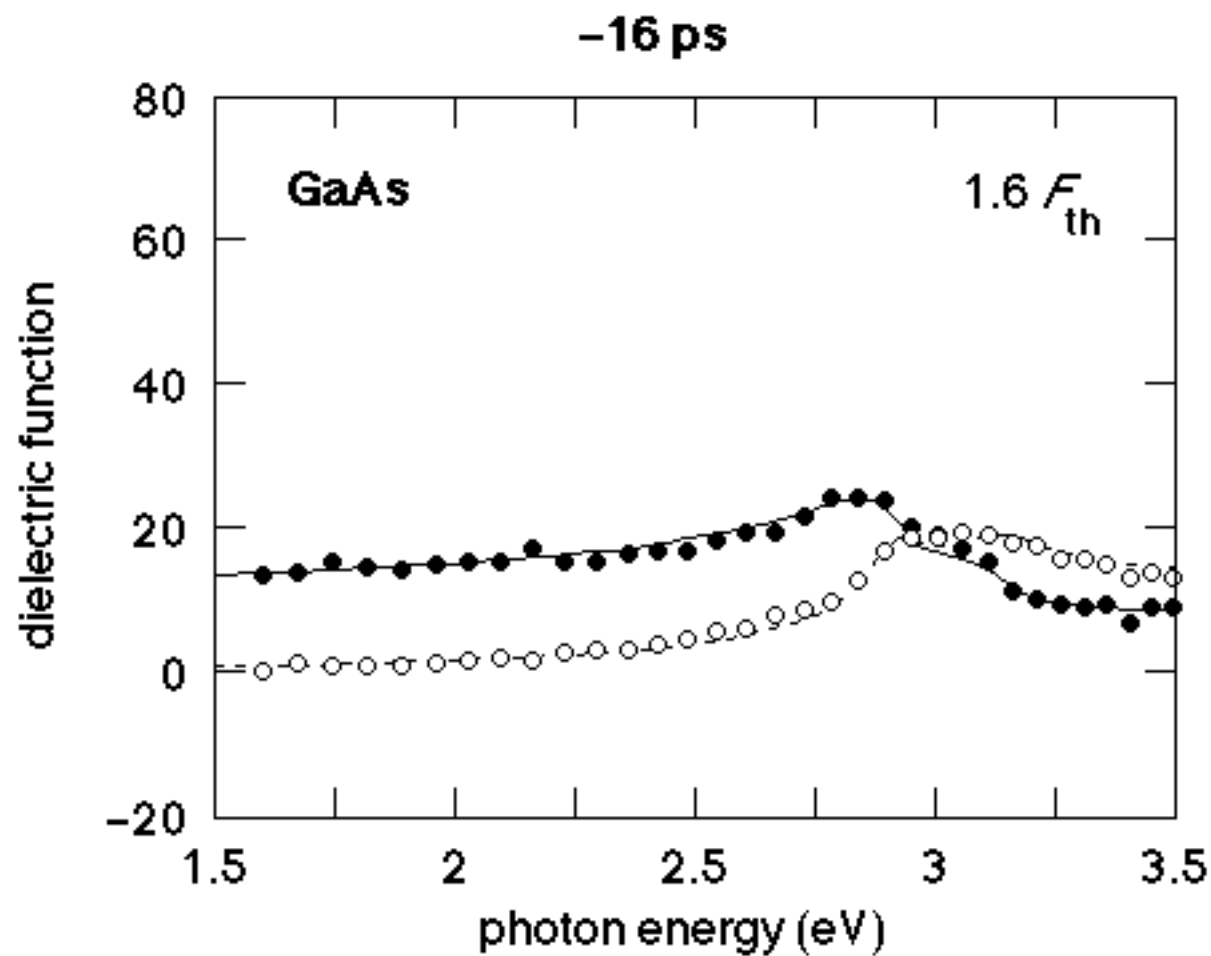
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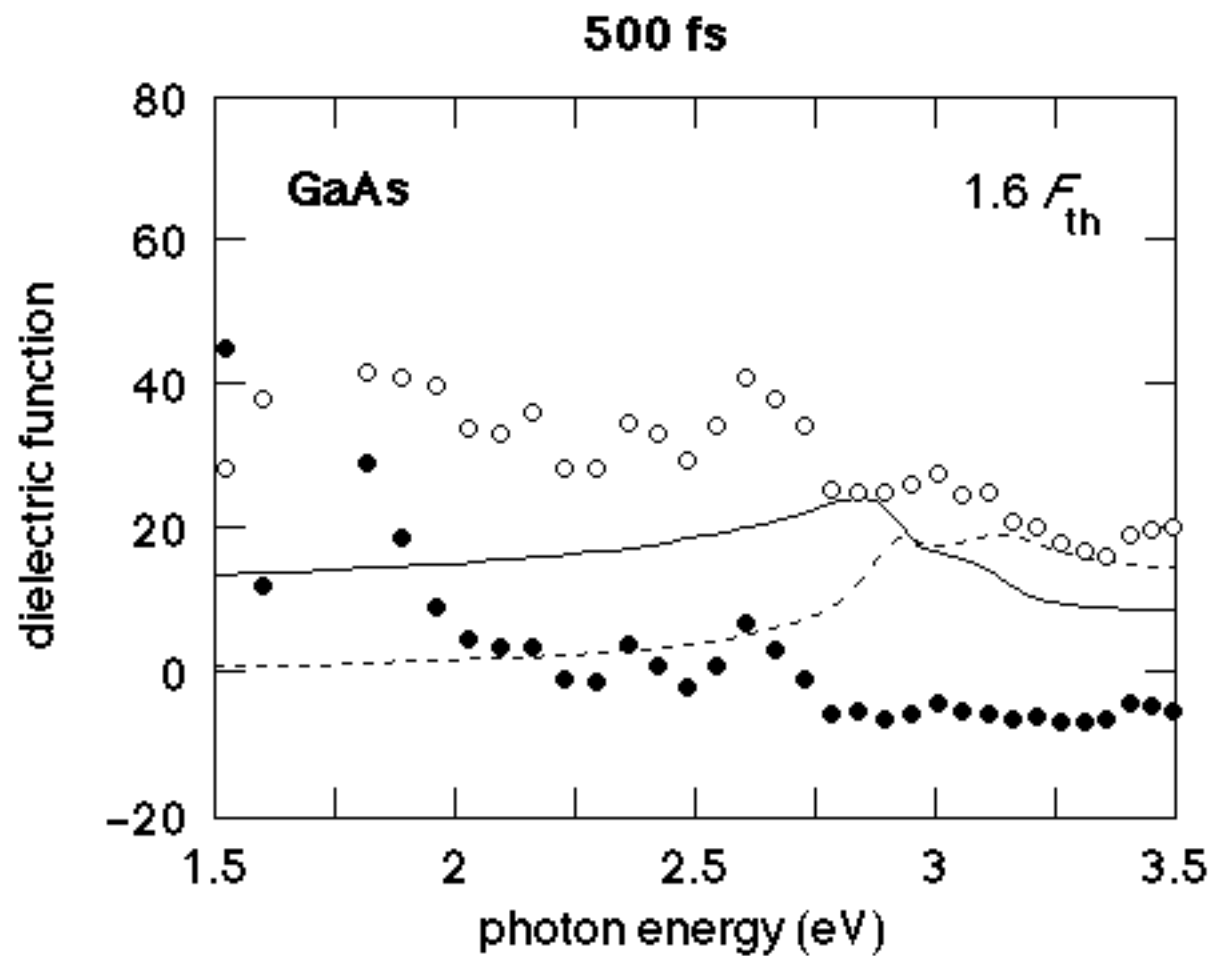
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