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ULTRAFAST LASER-INDUCED PHASE TRANSITIONS IN SEMICONDUCTORS

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Li Huang
Paul Callan
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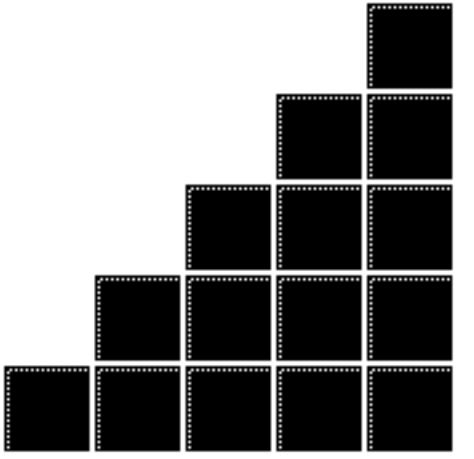
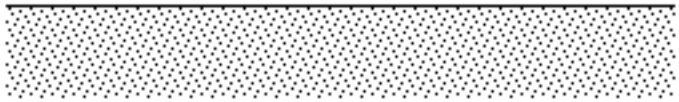
Boston College
18 September 1996



INTRODUCTION

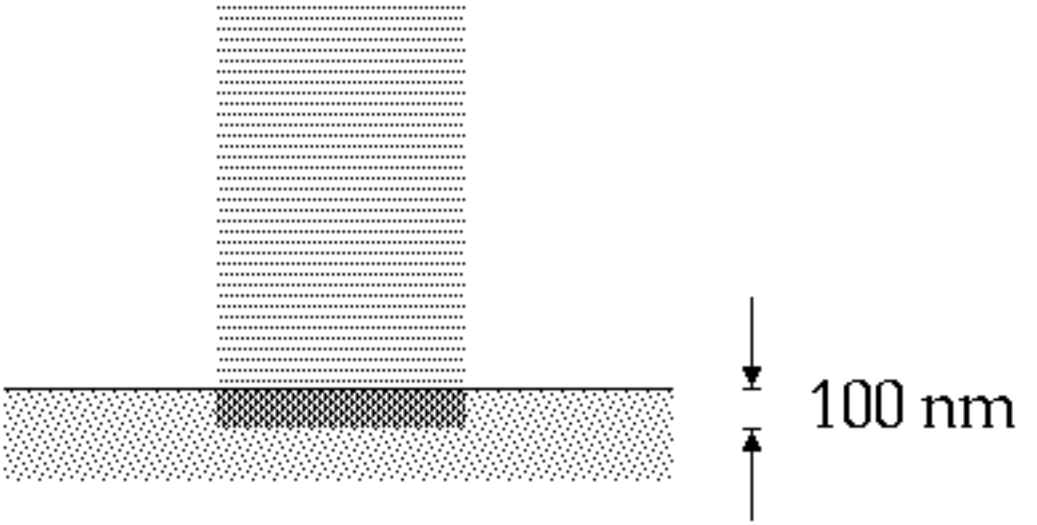
1976: (nanosecond) laser annealing of crystals damaged by ion bombardment

1985: femtosecond laser 'melting' of Si



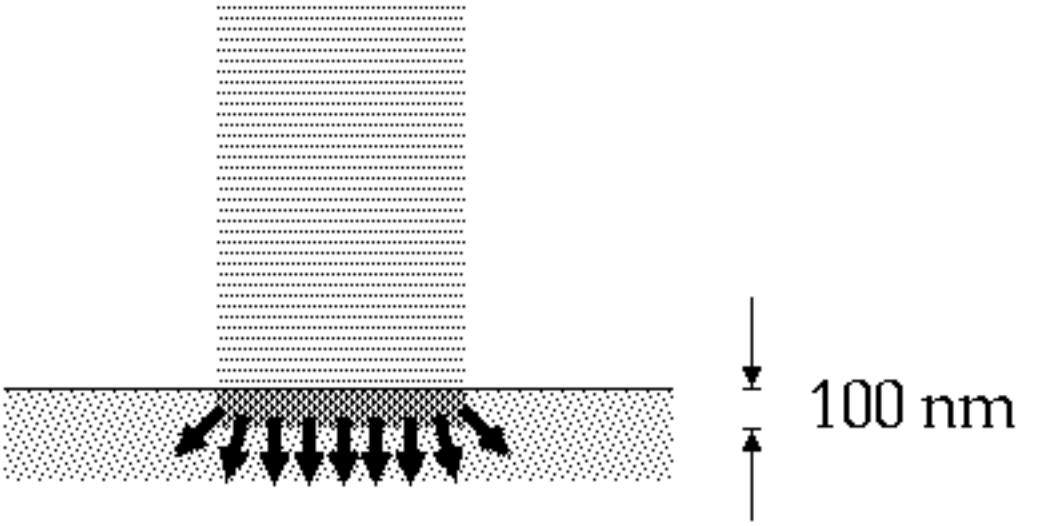
INTRODUCTION

laser deposits energy near surface...



INTRODUCTION

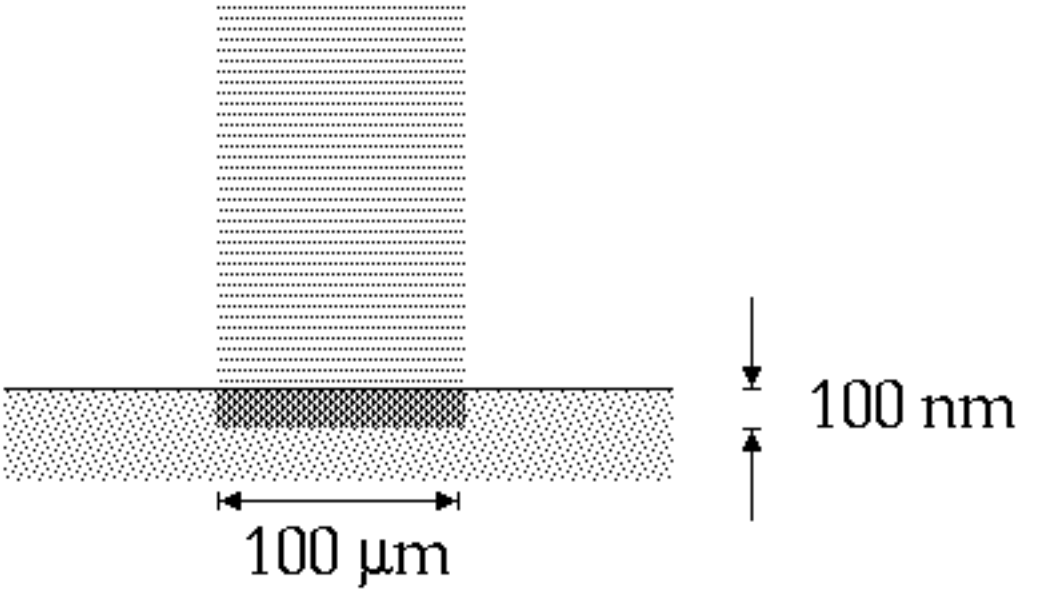
energy rapidly diffuses into bulk...



in 1 ns, diffusion length ≈ 100 nm

INTRODUCTION

For $t < 1$ ns can ignore diffusion



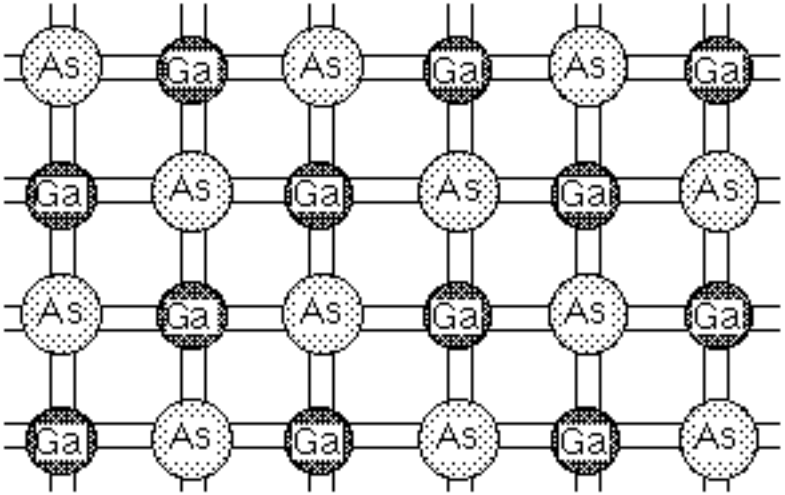
10 µJ into a volume of about 10^{-15} m^3!

❶ Background

❷ Single frequency work

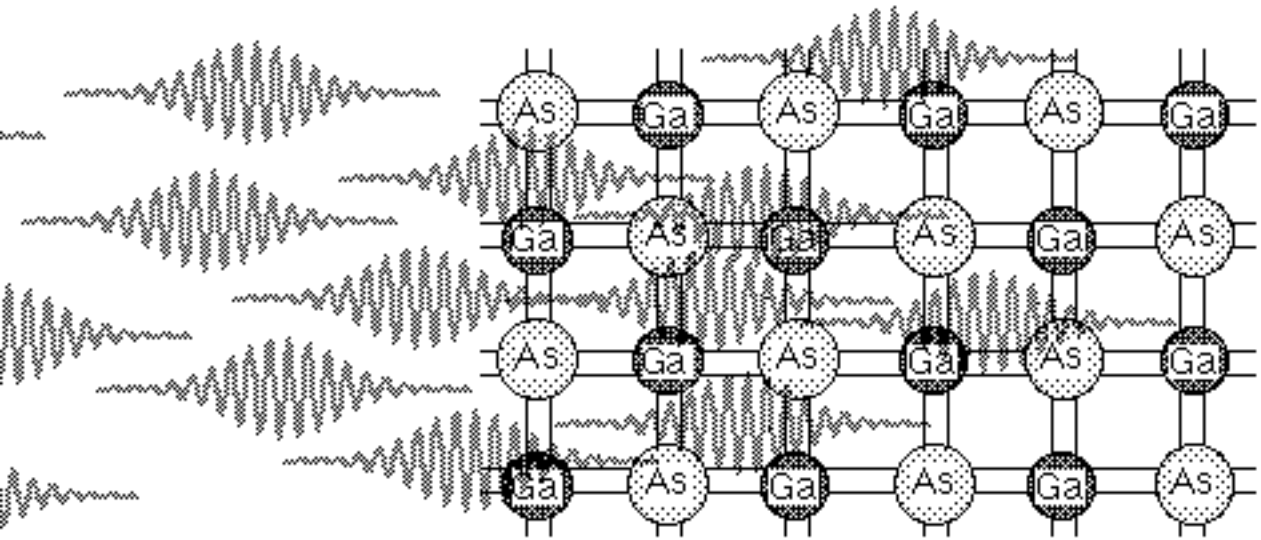
❸ Broadband work

EARLY MODELS



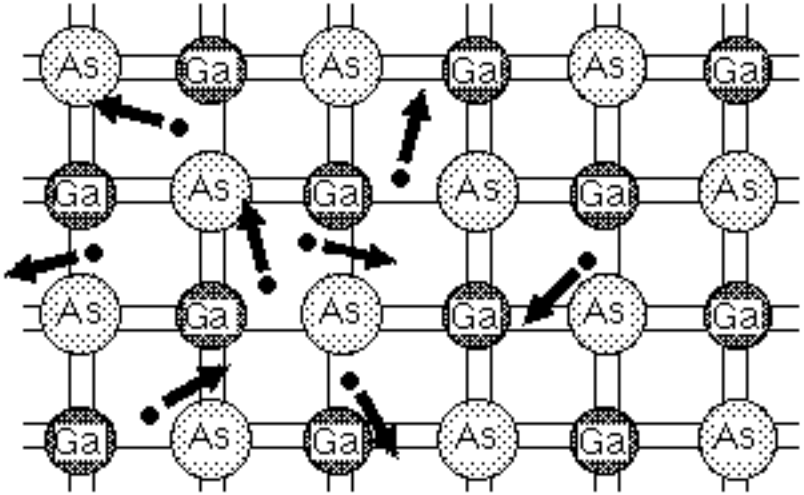
how does light melt a solid?

THERMAL MODEL



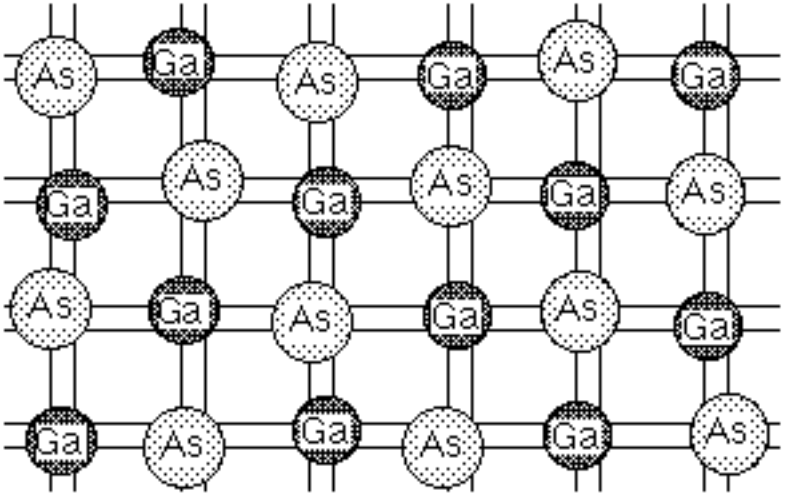
photons excite valence electrons...

THERMAL MODEL



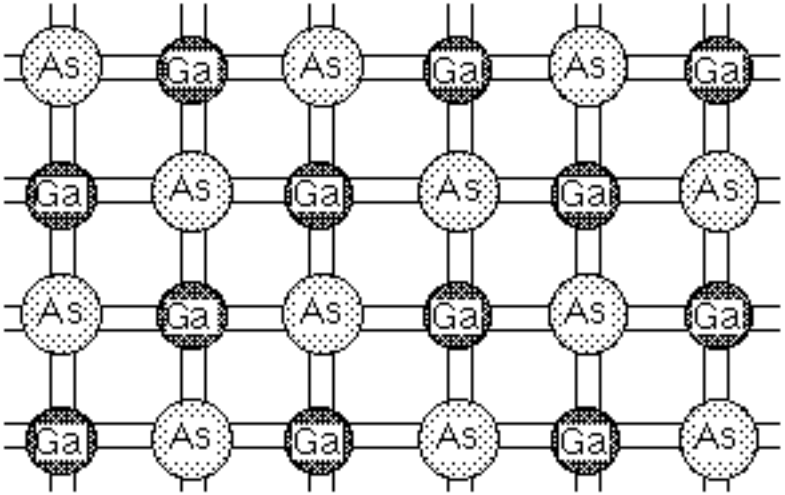
...and create hot electrons...

THERMAL MODEL



...which heat lattice

THERMAL MODEL



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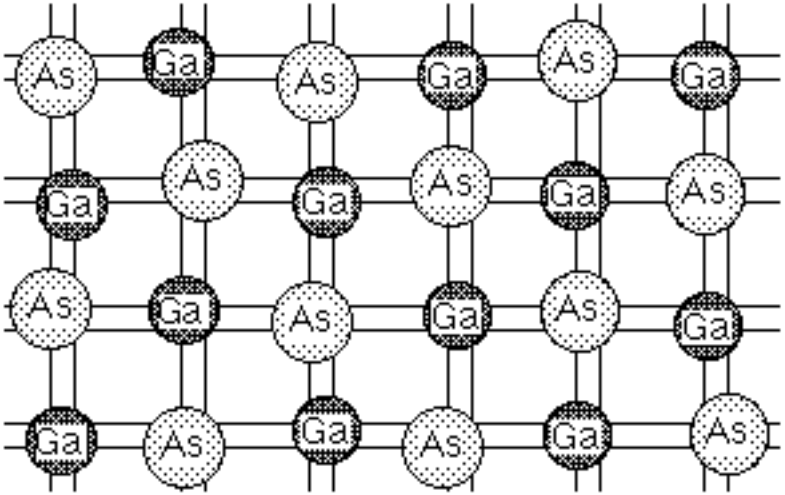
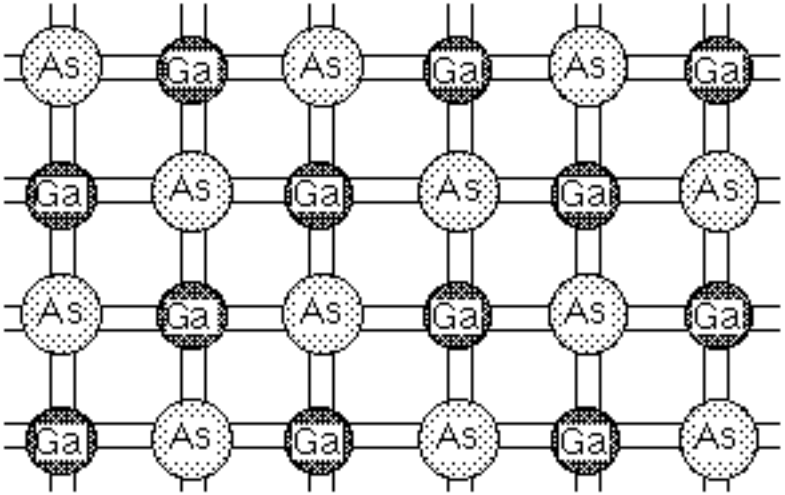
THERMAL MODEL



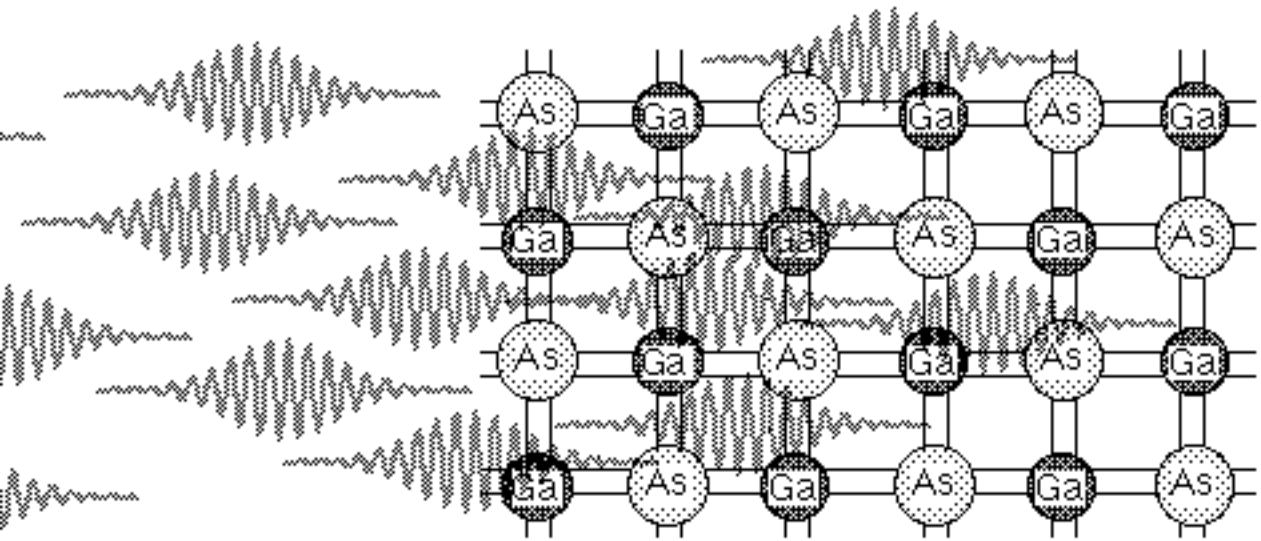
...which heat lattice

EARLY MODELS



...or...