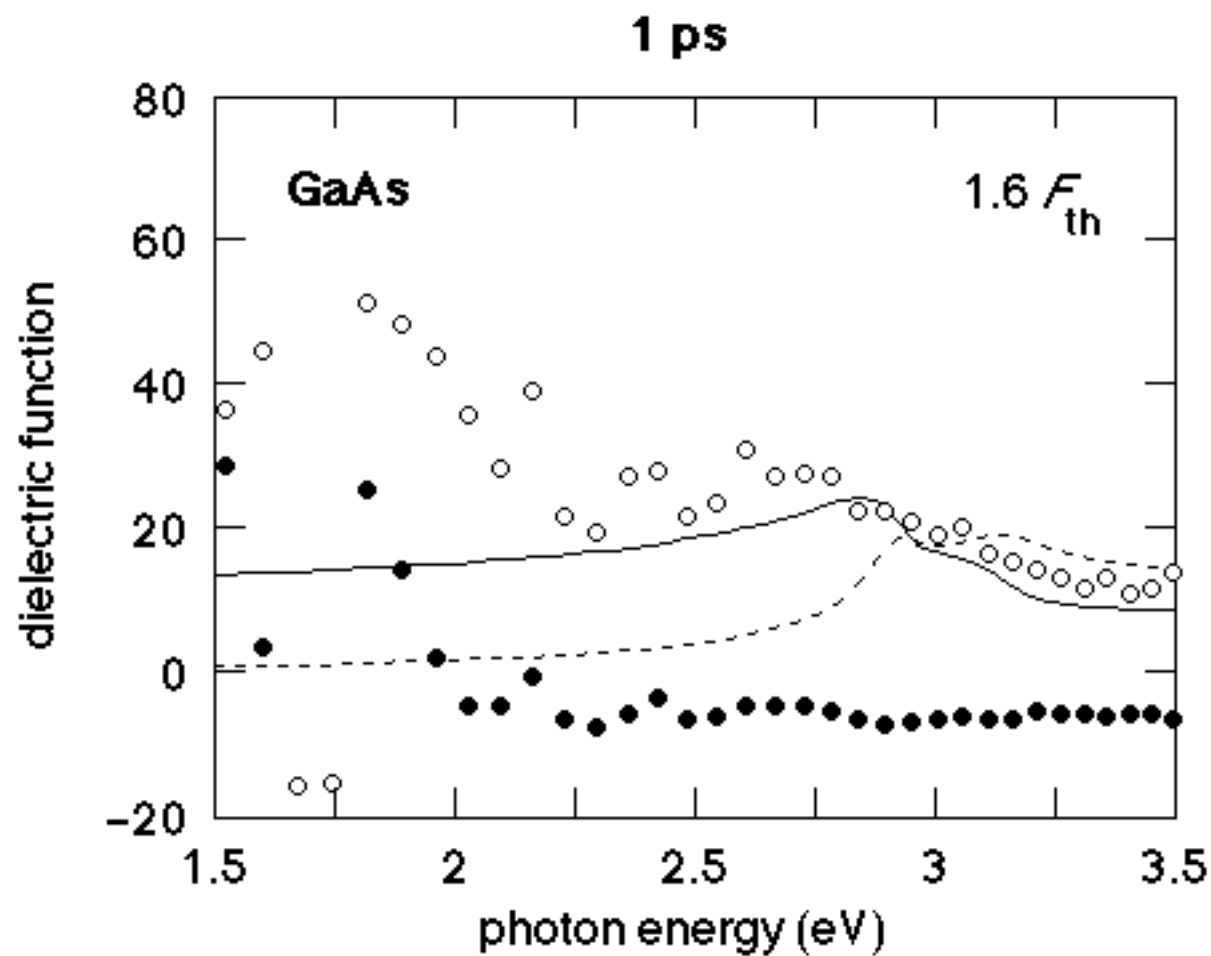
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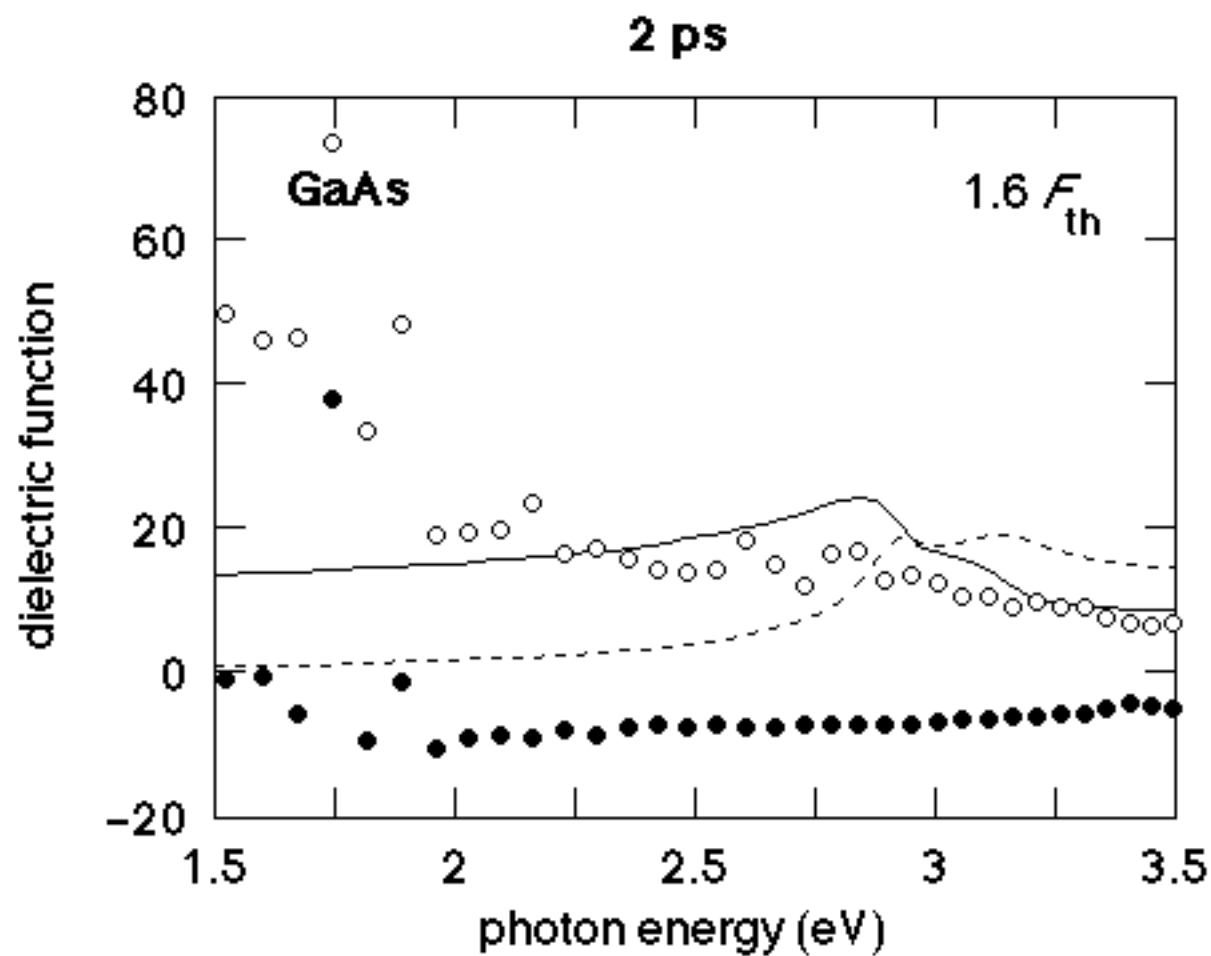
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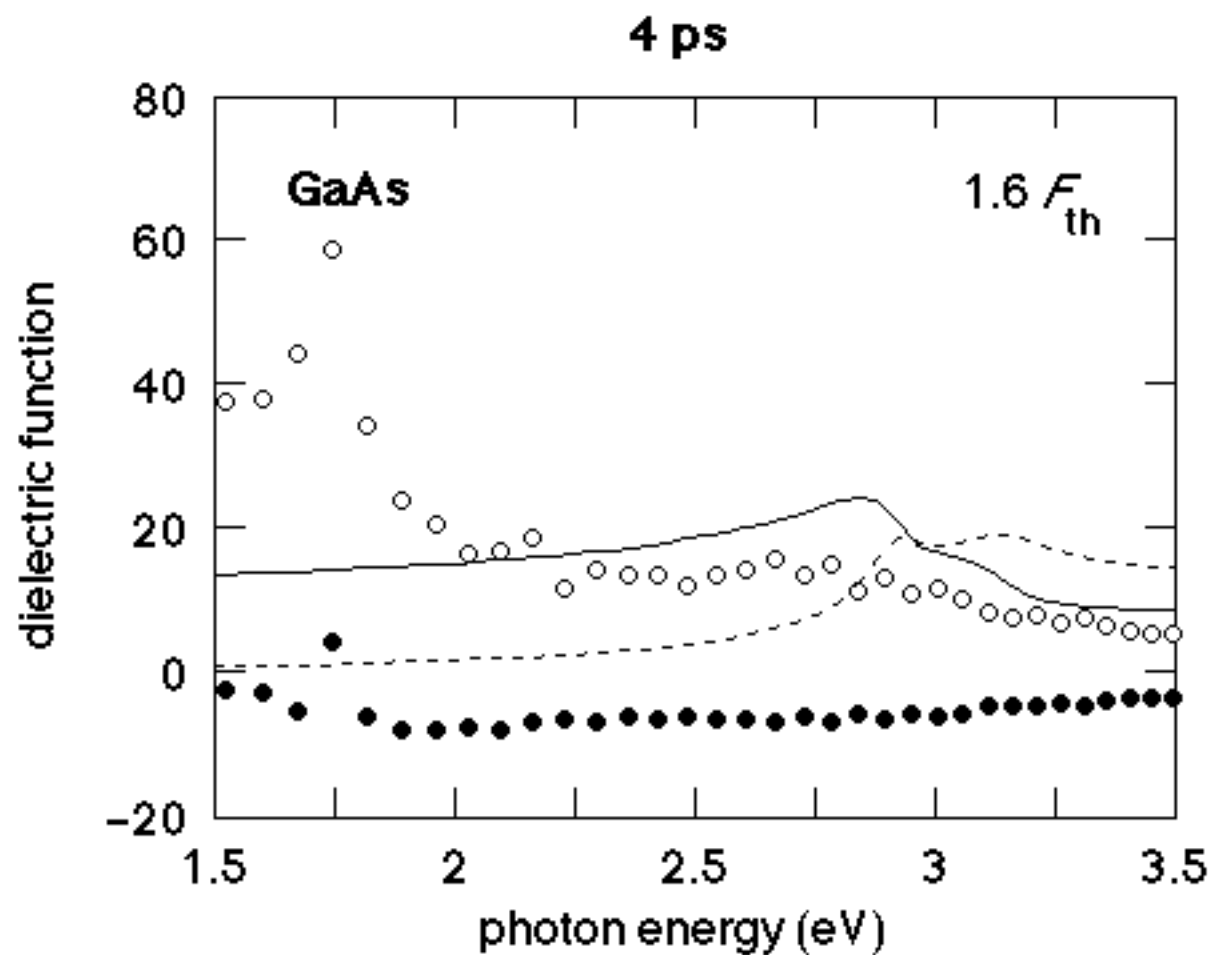
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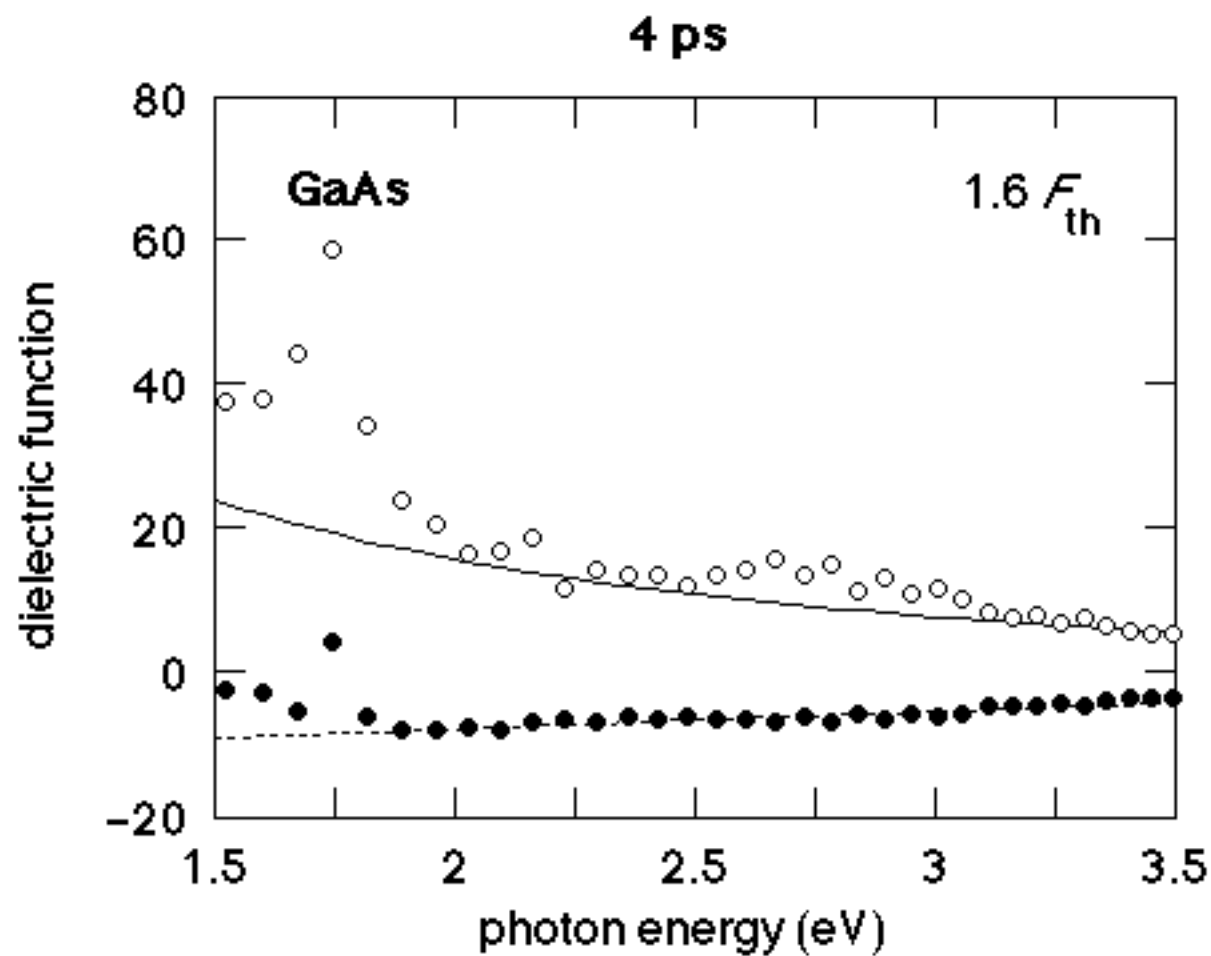
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