PLASMA ANNEALING



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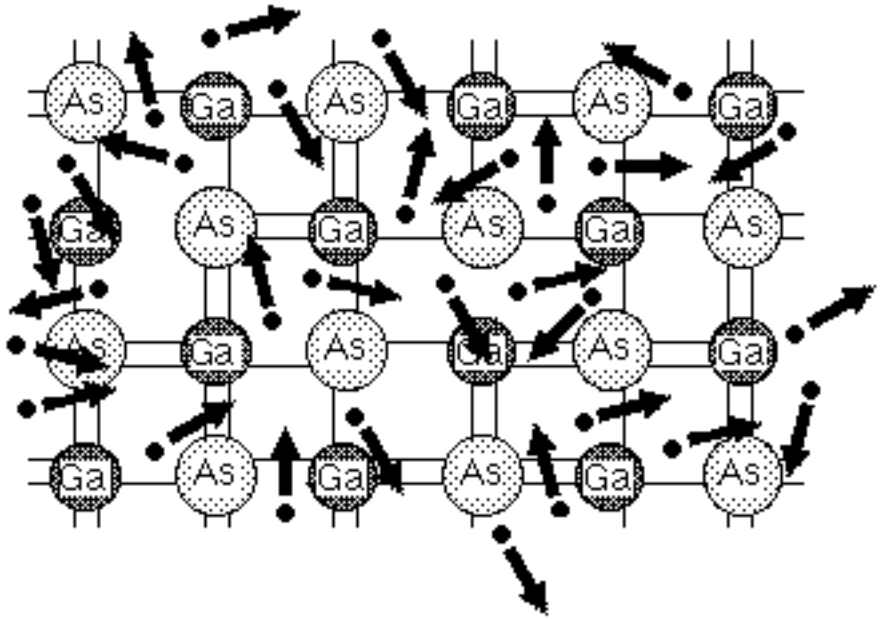
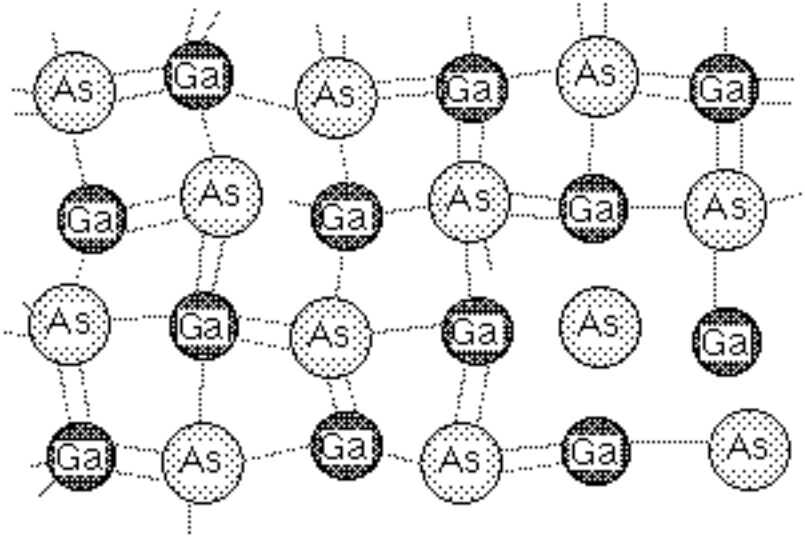


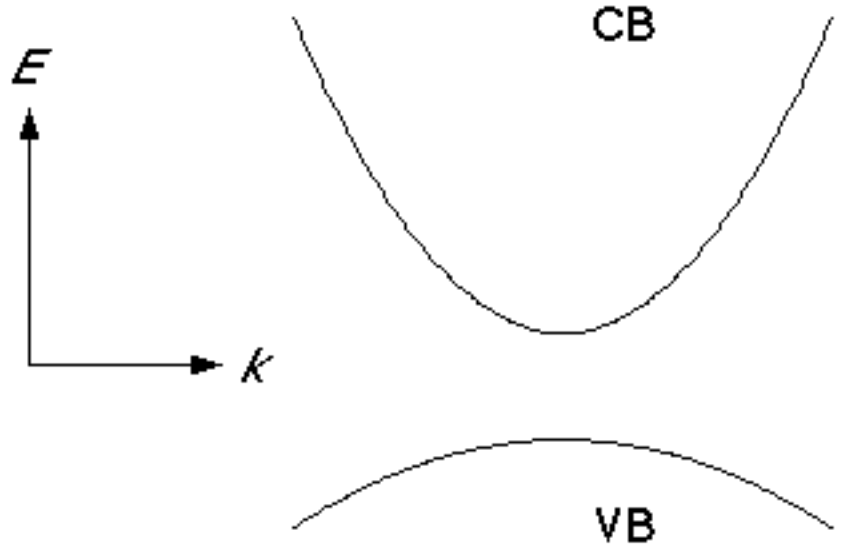
...creating a dense plasma...

PLASMA ANNEALING

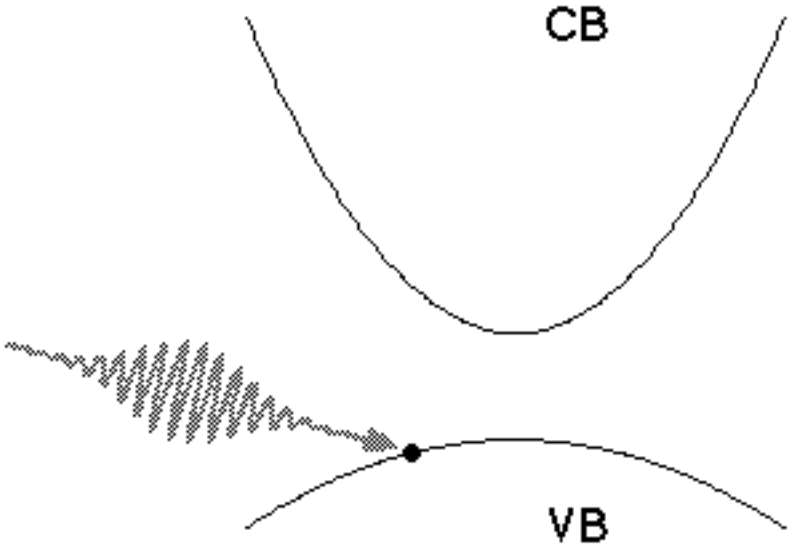


...and breaking bonds

EXCITATION

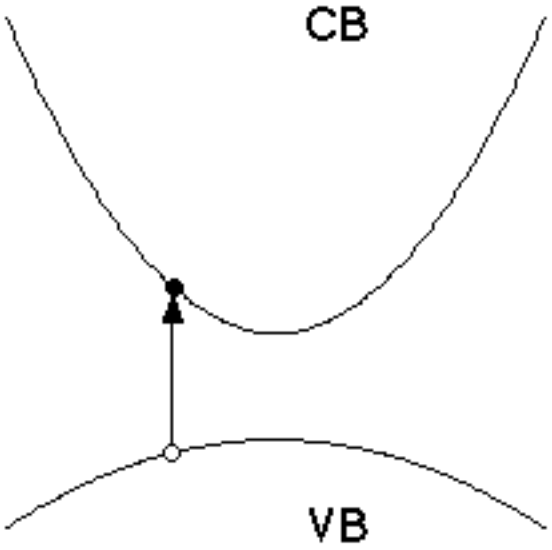


EXCITATION



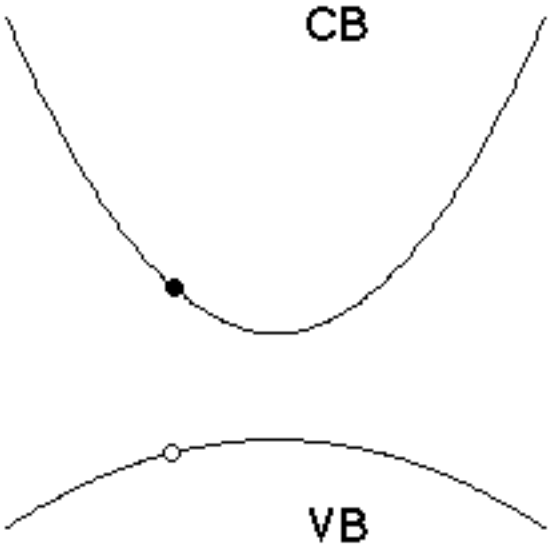
photon excites valence electron

EXCITATION

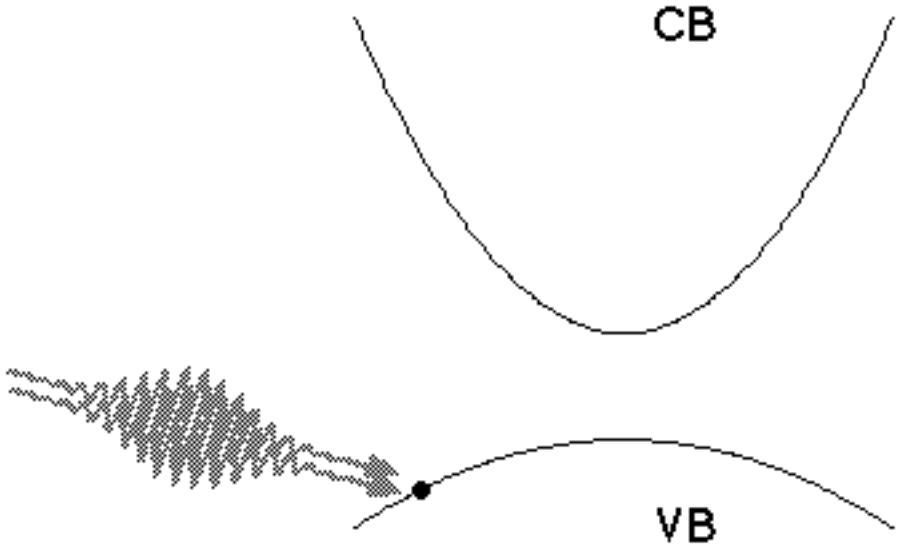


creating an electron-hole pair

EXCITATION

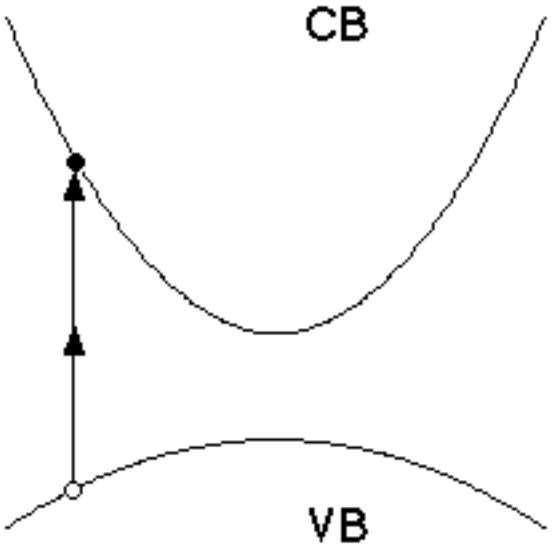


EXCITATION



multiphoton excitation

EXCITATION



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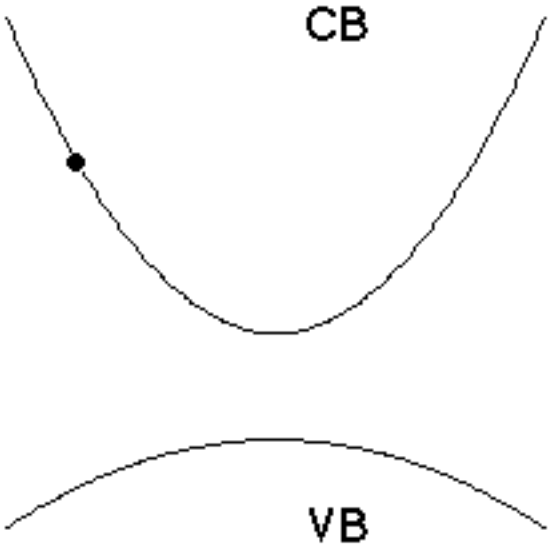
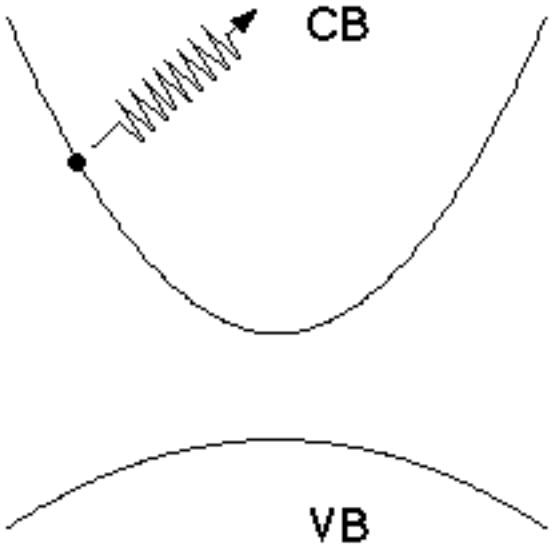
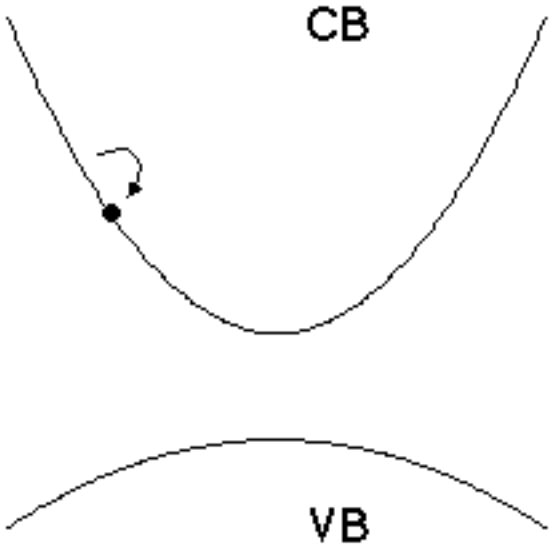


RELAXATION



emission of LO phonons

RELAXATION



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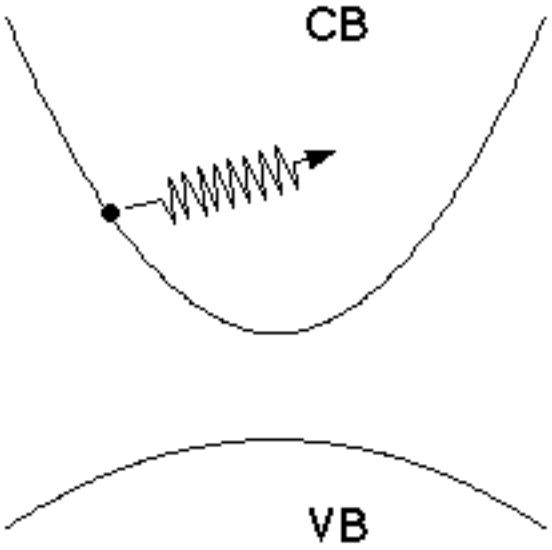

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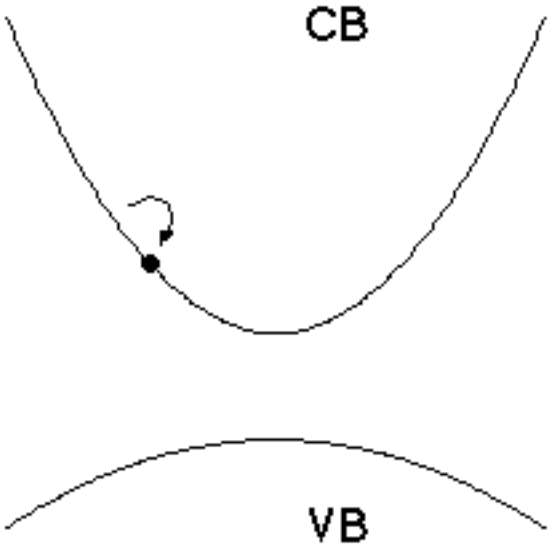

emission of LO phonons

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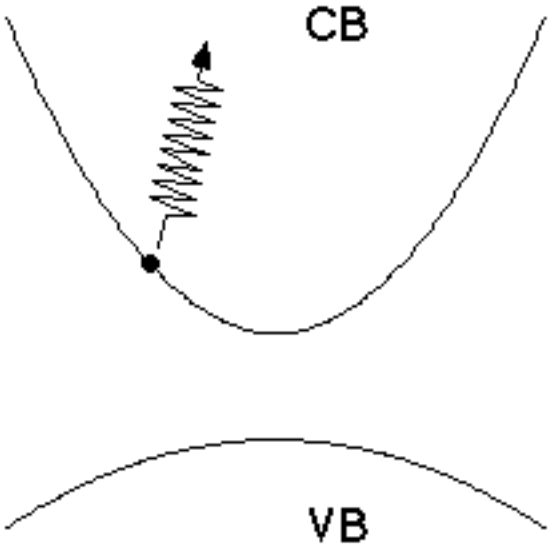

emission of LO phonons

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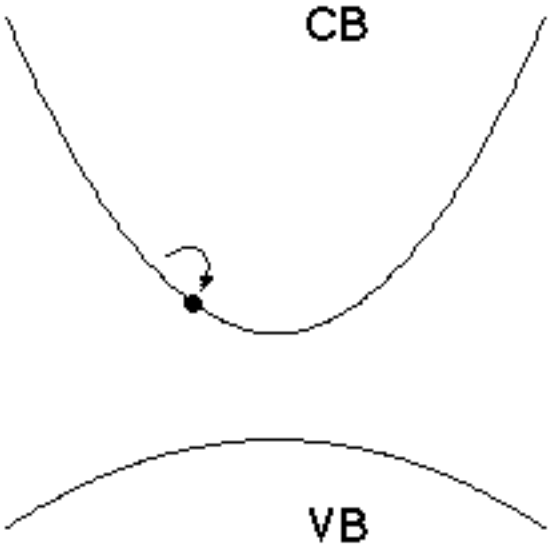

emission of LO phonons

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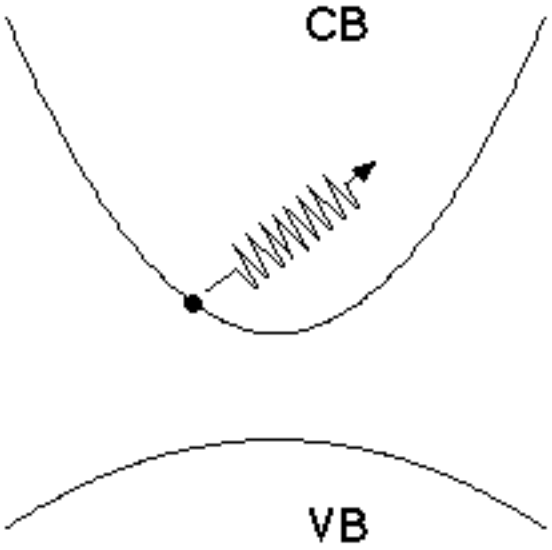

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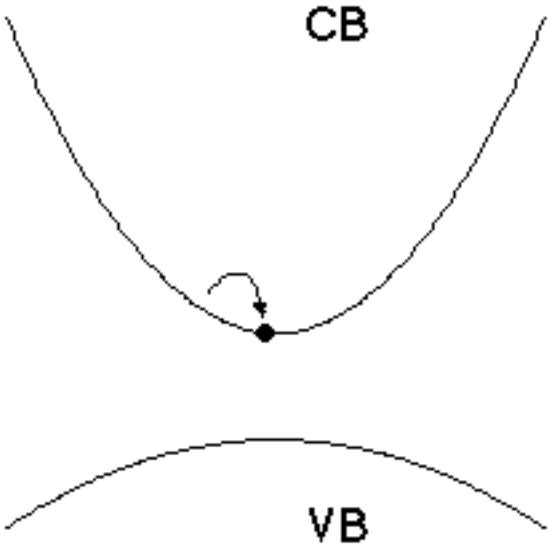

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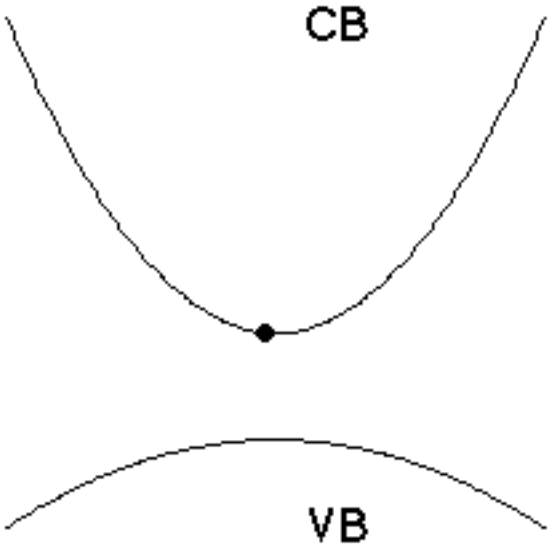
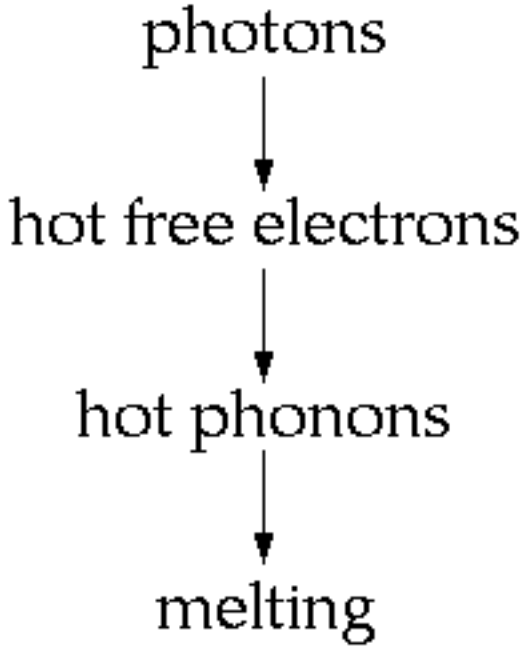


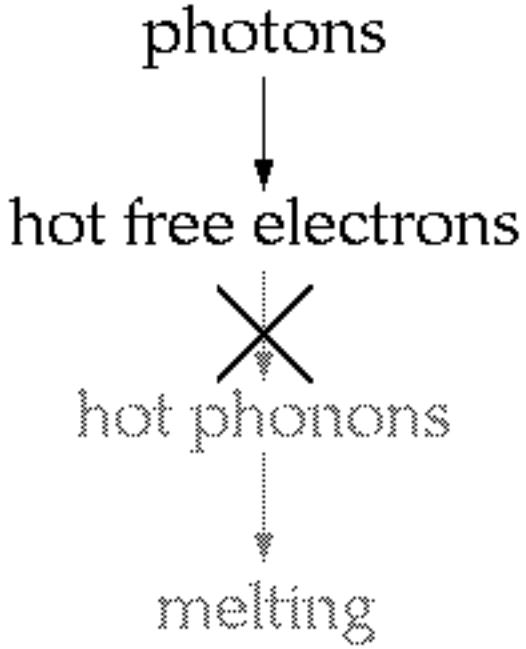
TIME SCALE

long pulses (τ_{exc} >> 1 ps): *thermal melting*



TIME SCALE

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



❶ Background

❷ Single frequency work

❸ Broadband work

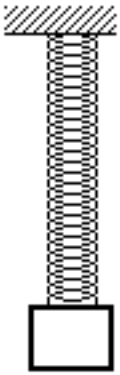
FEMTOSECOND WORK

Phonons	How can we drive them?
Beyond phonons	What happens when we break the limit?