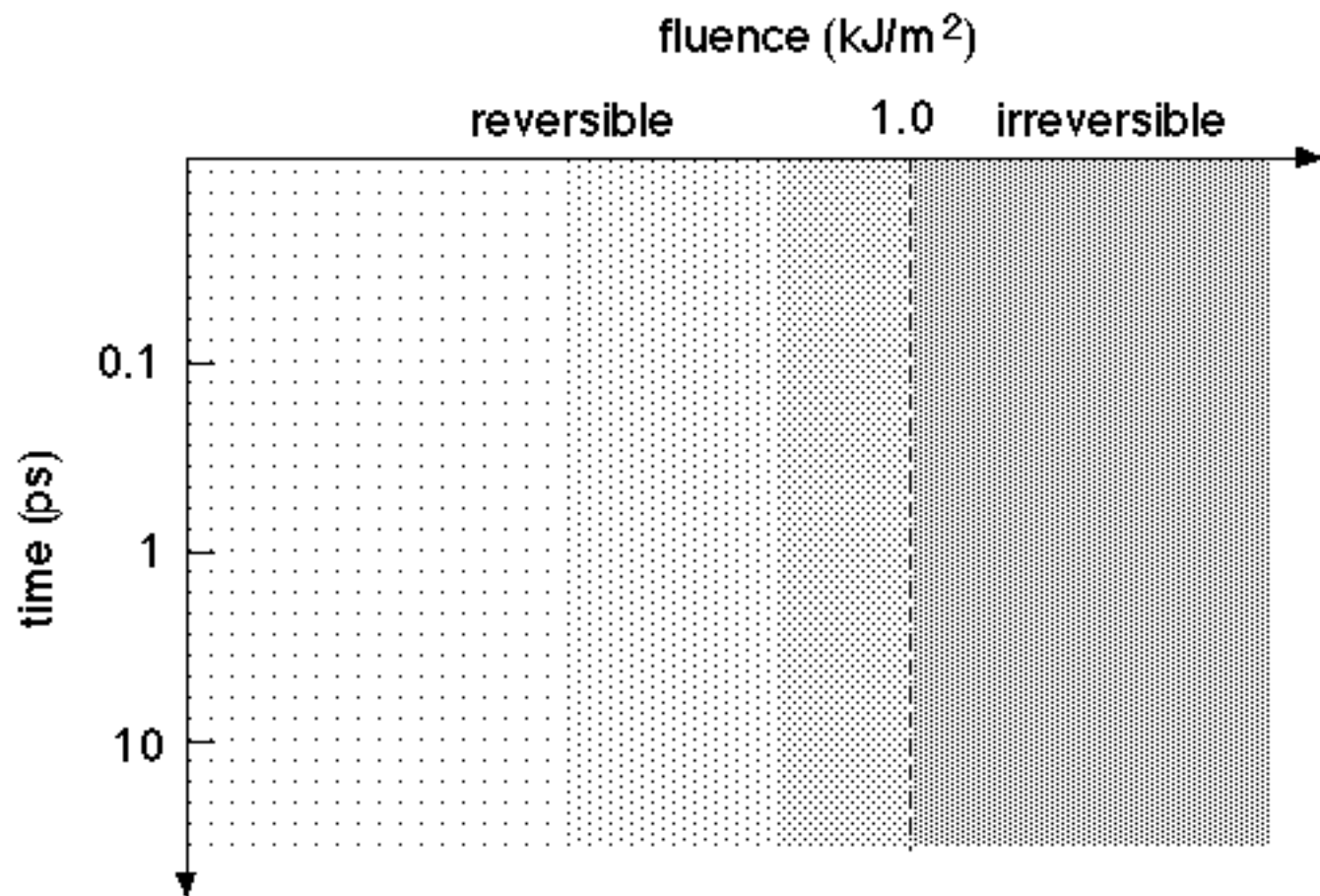
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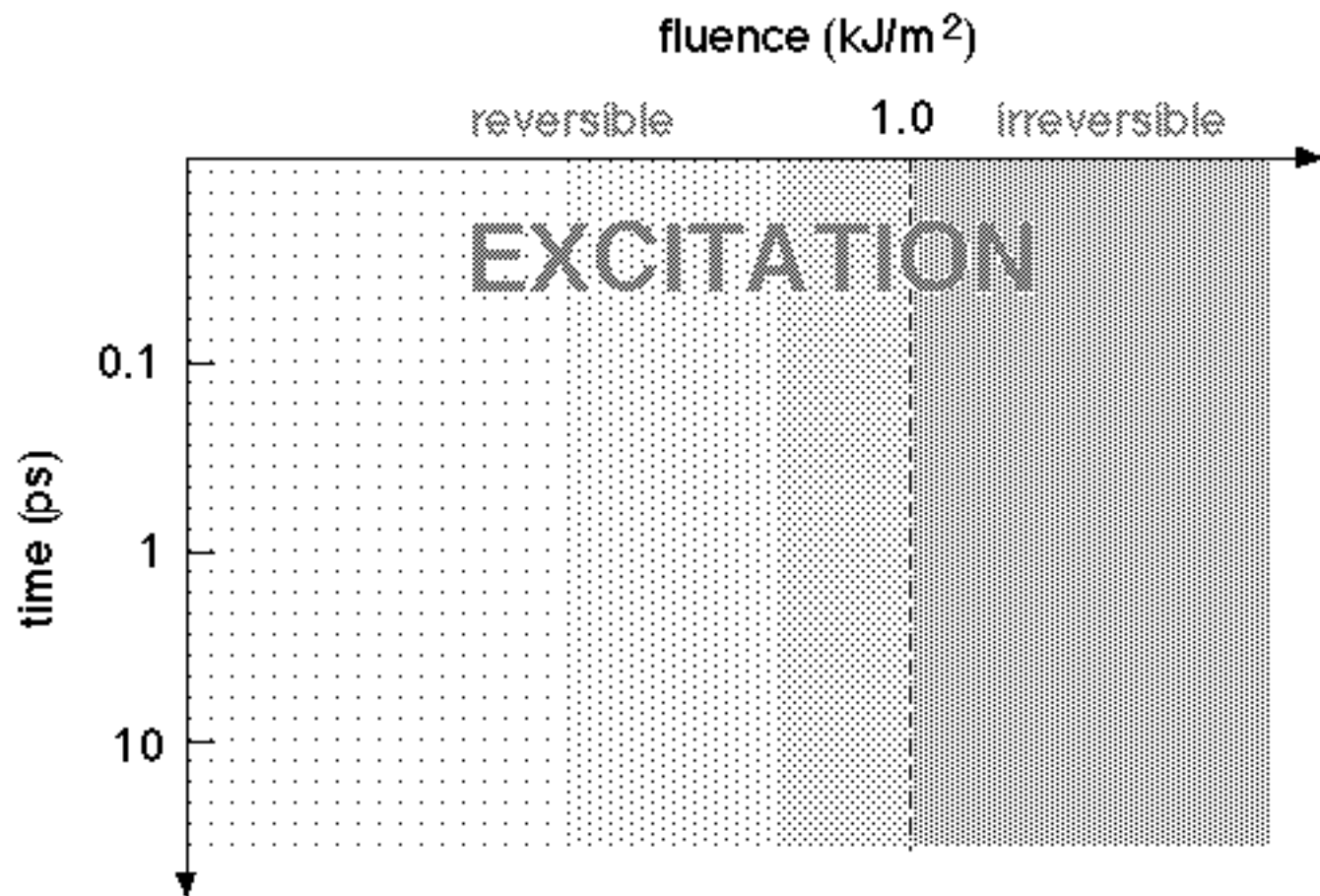
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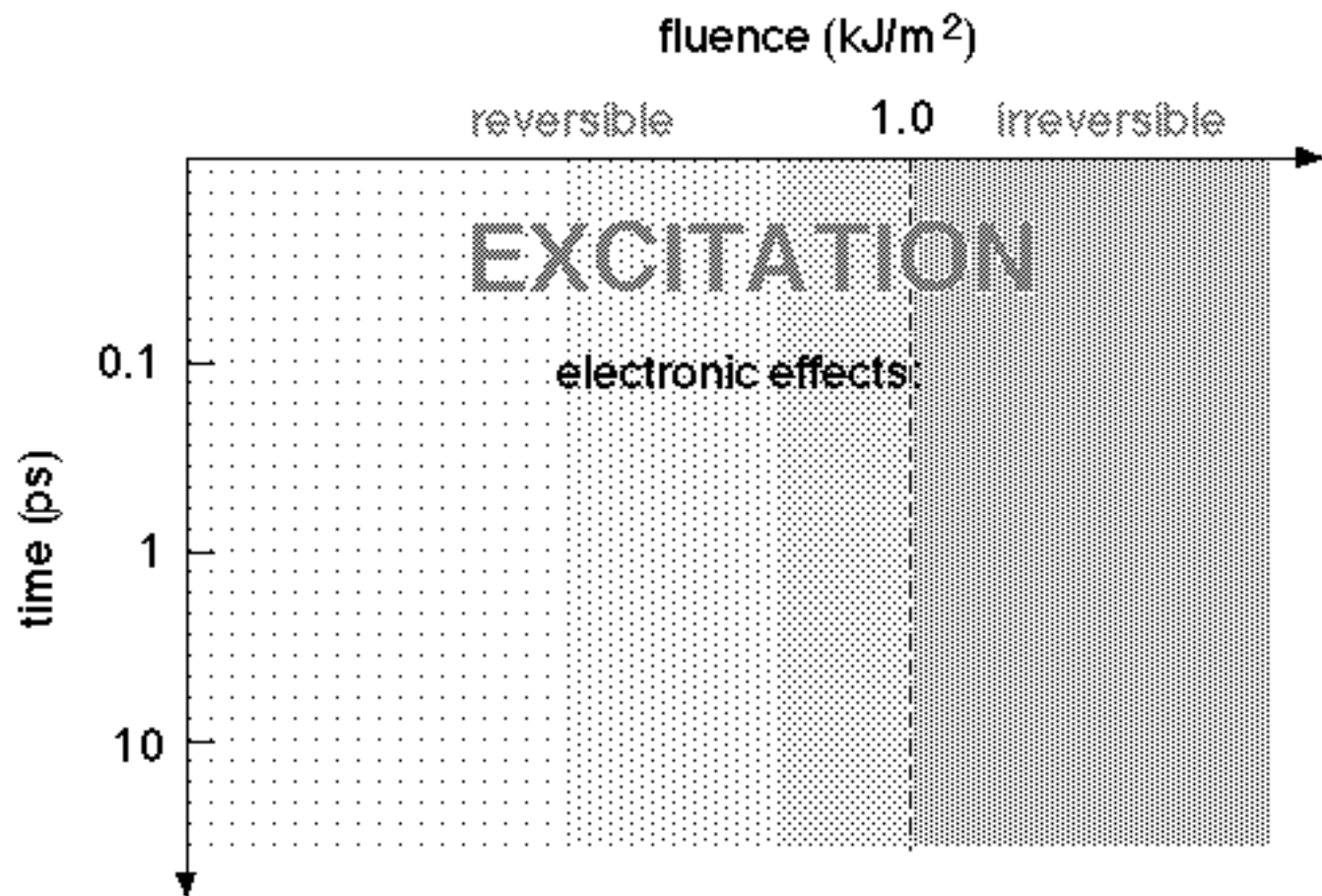
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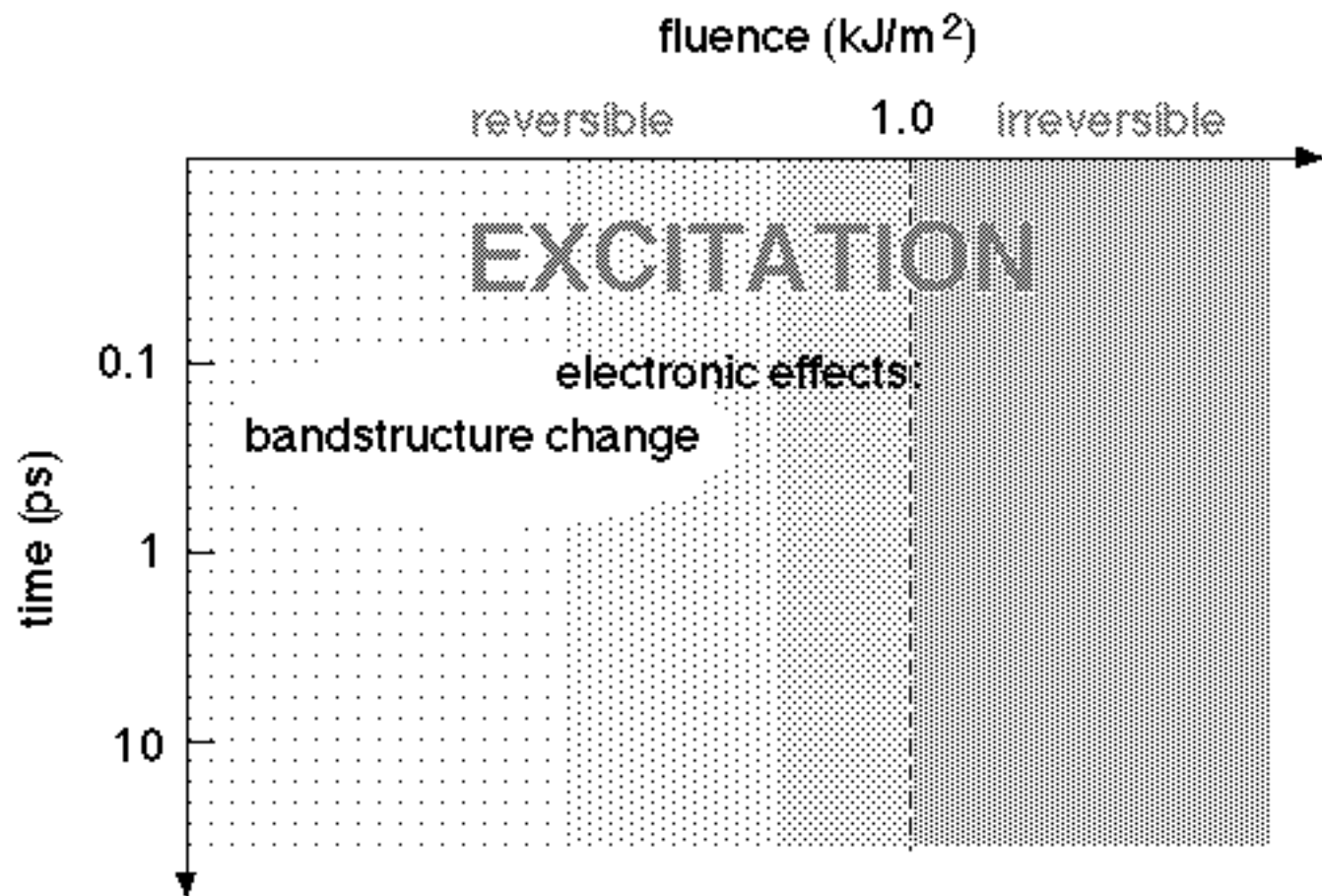
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