EXCITATION OF PHONONS

absorption	incoherent heating	$\tau_{e\text{-}ph} \approx 2$ ps $>> T_{ph}$
SRS, CARS	coherent driving	$\tau_{exc} \geq 6$ ps $>> T_{ph}$
ISRS	impulsive excitation	$\tau_{exc} \approx 0.1$ ps $\leq T_{ph}$

IMPULSIVE RAMAN SCATTERING



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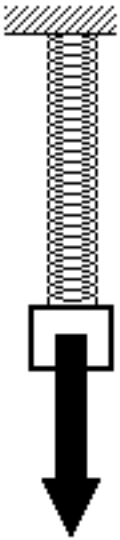

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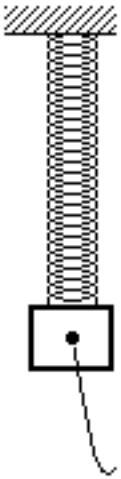

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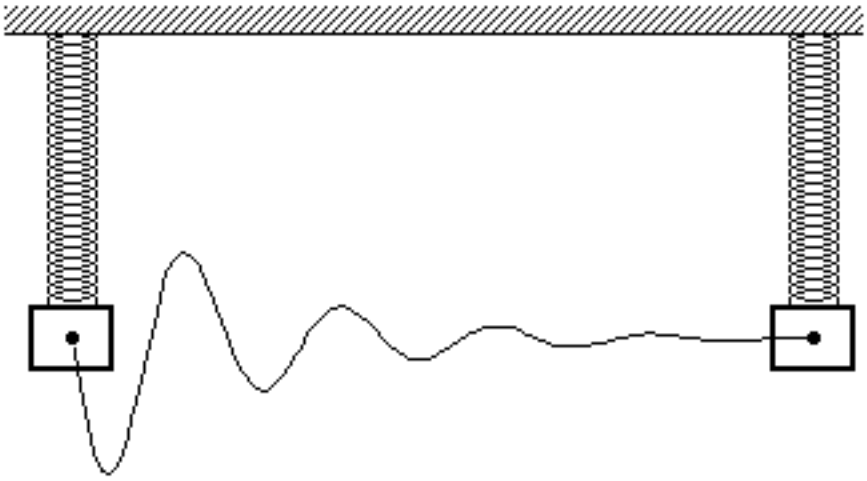

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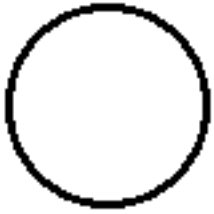

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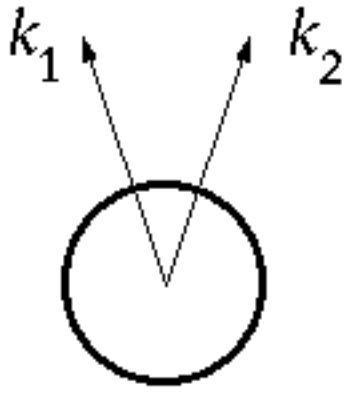

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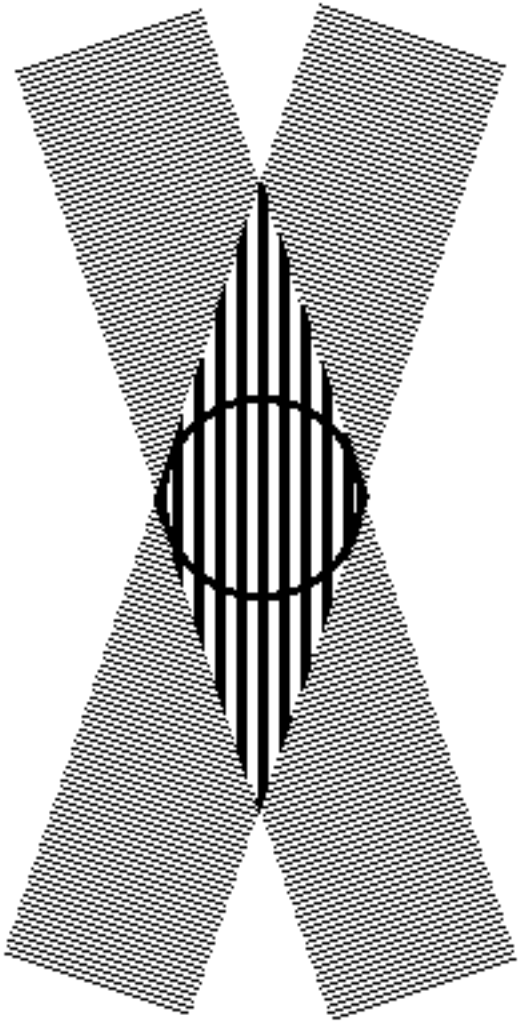

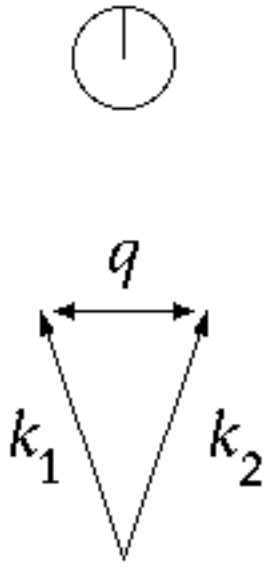

$$k_1 - k_2 = \pm q$$

IMPULSIVE RAMAN SCATTERING

create decaying standing wave...

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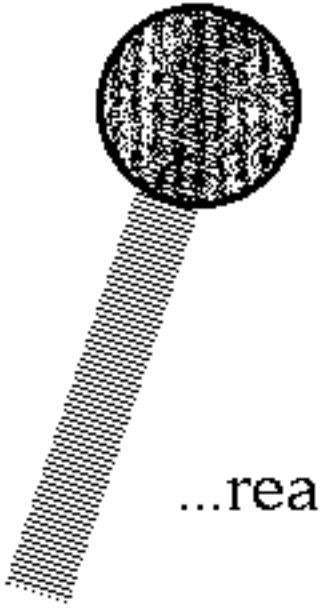
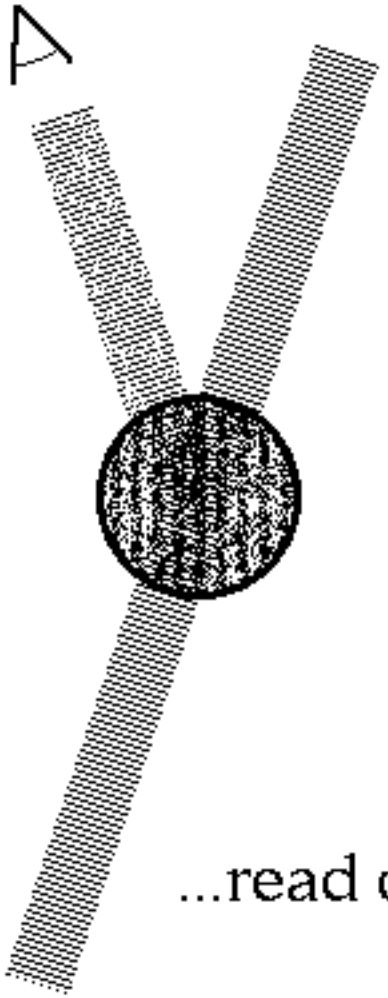


...read out with third beam

IMPULSIVE RAMAN SCATTERING



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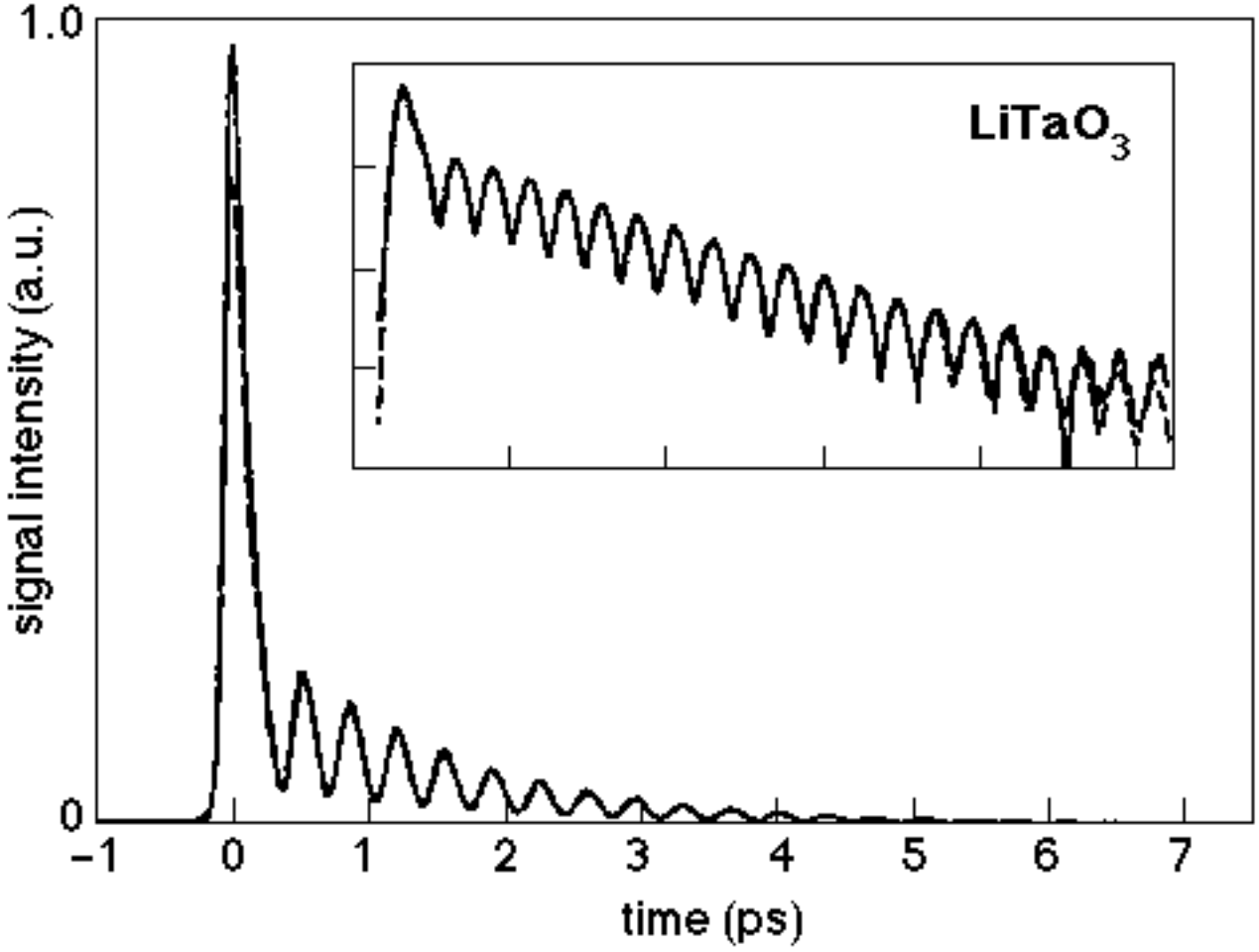
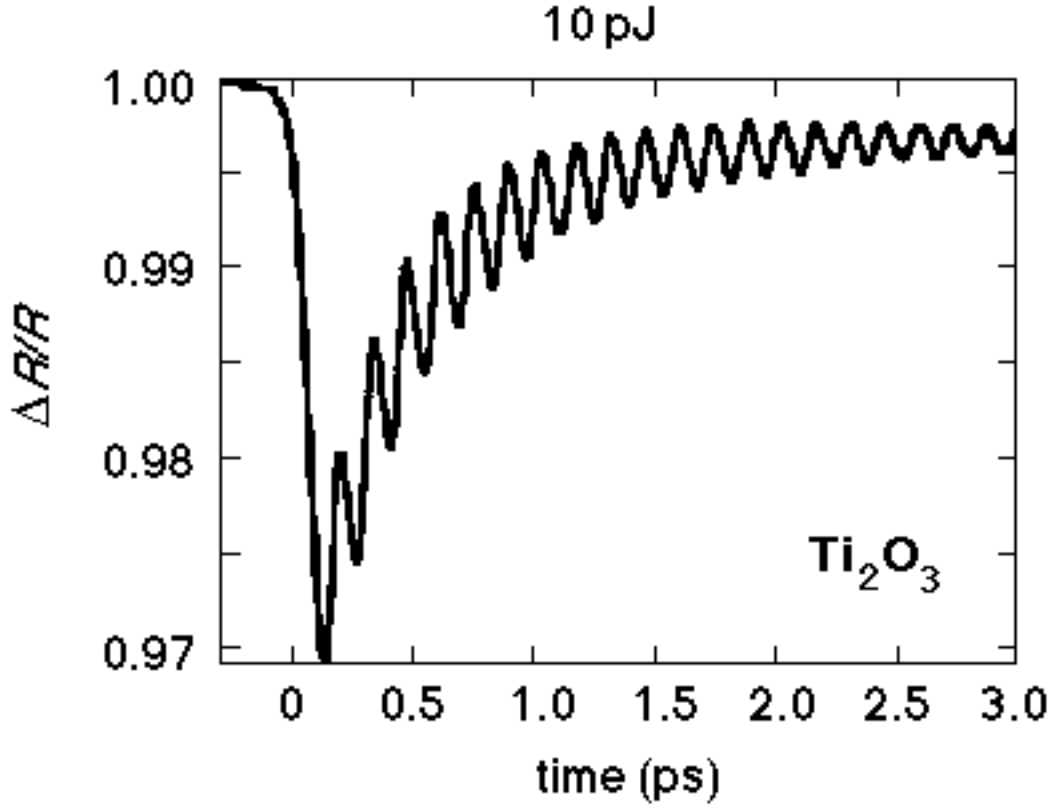


L. Dhar *et al.*, *Chem. Rev.* **94** (1994) 157

LARGE AMPLITUDE OSCILLATIONS



T.K. Cheng *et al., Appl. Phys. Lett.* **57** (1990) 1004
H.J. Zeiger *et al., Phys. Rev. B* **45** (1992) 768

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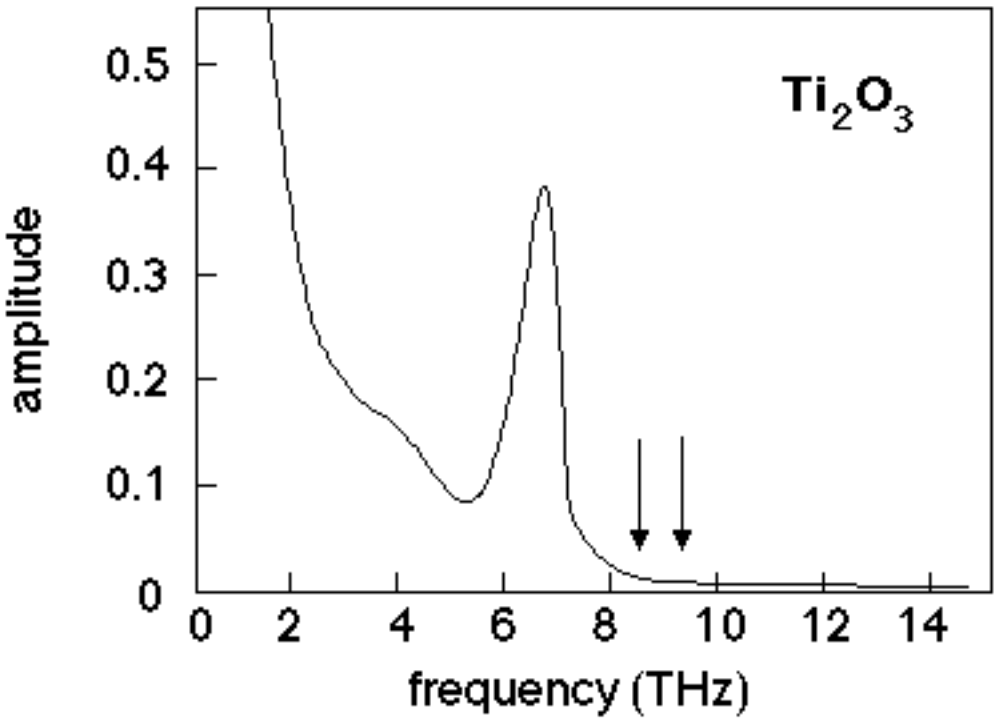

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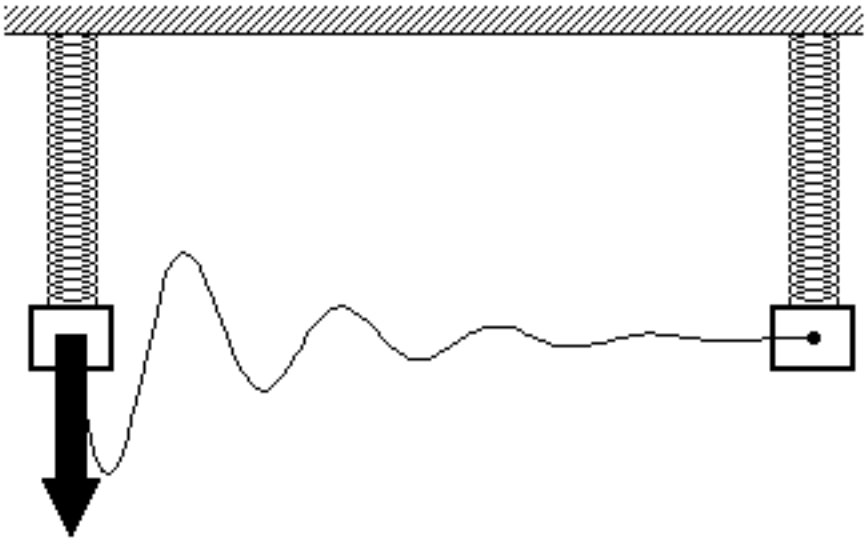
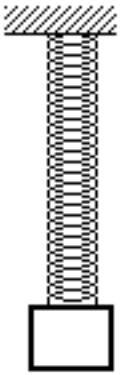


- need to match q
- sinusoidal oscillation

DISPLACIVE EXCITATION



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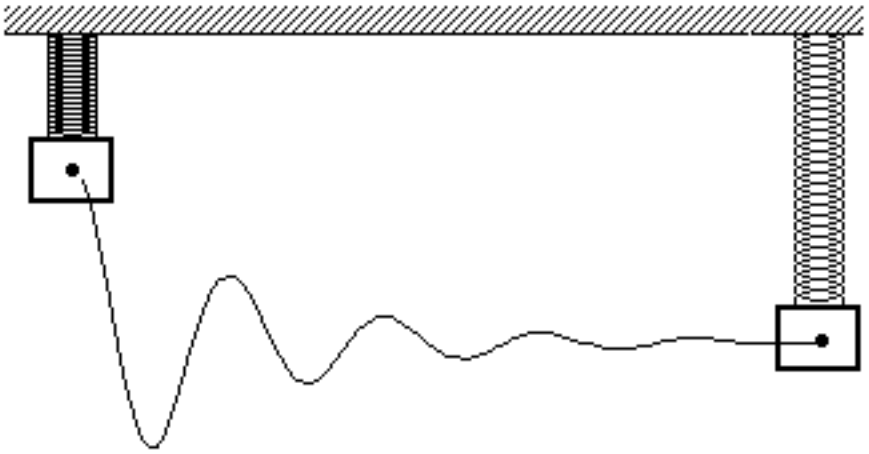
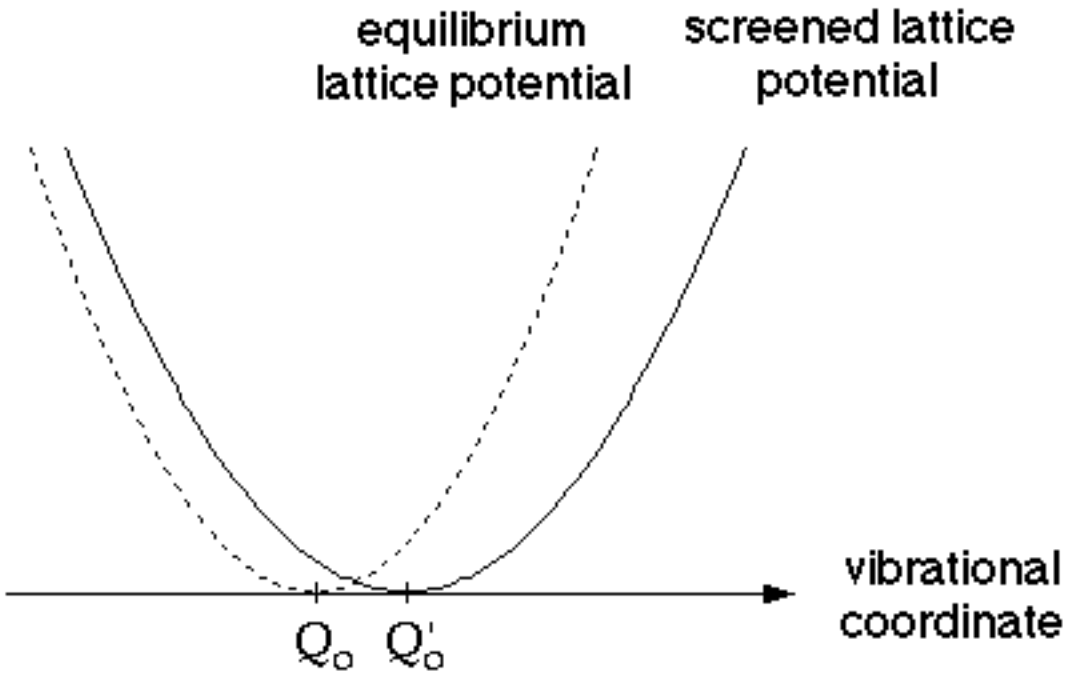
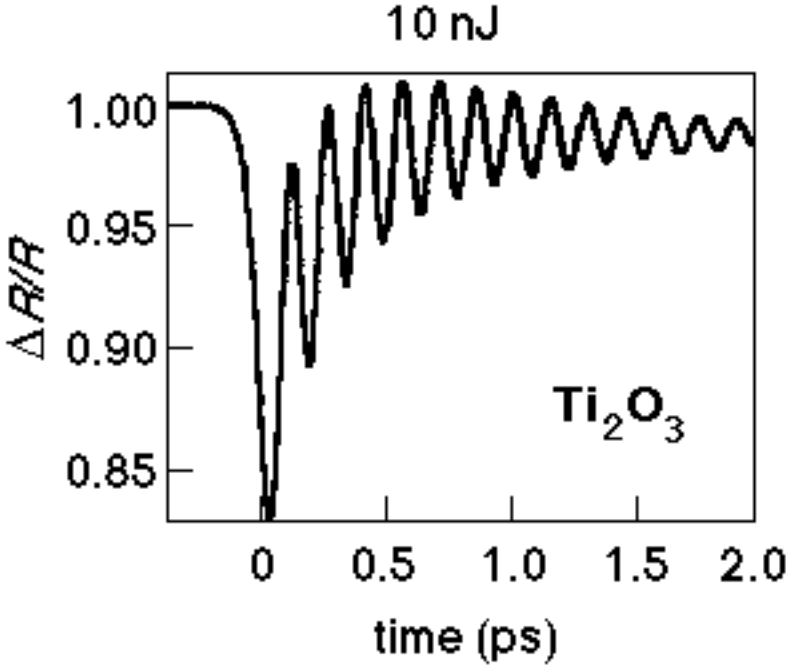
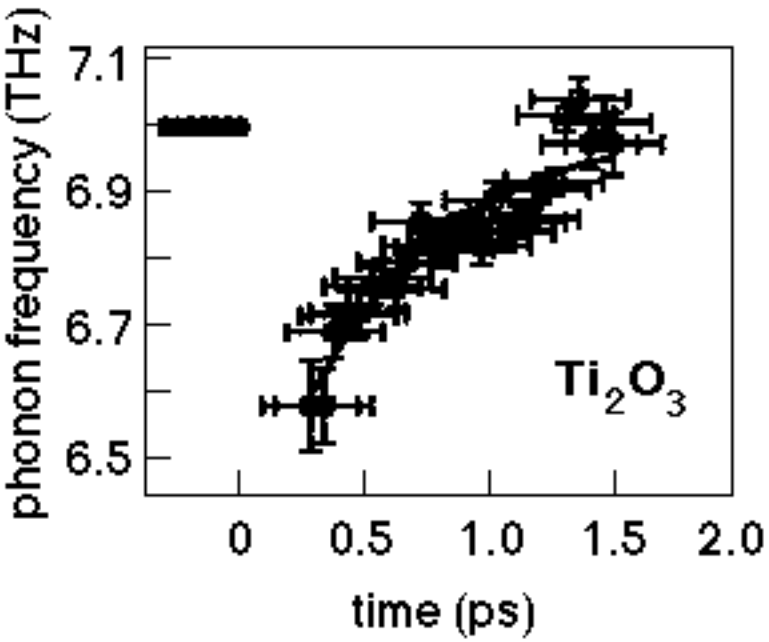