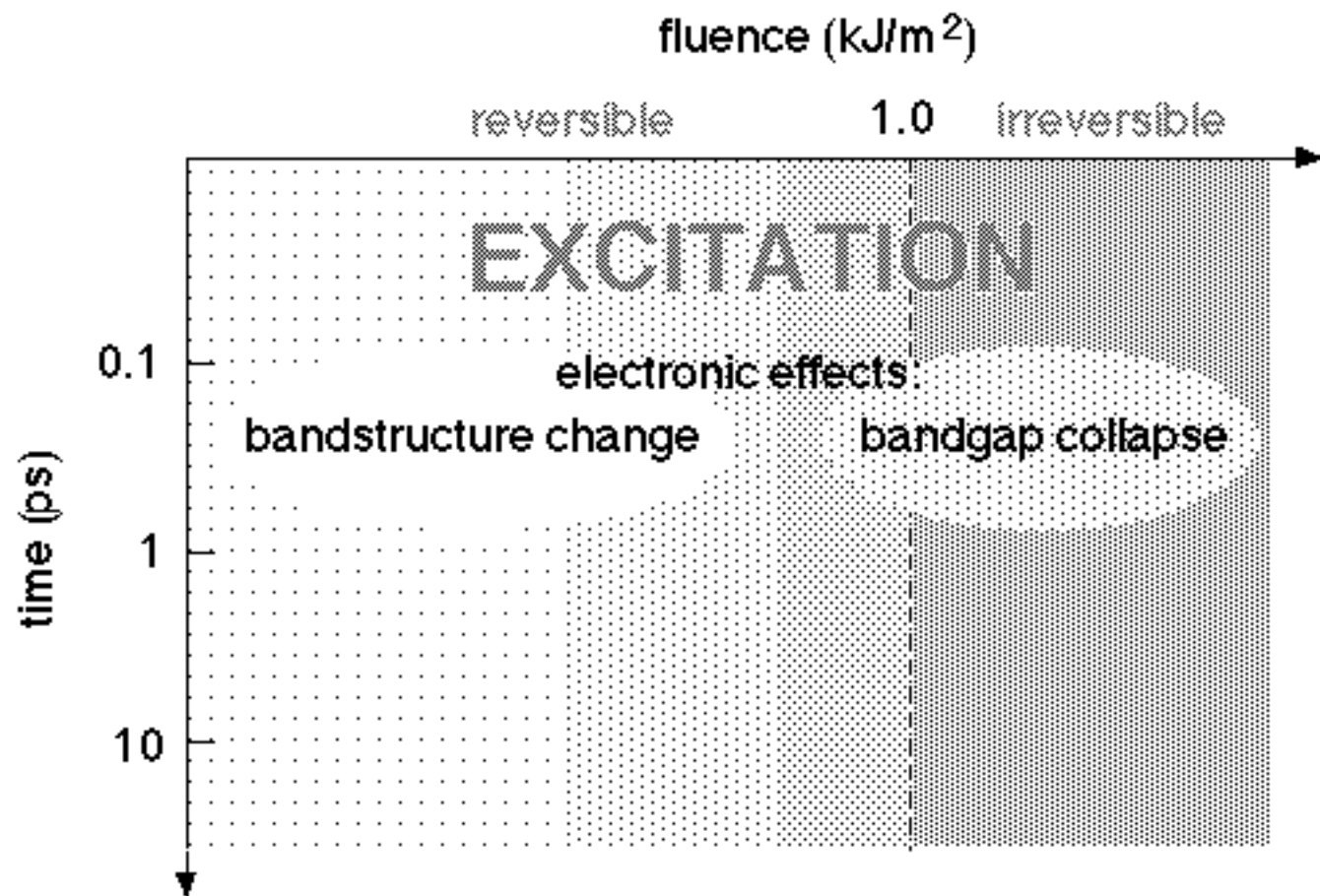
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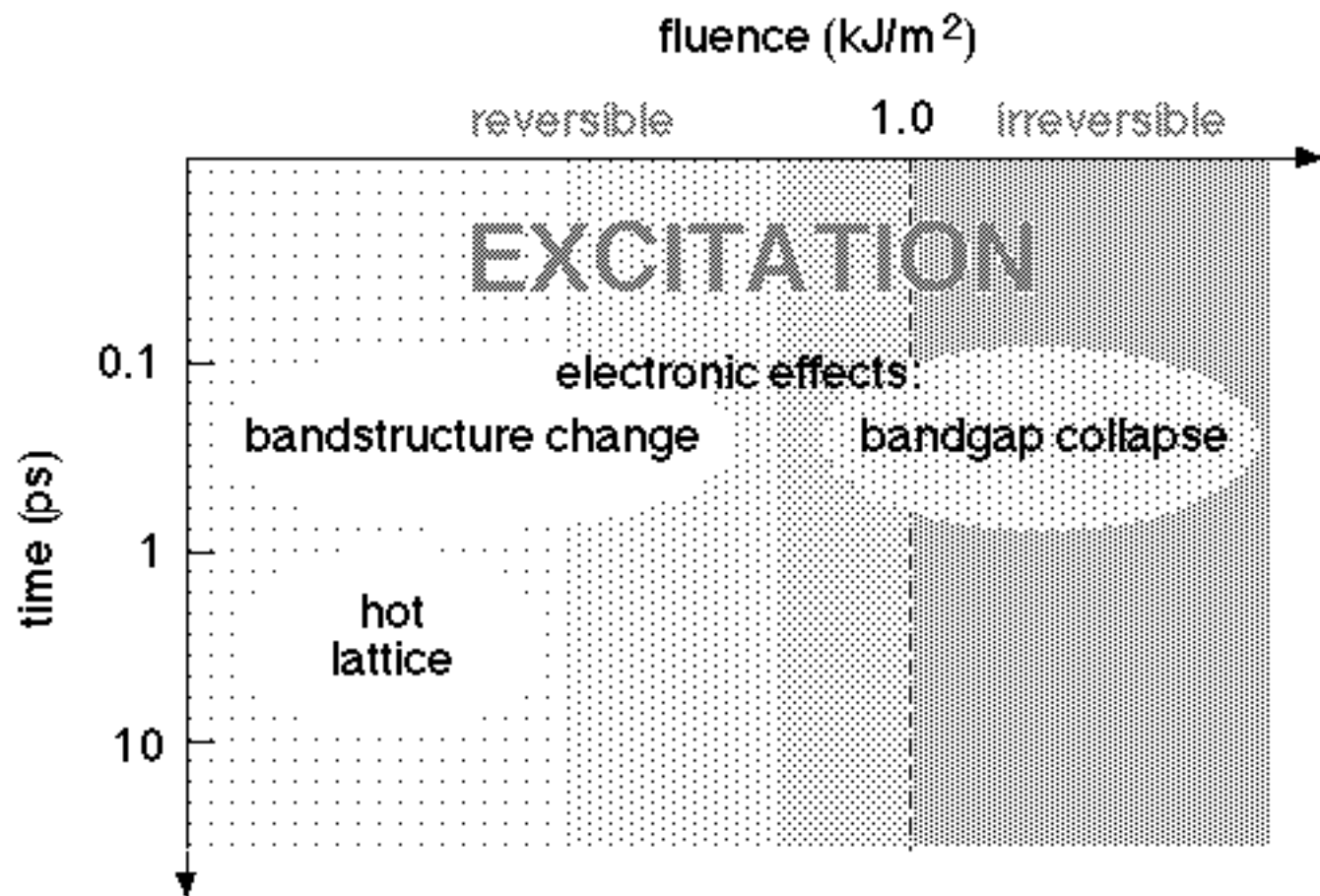
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