DISPLACIVE EXCITATION



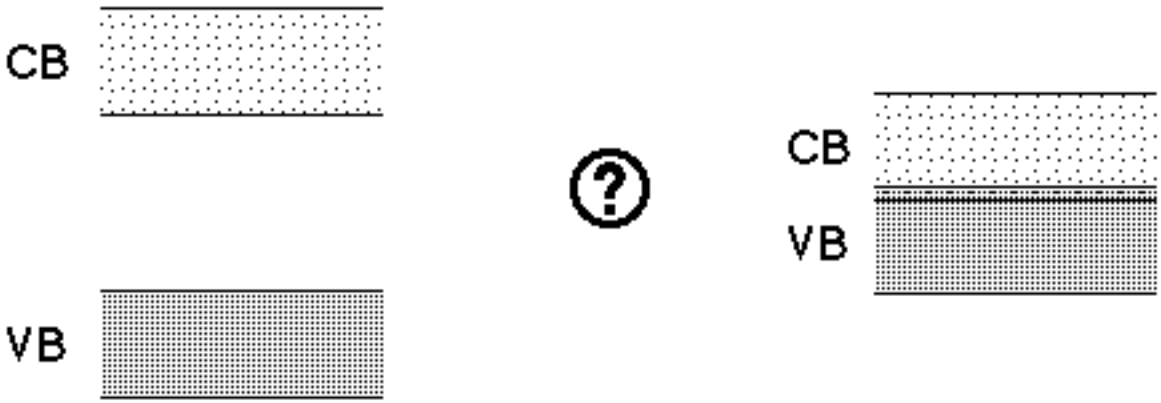
DISPLACIVE EXCITATION





T.K. Cheng *et al., Appl. Phys. Lett.* **62** (1993) 1901

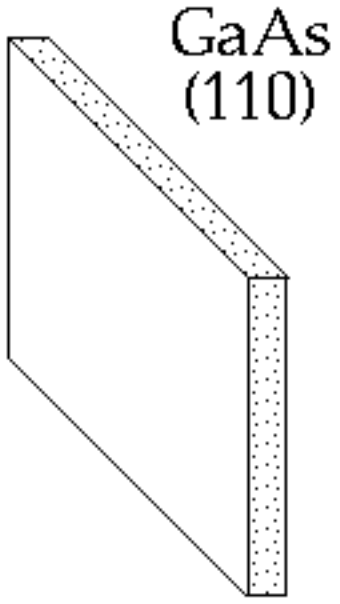
intense pulses can 'melt' GaAs, Si, but how?



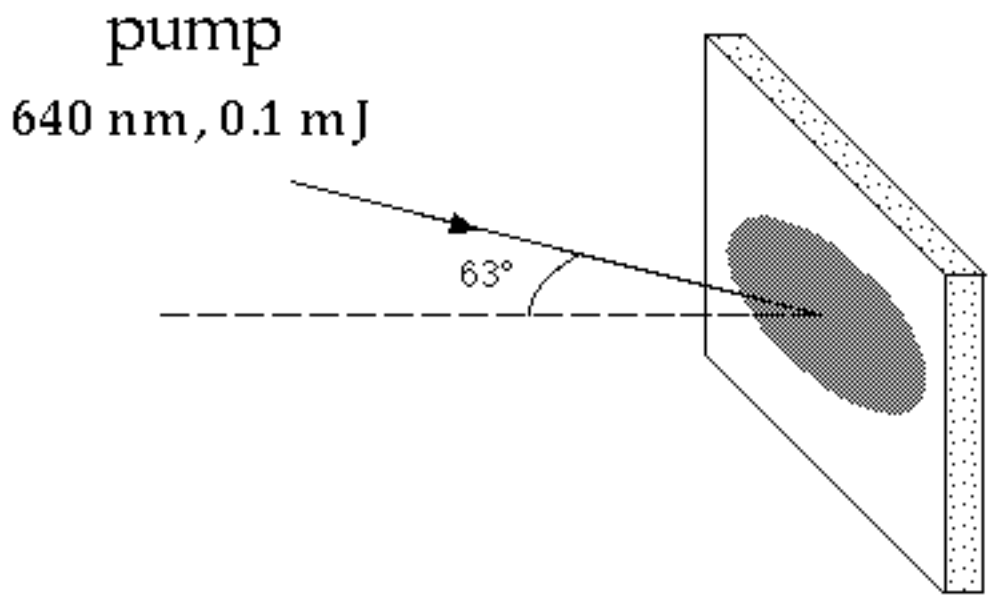
BEYOND PHONONS

intrinsic properties		measured properties
$\varepsilon', \varepsilon''(\omega)$	⟶	R
$\varepsilon', \varepsilon''(\omega)$ $\varepsilon', \varepsilon''(2\omega)$ $\chi_{(2)}$	⟶	SHG

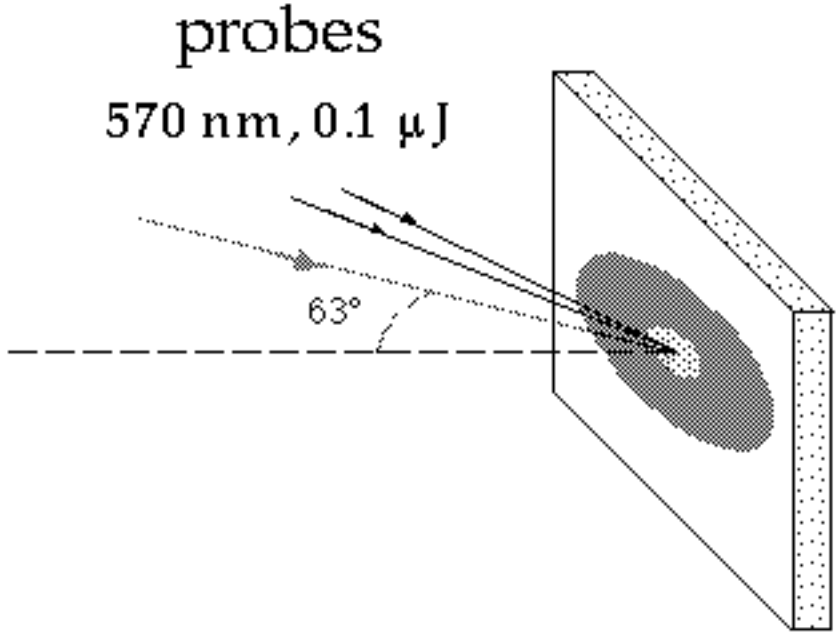
2.2 eV DIELECTRIC CONSTANT



2.2 eV DIELECTRIC CONSTANT

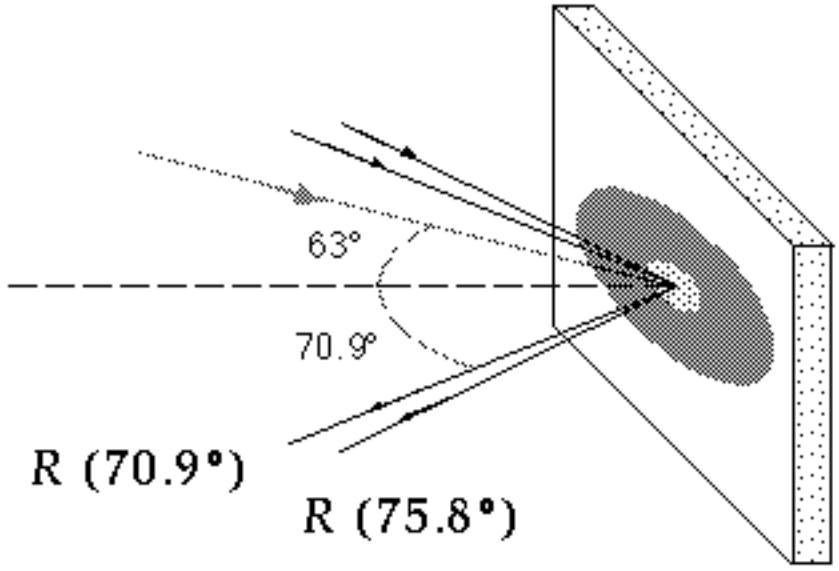


2.2 eV DIELECTRIC CONSTANT

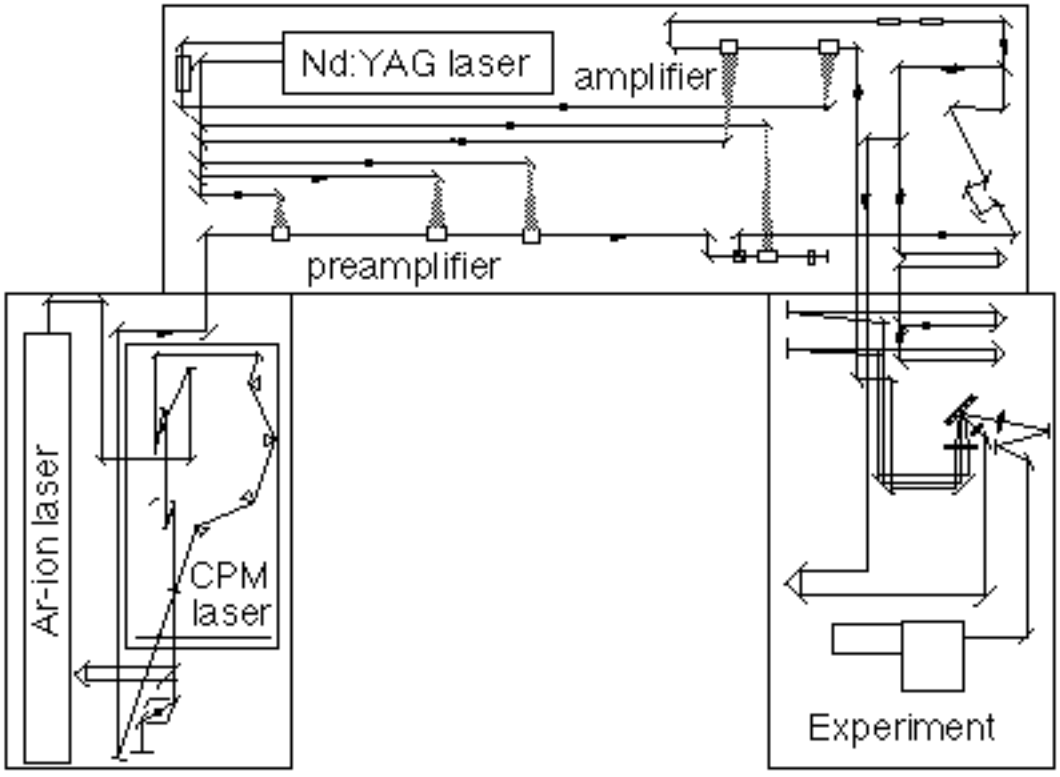


all beams
70 fs, *p*-pol

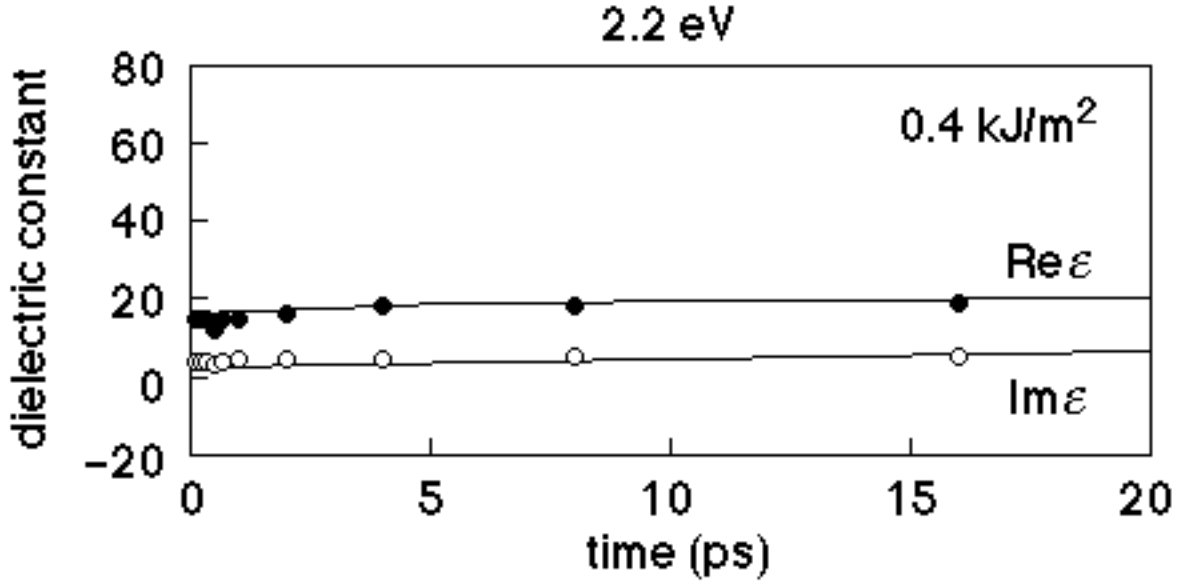
2.2 eV DIELECTRIC CONSTANT



SETUP


Nd:YAG laser
amplifier
preamplifier
Ar-ion laser
CPM
laser
Experiment

TIME DEPENDENCE



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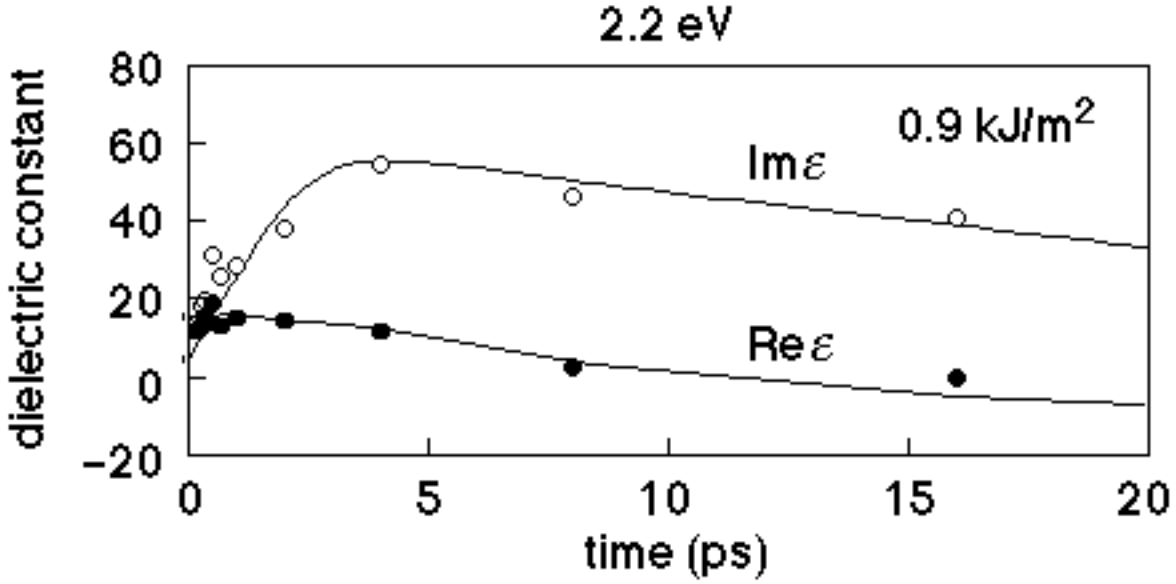
TIME DEPENDENCE



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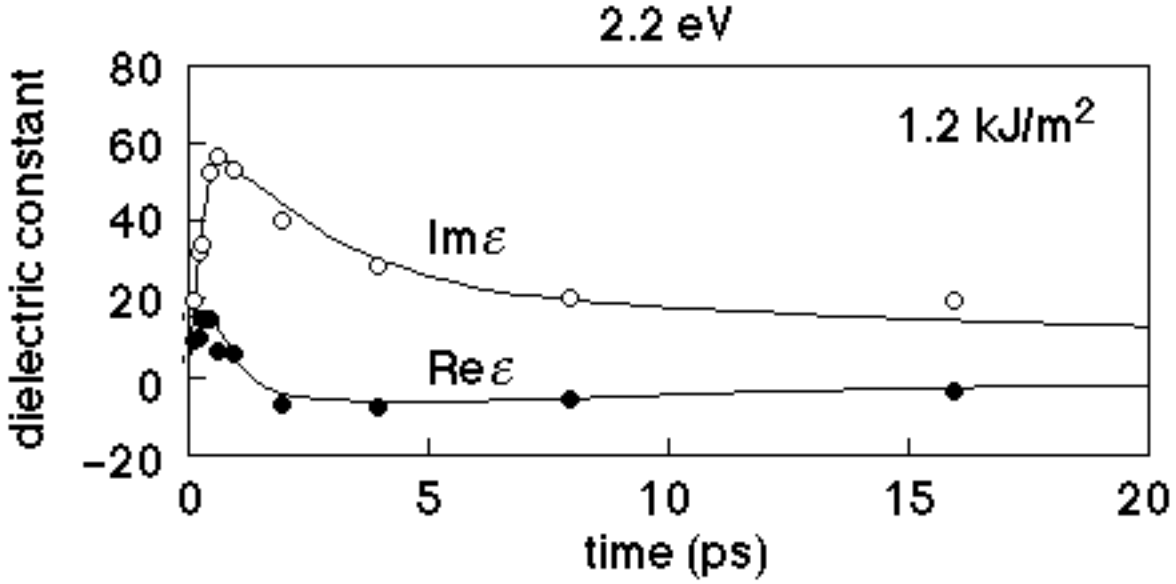
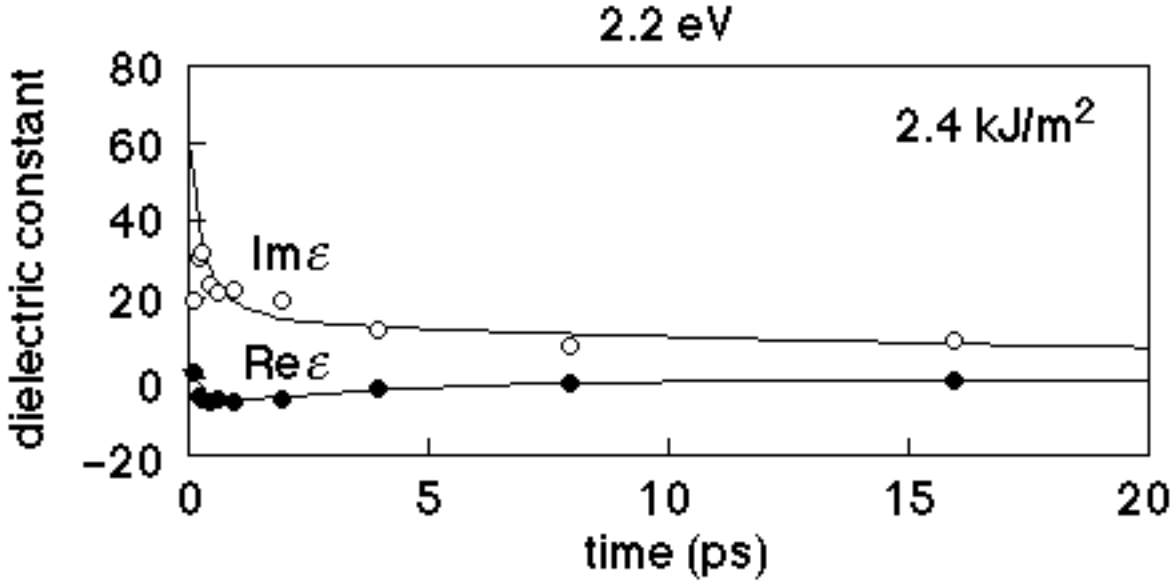


Phys. Rev. B **51** (1995) 6959

TIME DEPENDENCE



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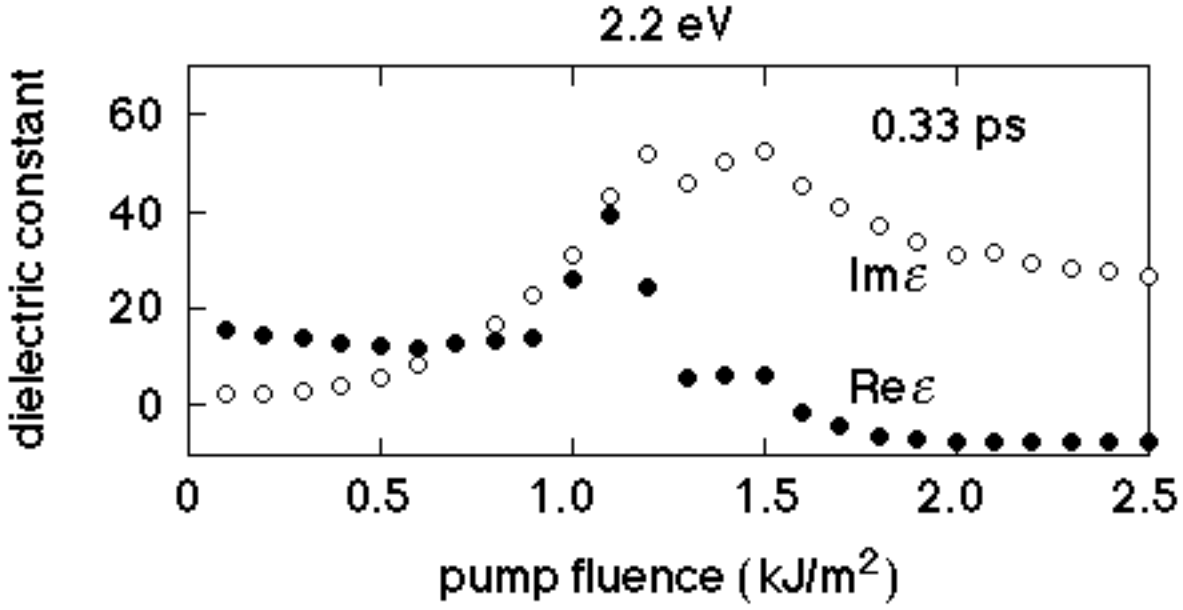
FLUENCE DEPENDENCE



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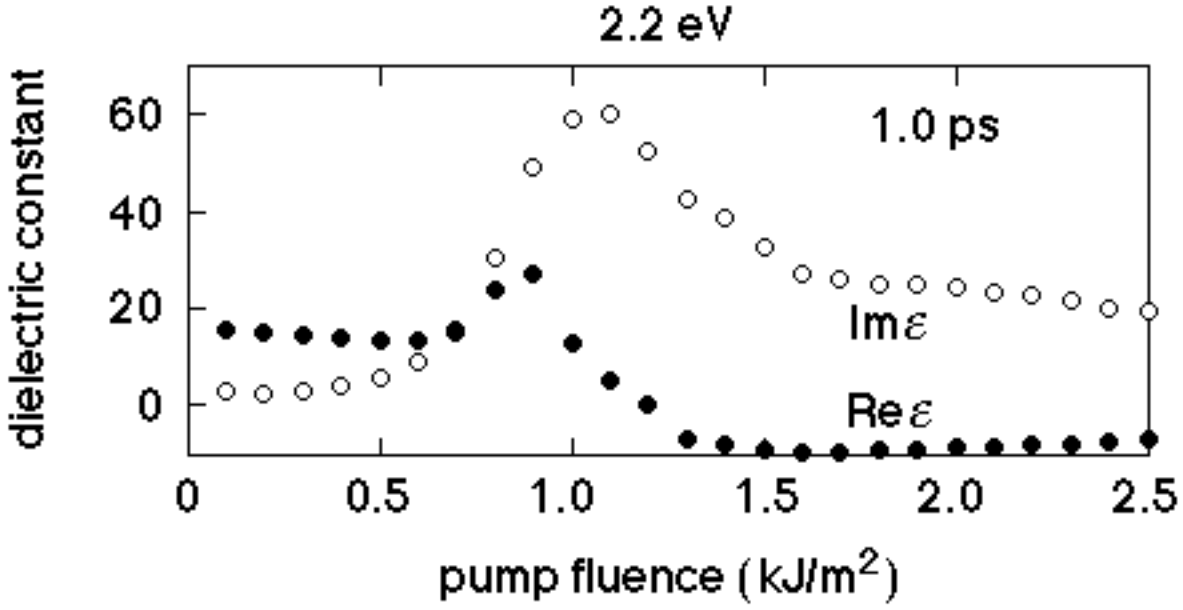

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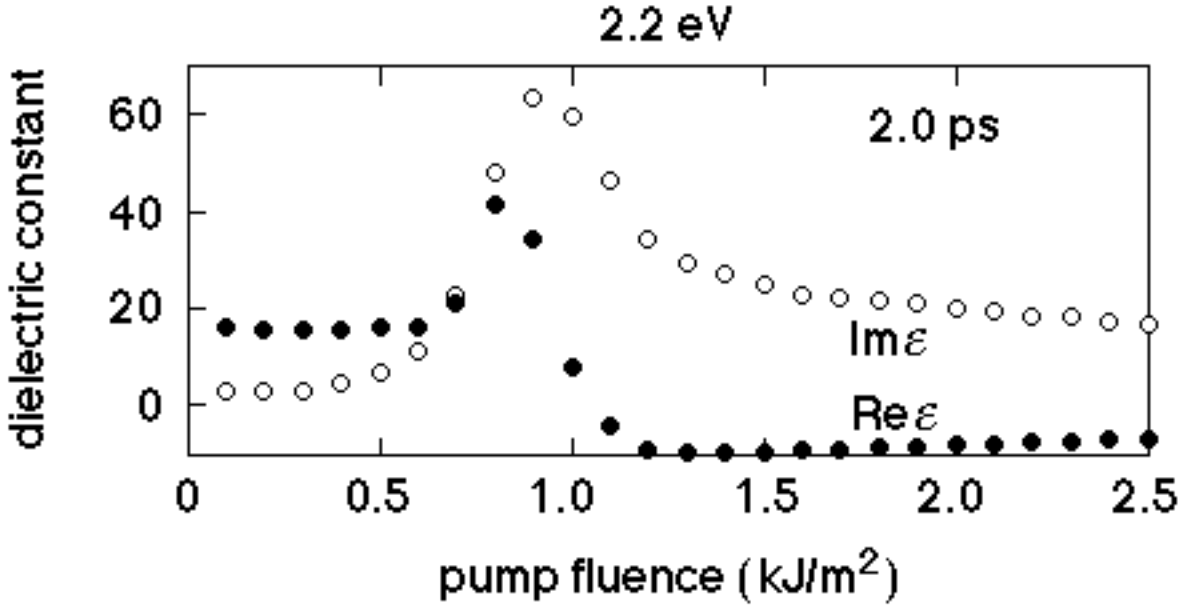

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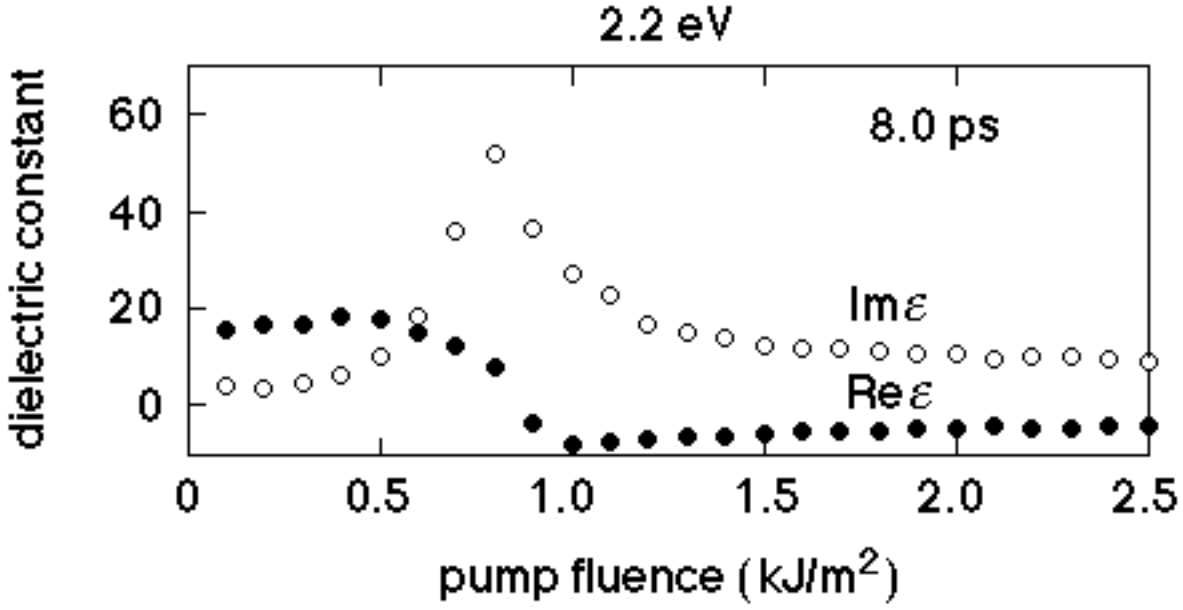

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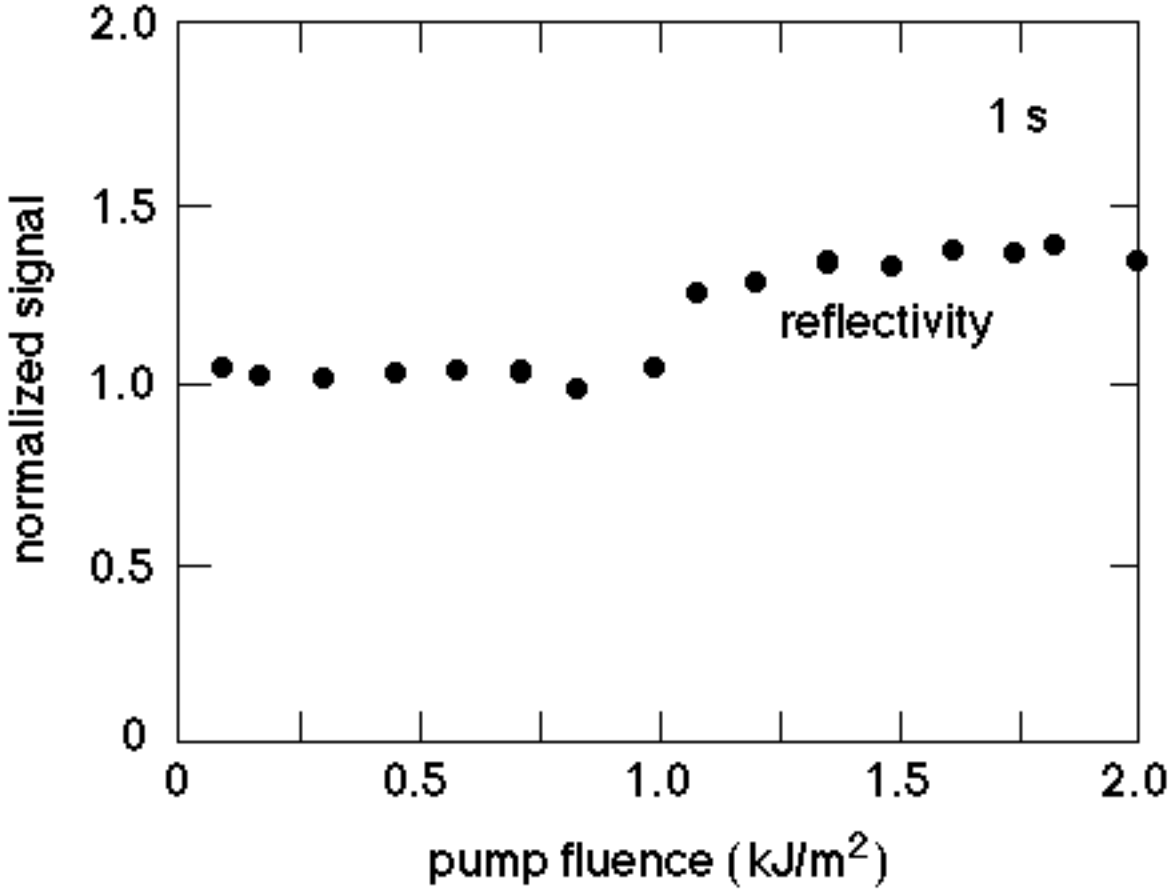
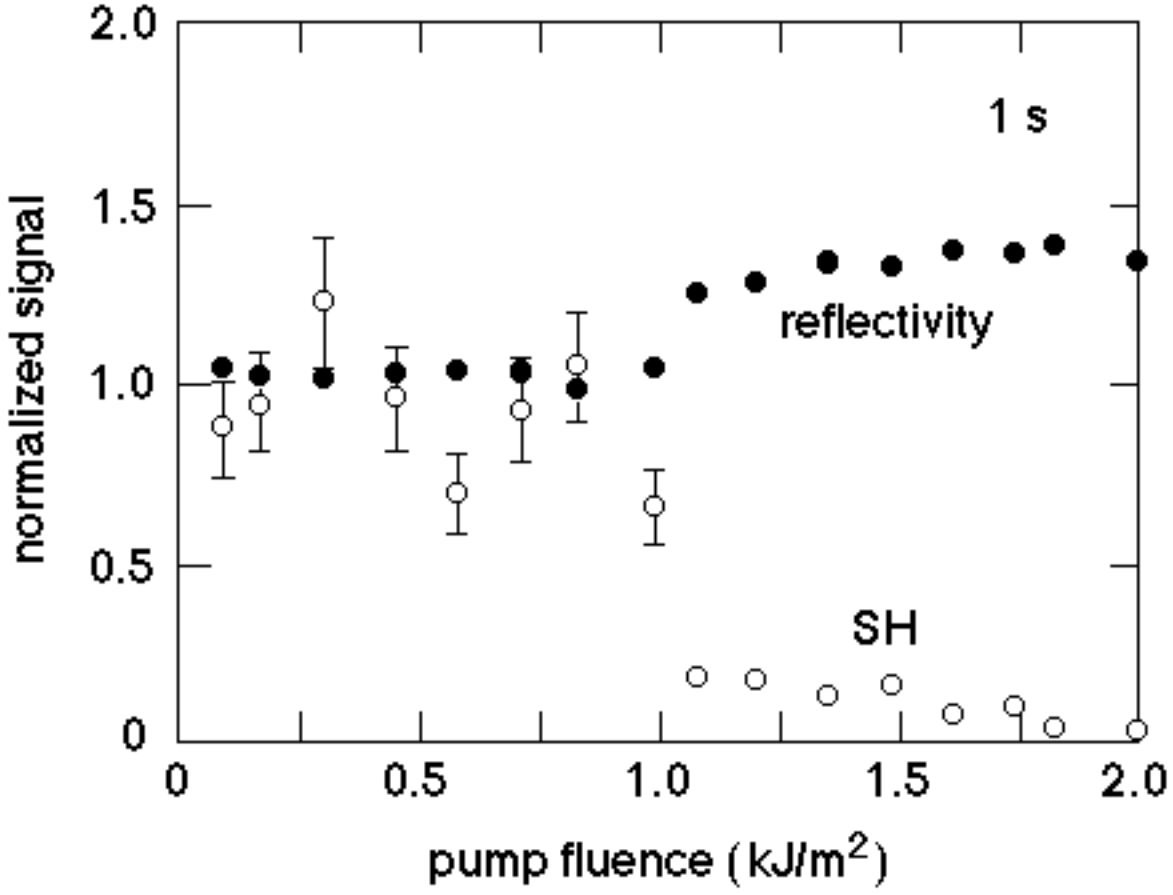
REVERSIBLE vs. IRREVERSIBLE



Phys. Rev. B **51** (1995) 6959

REVERSIBLE vs. IRREVERSIBLE



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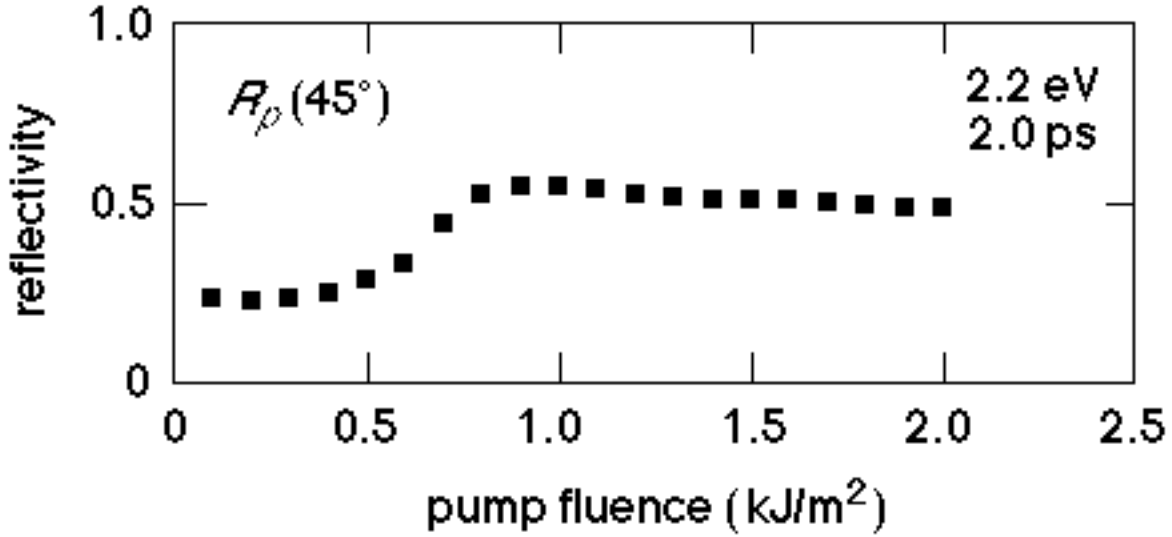
SIMPLE MODEL



Drude model:

$$\mathrm{Re}(\varepsilon) = 1 - \frac{Ne^2}{m\varepsilon_\circ}\frac{1}{\omega^2+\gamma^2} \qquad \mathrm{Im}(\varepsilon) = \frac{Ne^2}{m\varepsilon_\circ}\frac{\gamma\omega}{\omega^4+\gamma^2\omega^2}$$

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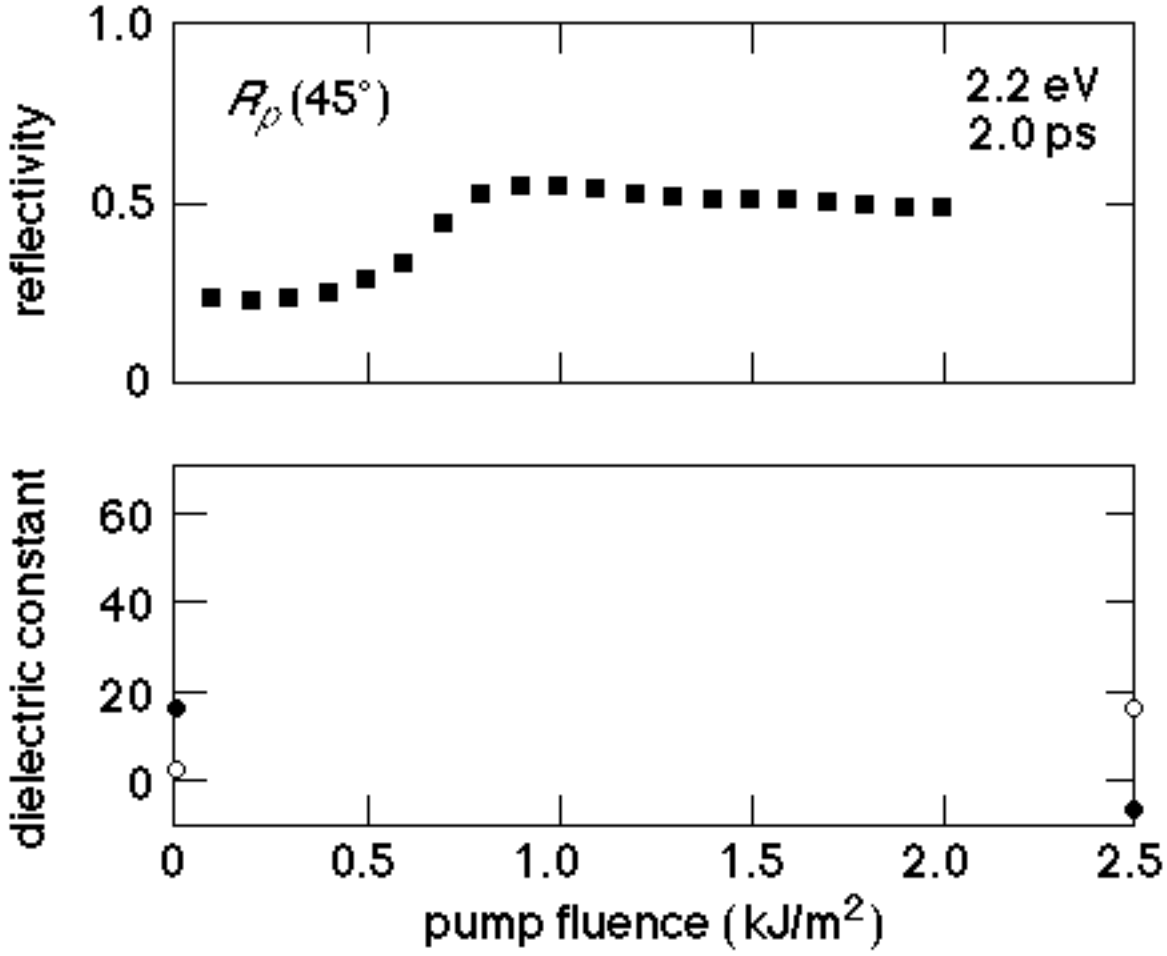
SIMPLE MODEL