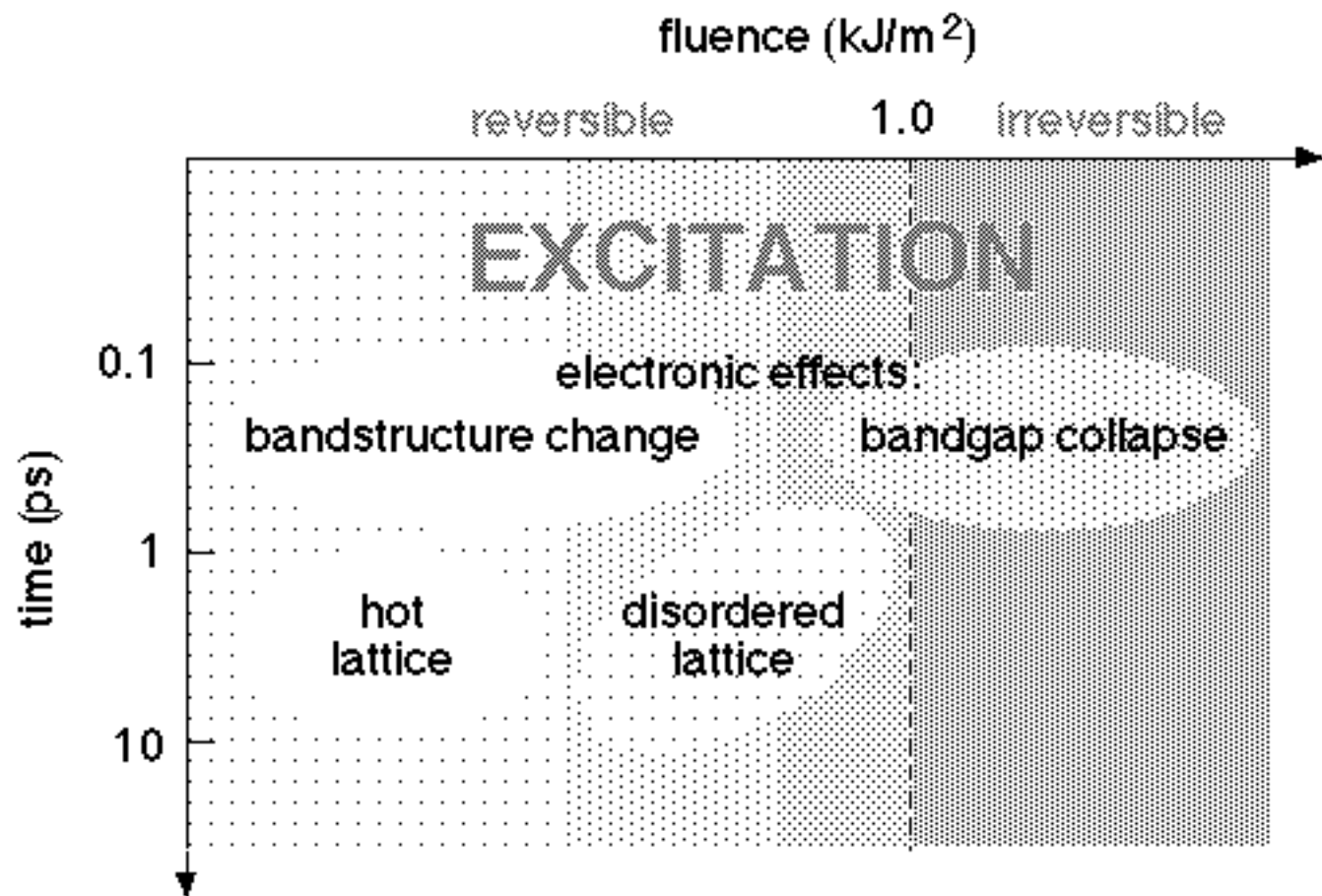
BROADBAND DATA



BROADBAND DATA



BROADBAND DATA



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



Acknowledgments

Prof. Bloembergen
Prof. Ehrenreich
Prof. Kaxiras
Prof. Aziz

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NSF DMR 89-20490

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