Annu. Rev. Mater. Sci. **25** (1995) 223

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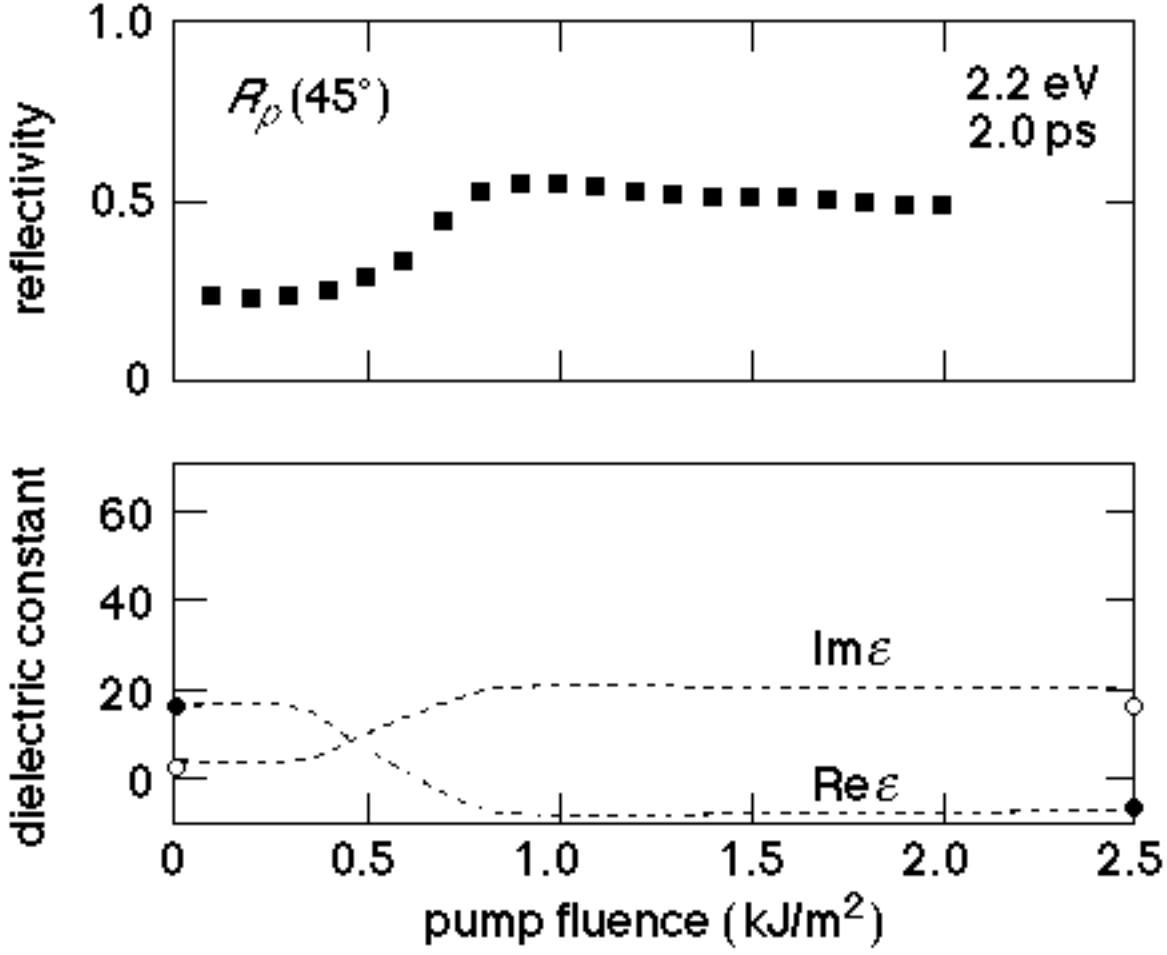

Annu. Rev. Mater. Sci. **25** (1995) 223

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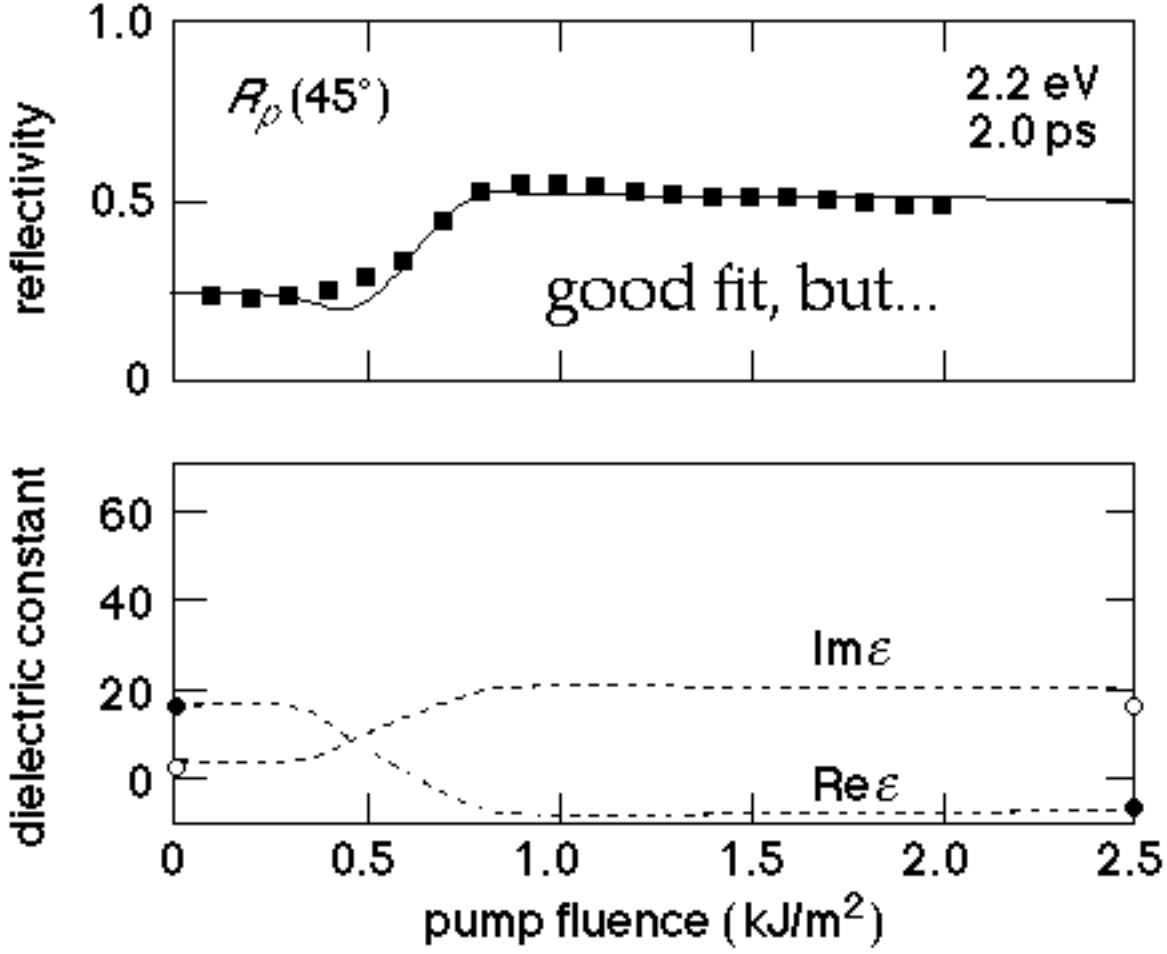

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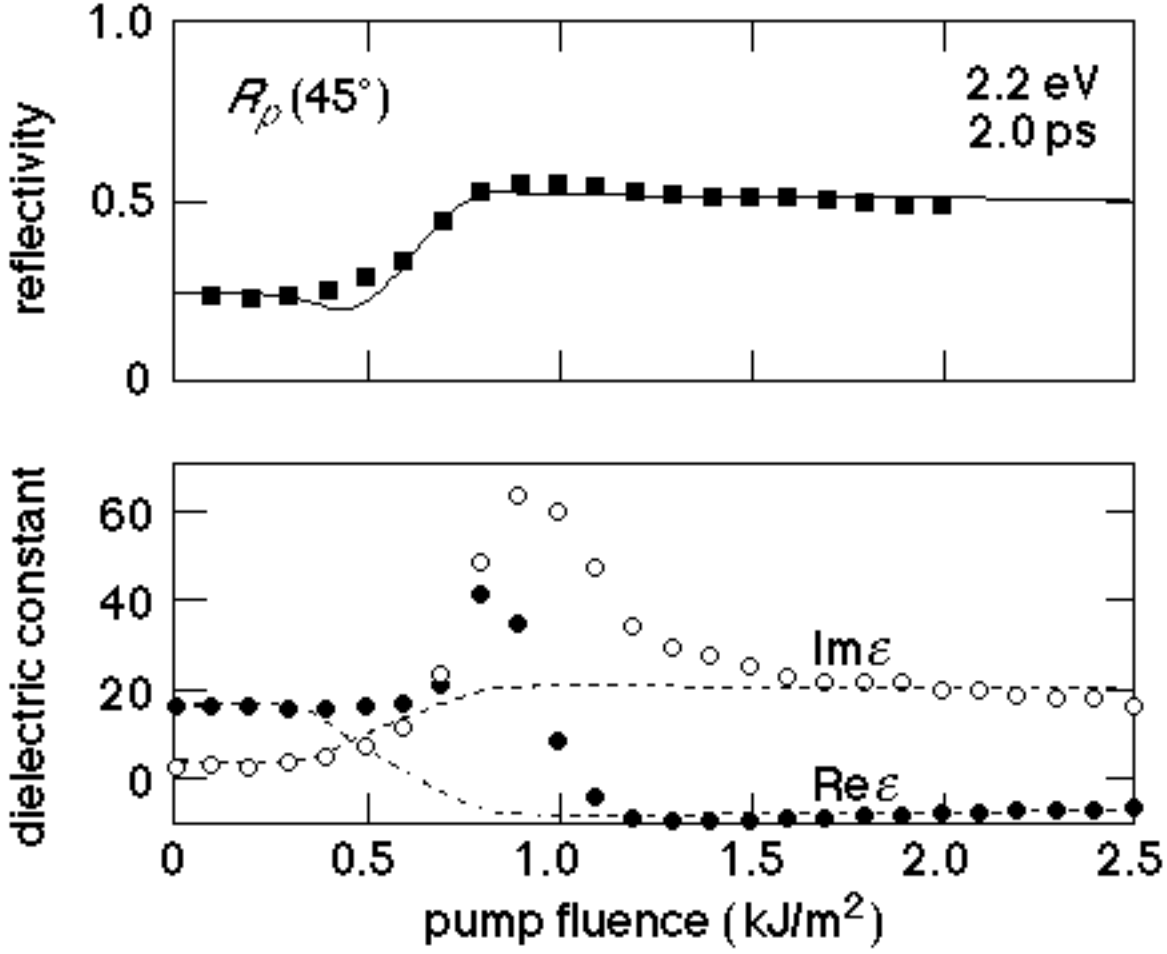

Annu. Rev. Mater. Sci. **25** (1995) 223

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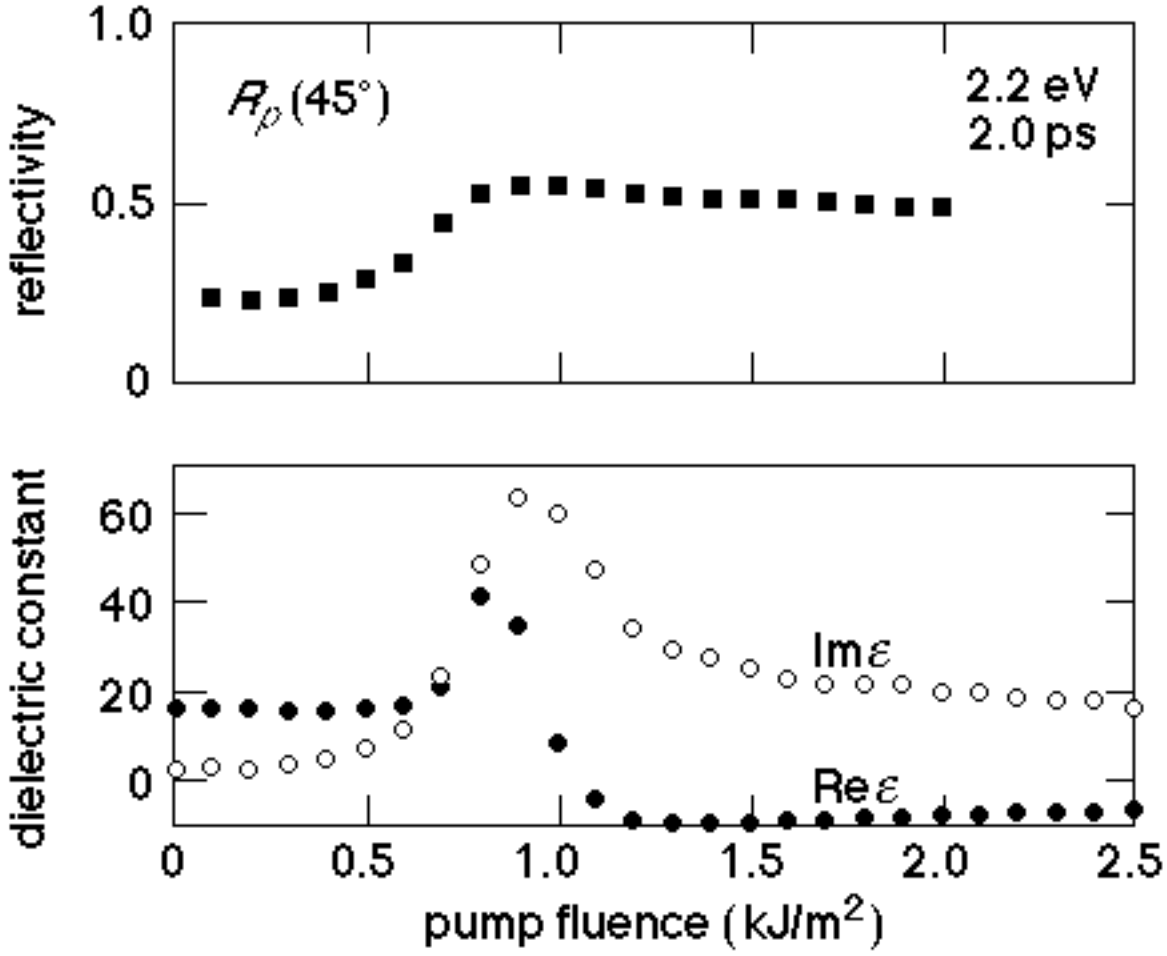

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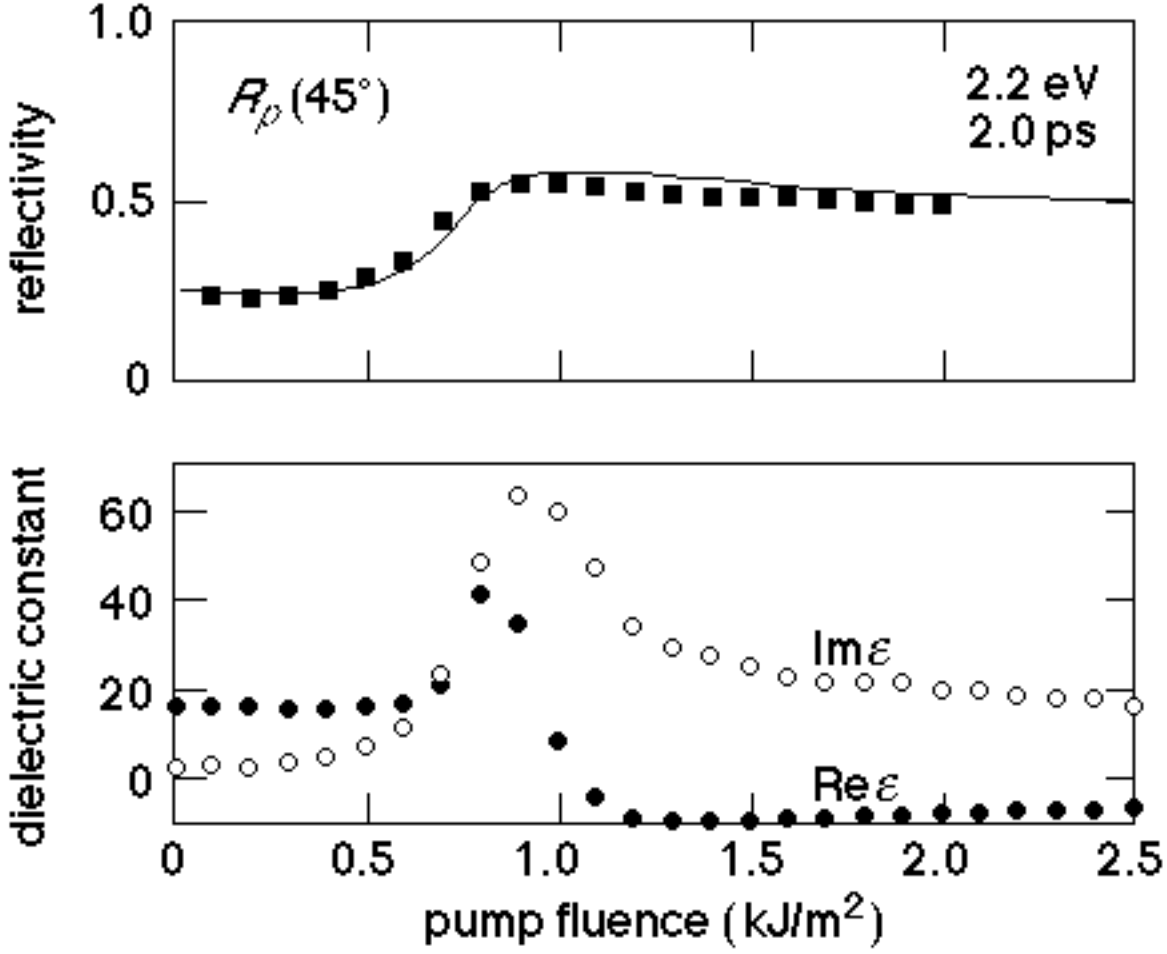
SIMPLE MODEL



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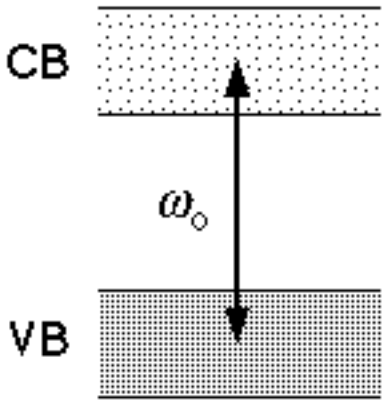
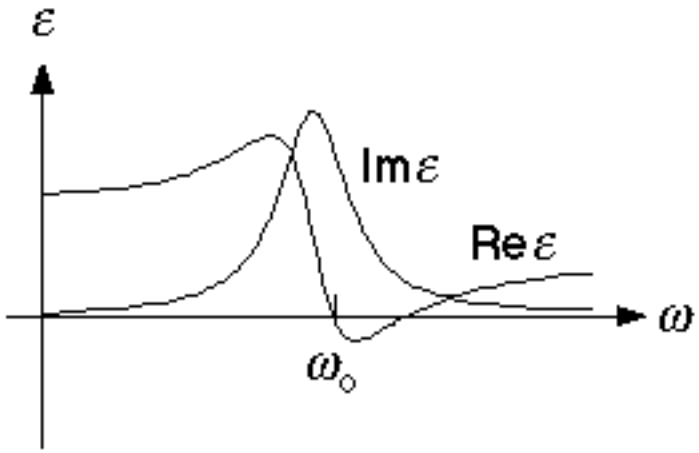
WHAT IS HAPPENING?

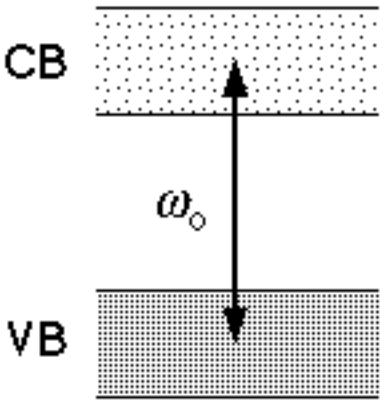


semiconductor

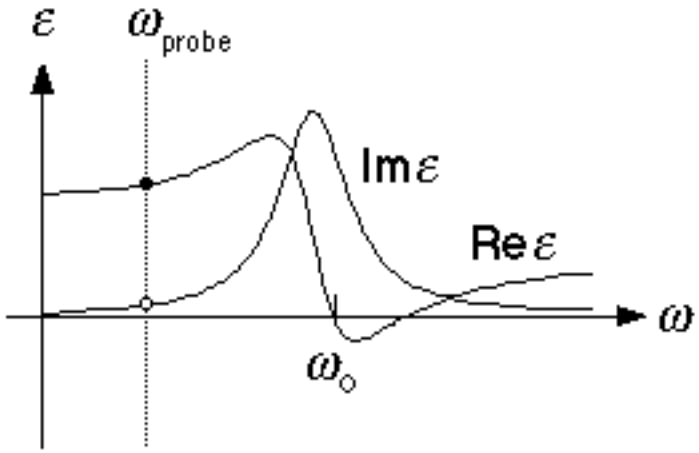


$\omega_o \approx 4.5$ eV

WHAT IS HAPPENING?

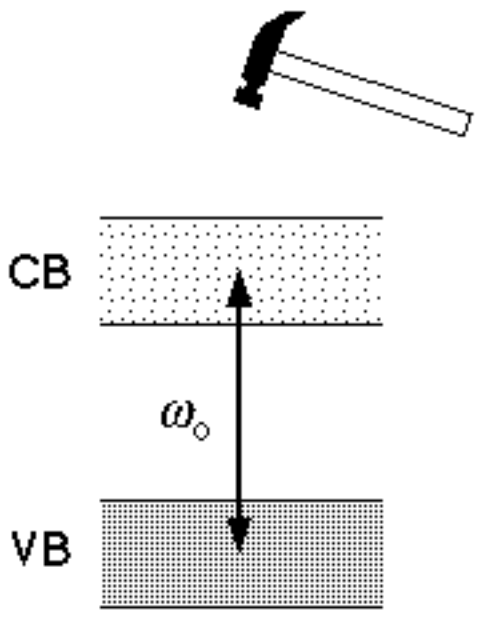


semiconductor

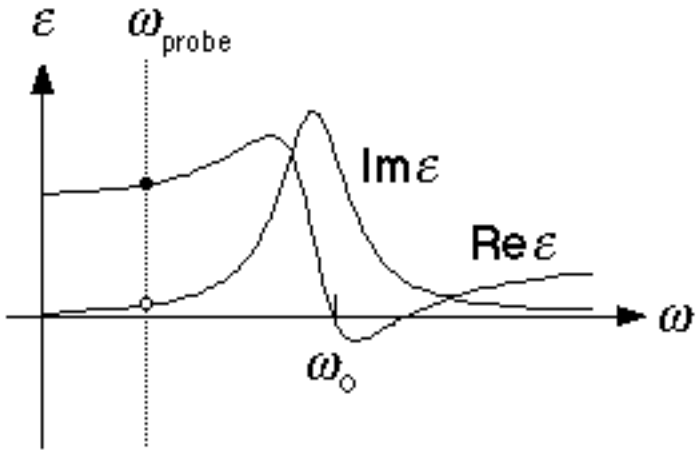


$\omega_{probe} = 2.2$ eV

WHAT IS HAPPENING?



semiconductor



pump pulse excites valence electrons...

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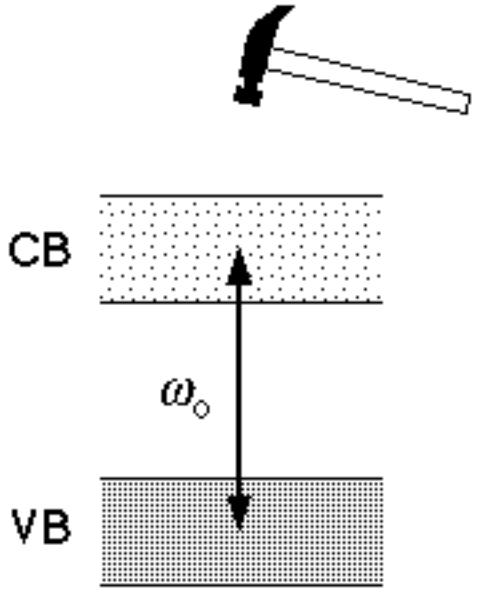
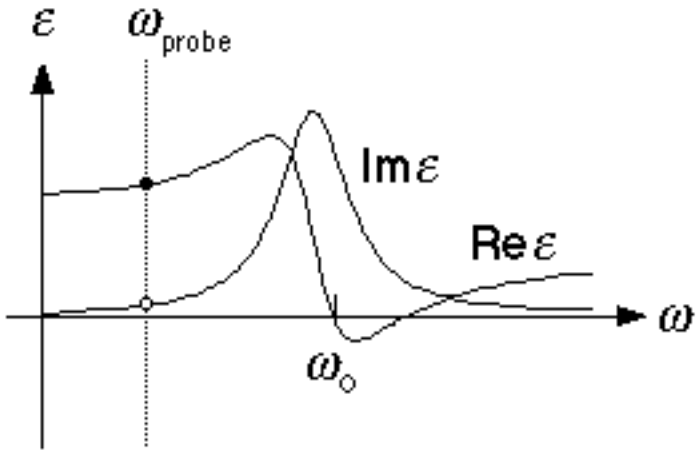


semiconductor



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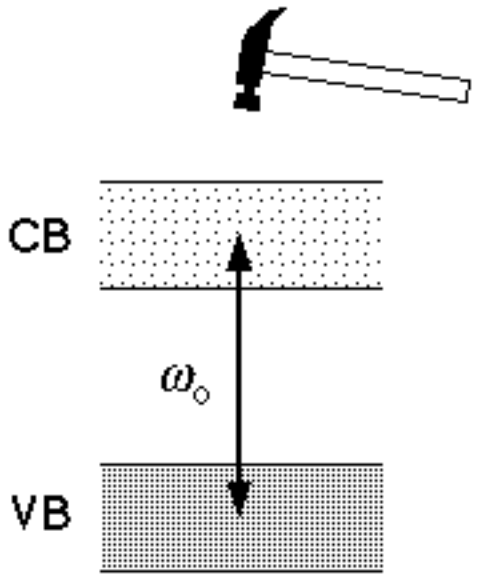
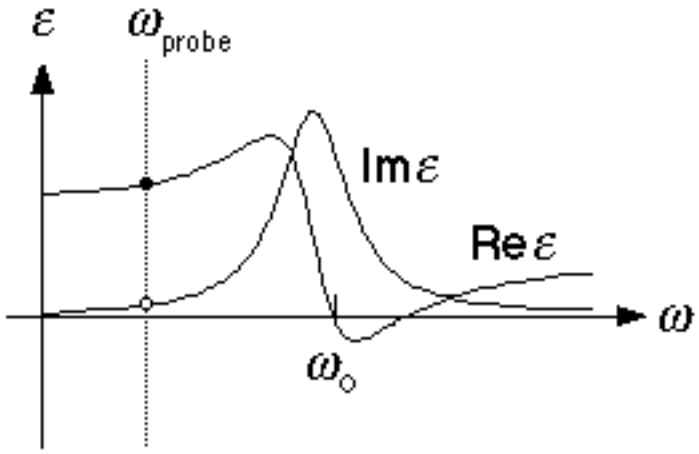


semiconductor



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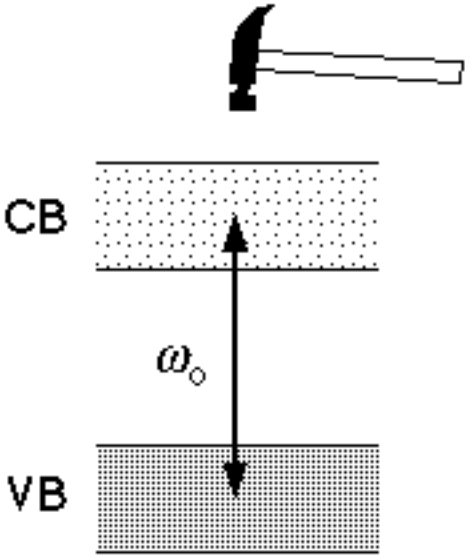
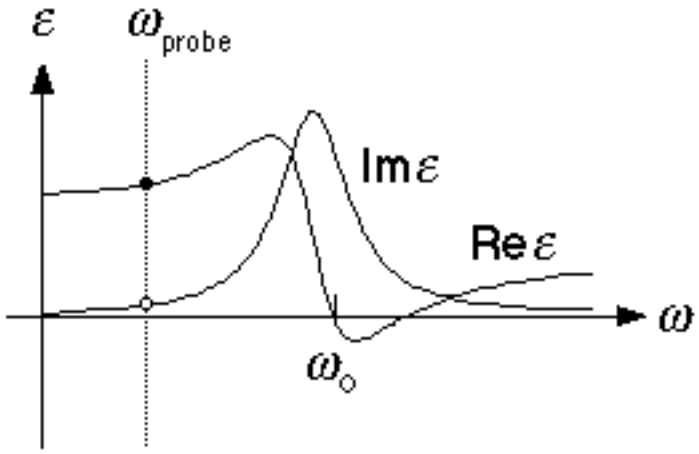


semiconductor



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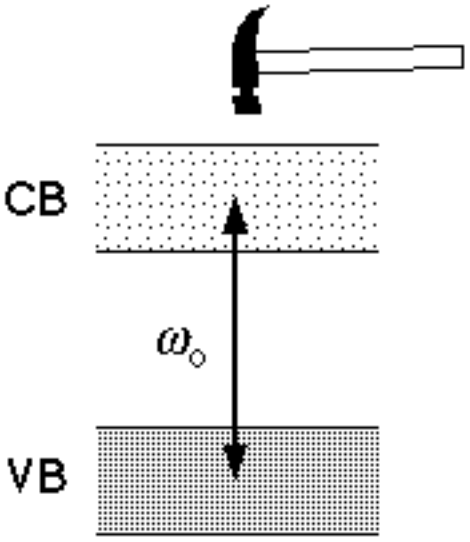

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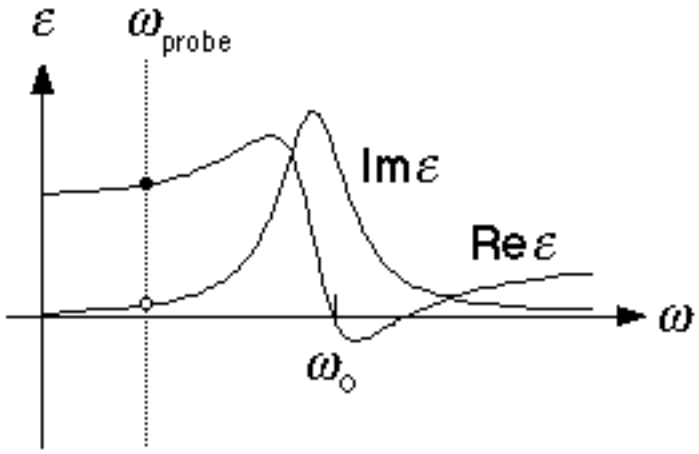

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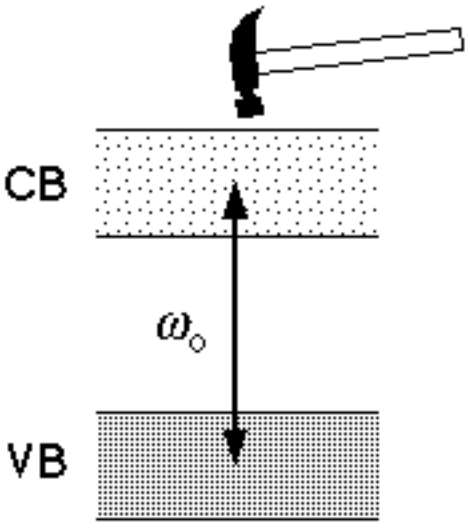
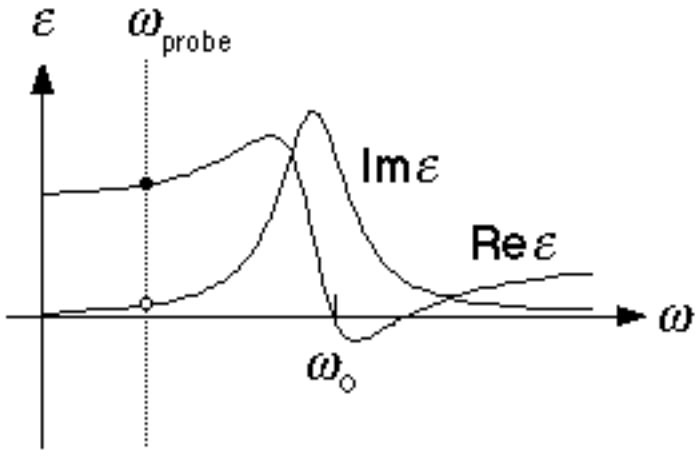


semiconductor



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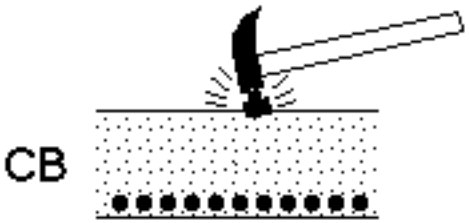

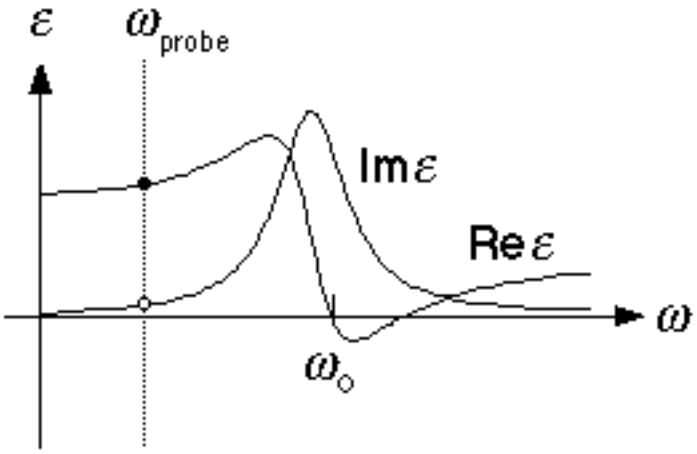

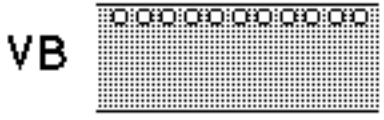

semiconductor

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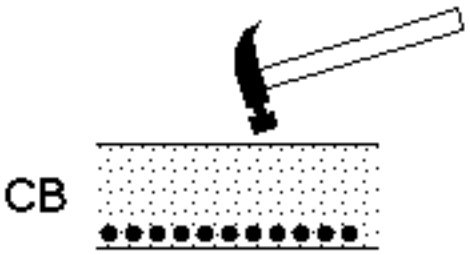
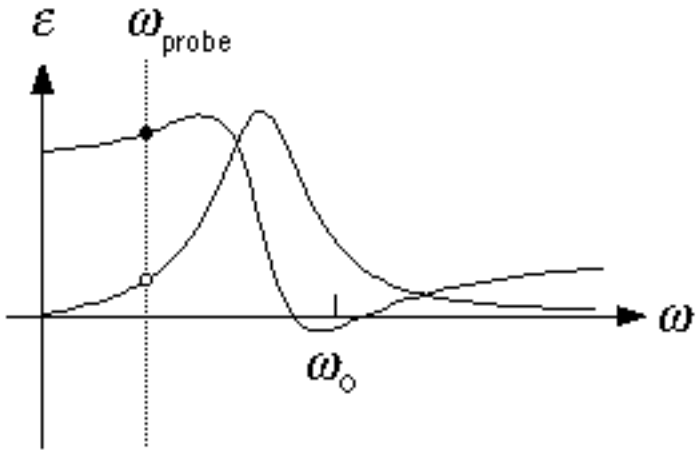
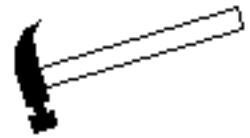


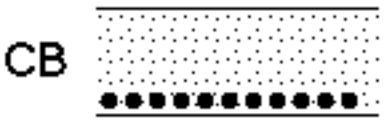
semiconductor



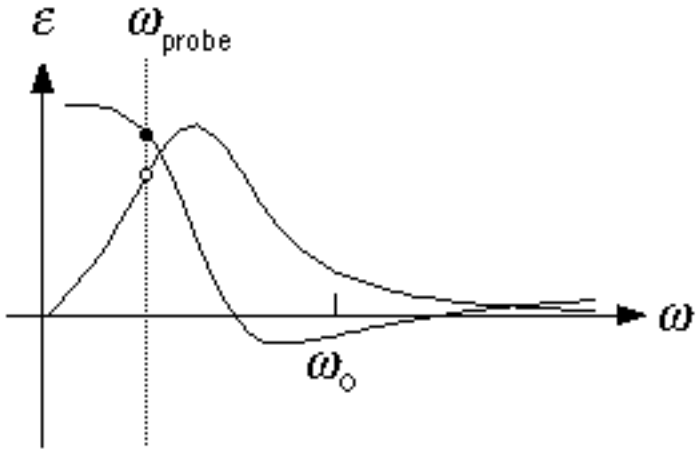
...and induces change in bandstructure

WHAT IS HAPPENING?



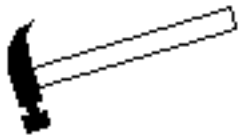


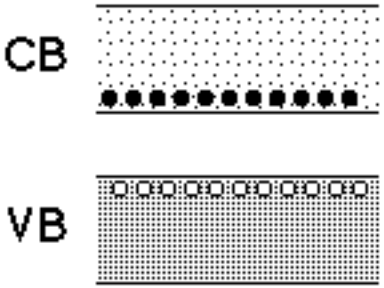
semiconductor



...and induces change in bandstructure

WHAT IS HAPPENING?





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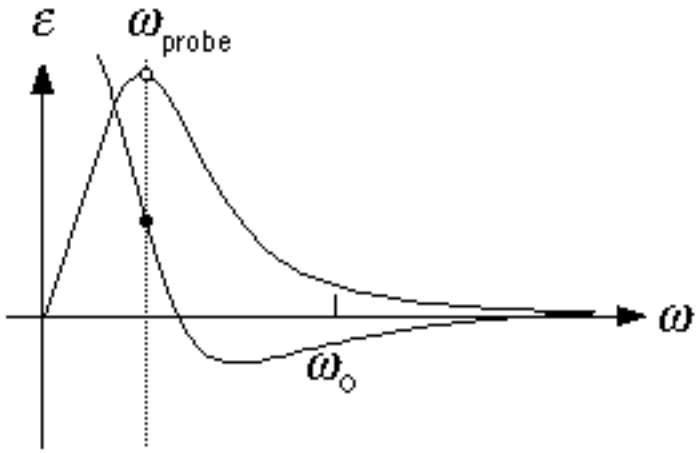

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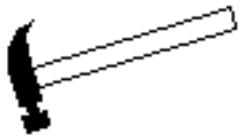

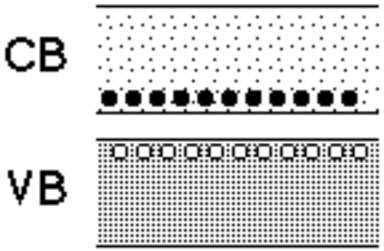

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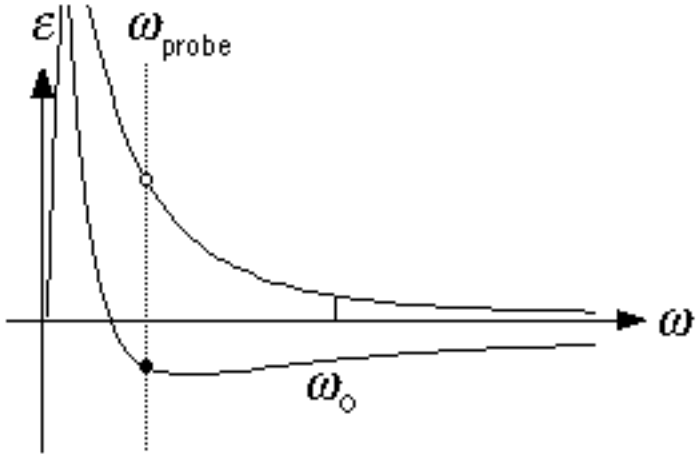

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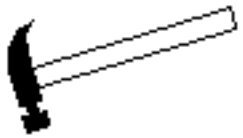

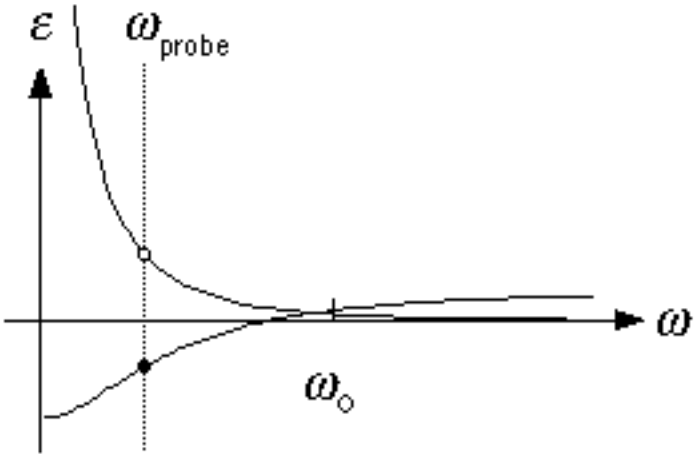

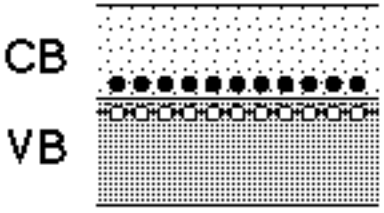

semimetal

...and induces change in bandstructure

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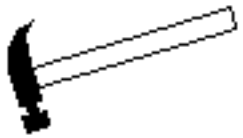

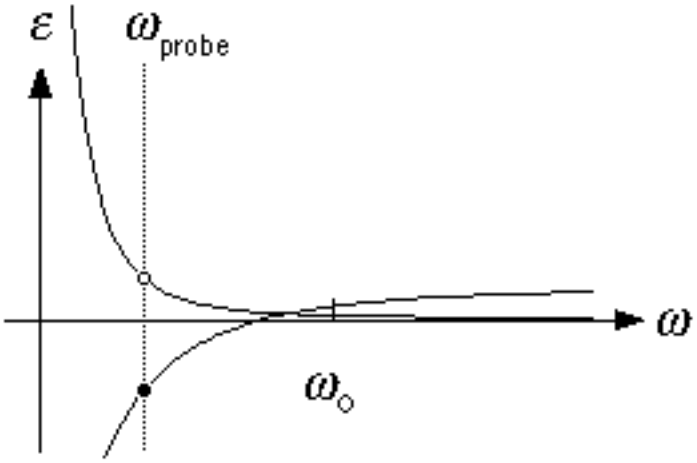

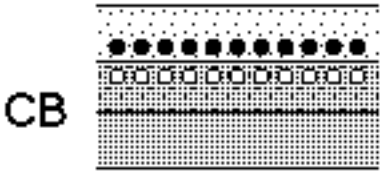

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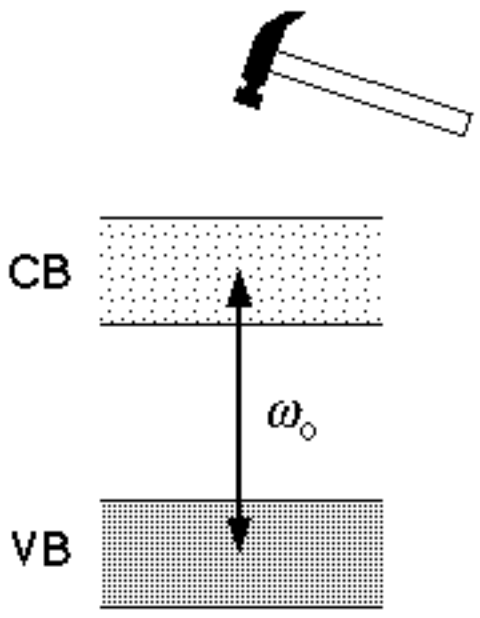

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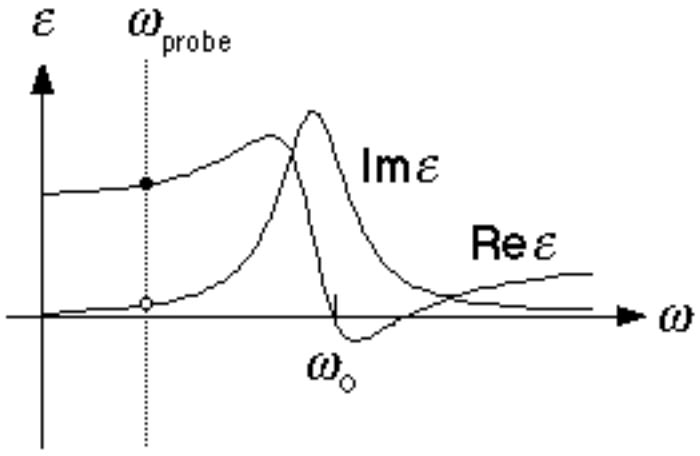

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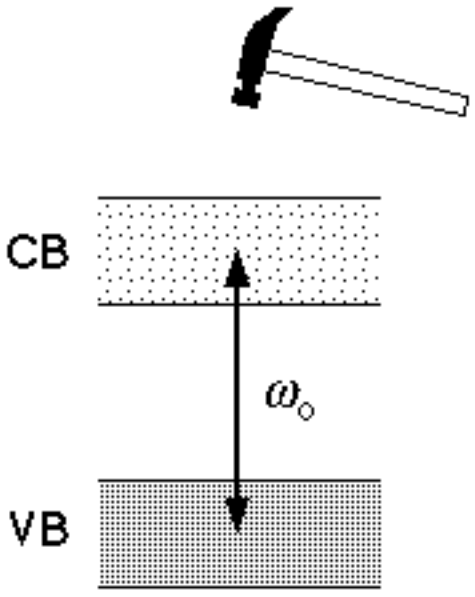

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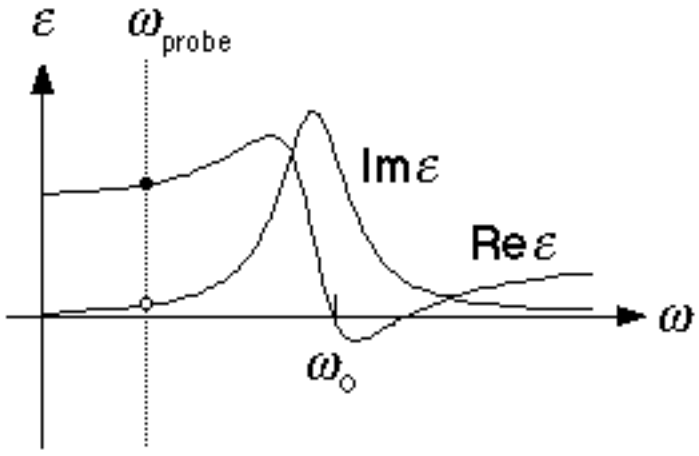

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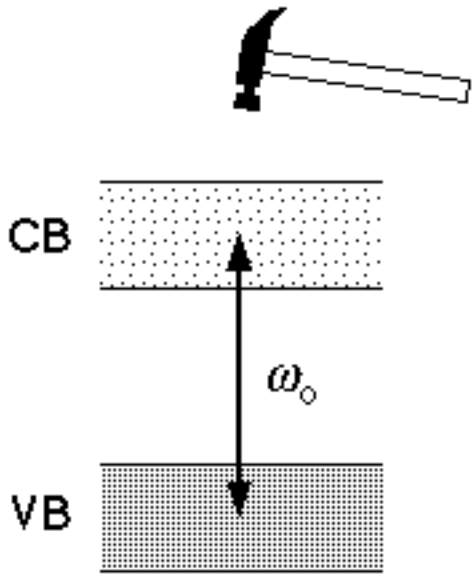

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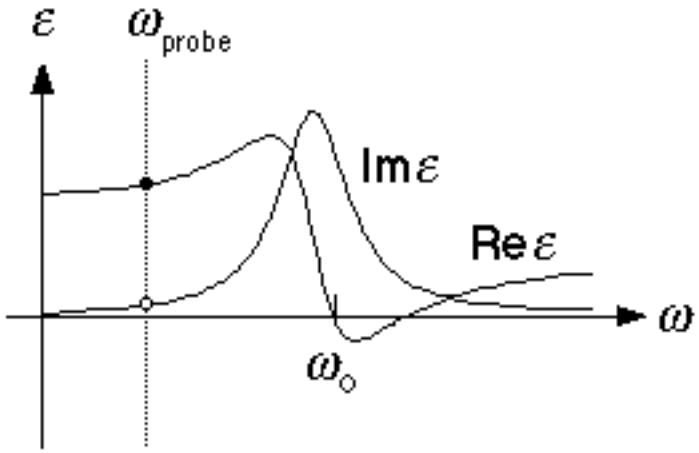

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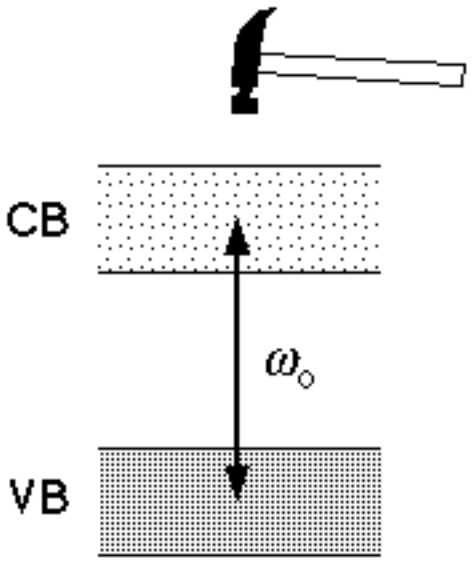

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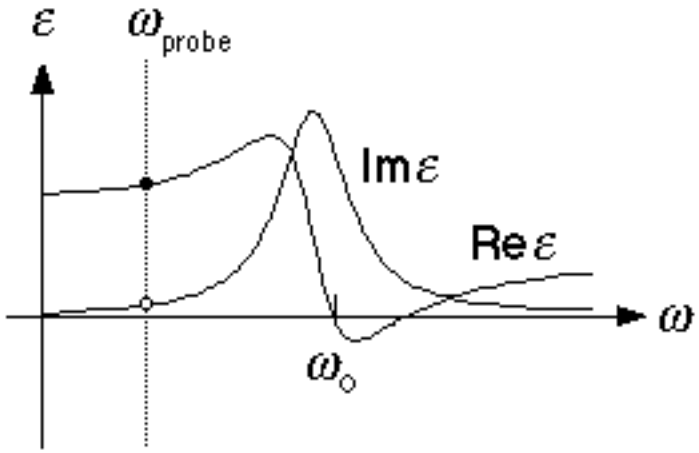

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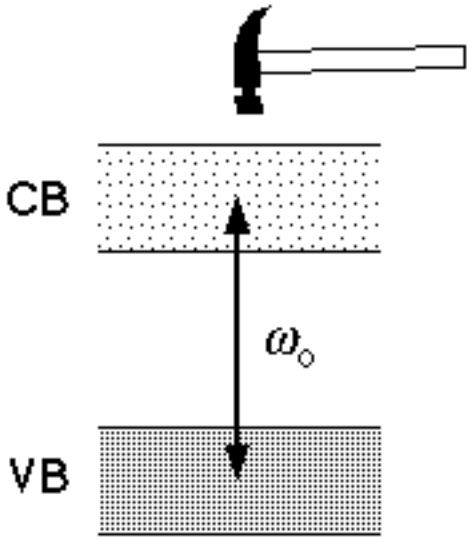

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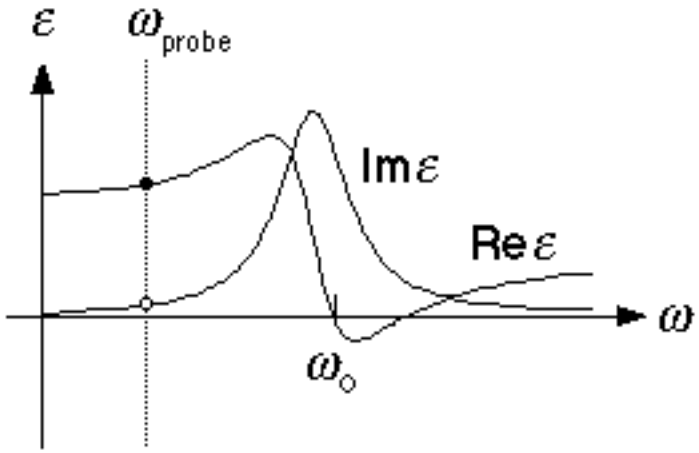

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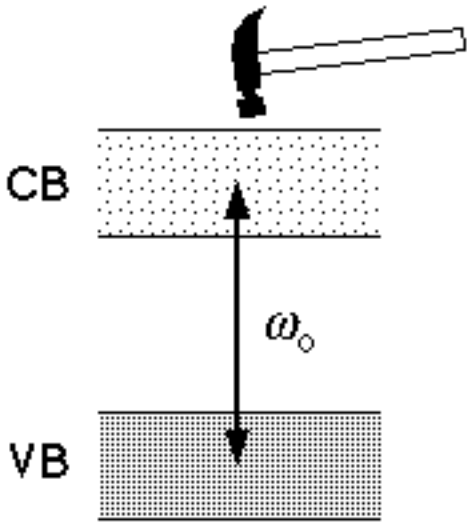

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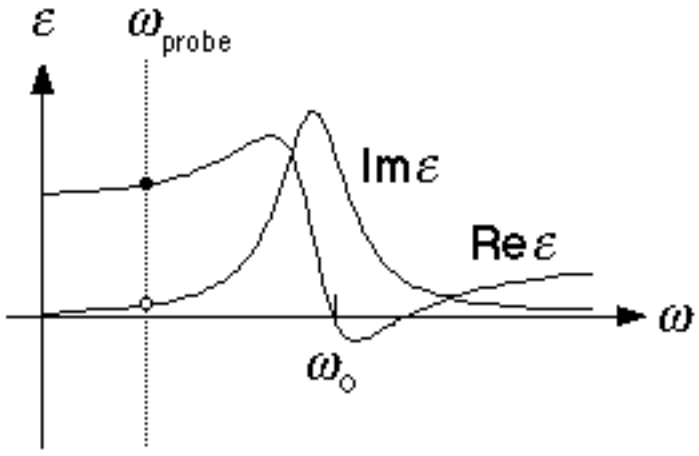

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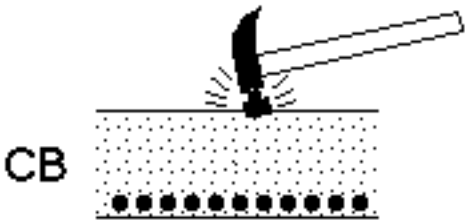

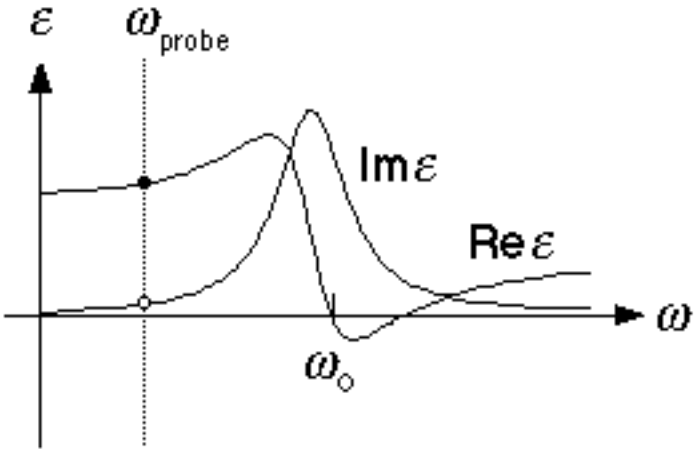

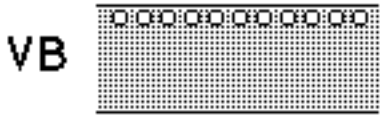

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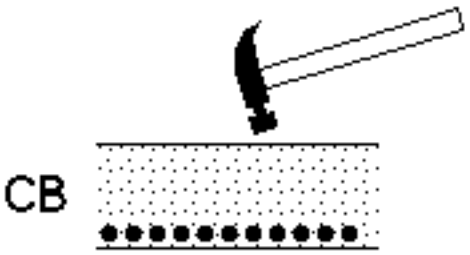

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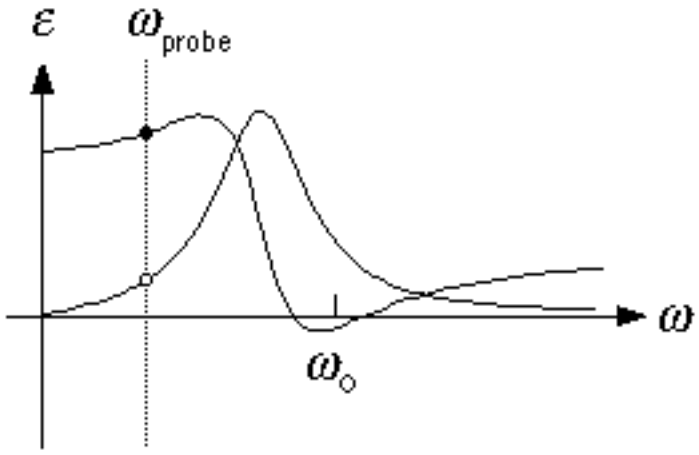

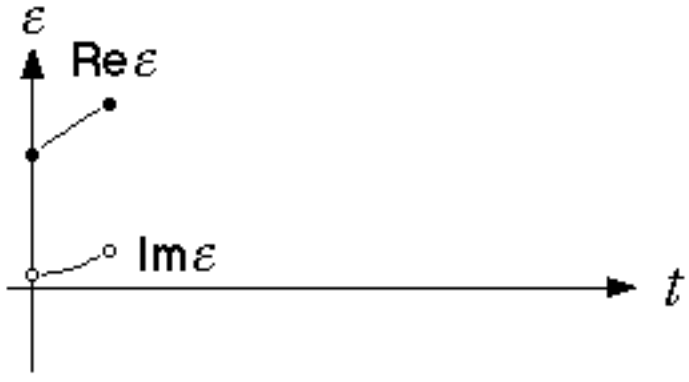

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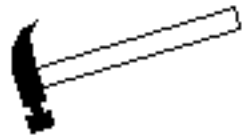

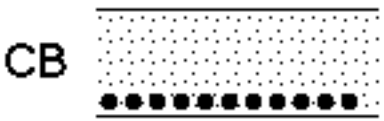

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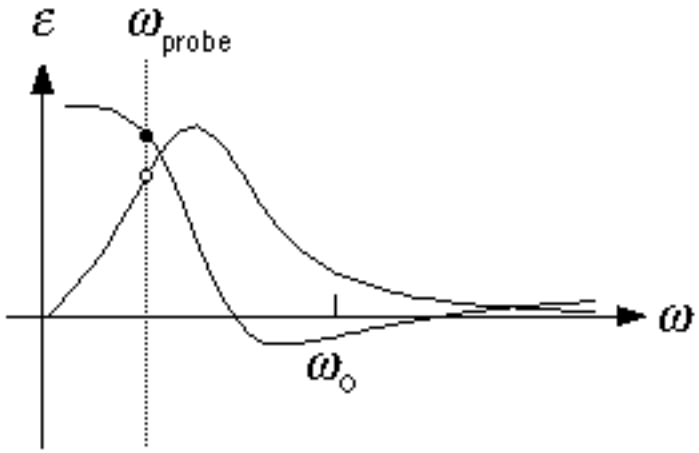

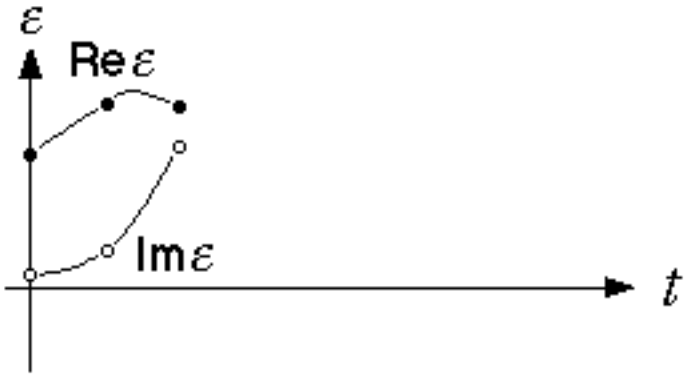

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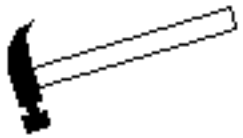

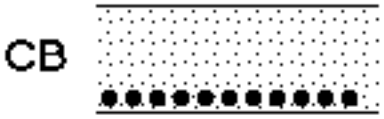

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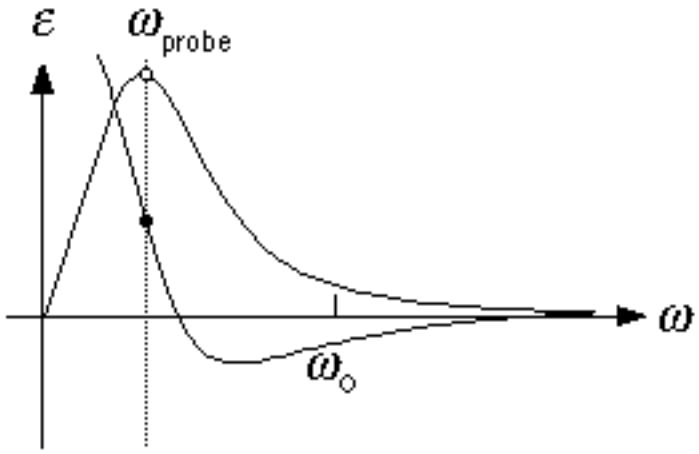

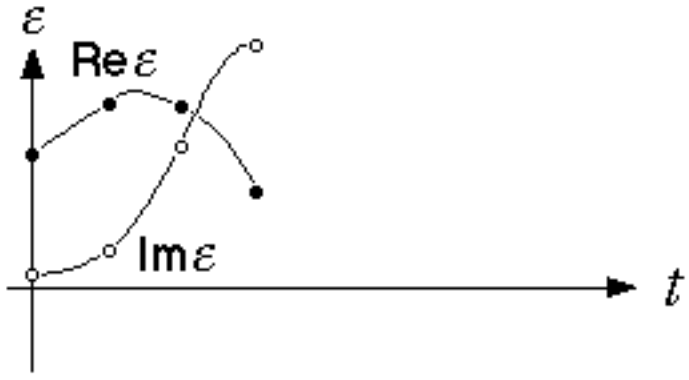

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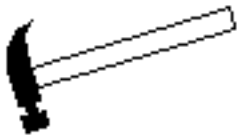

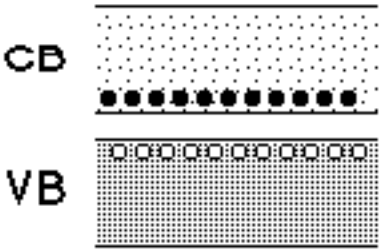

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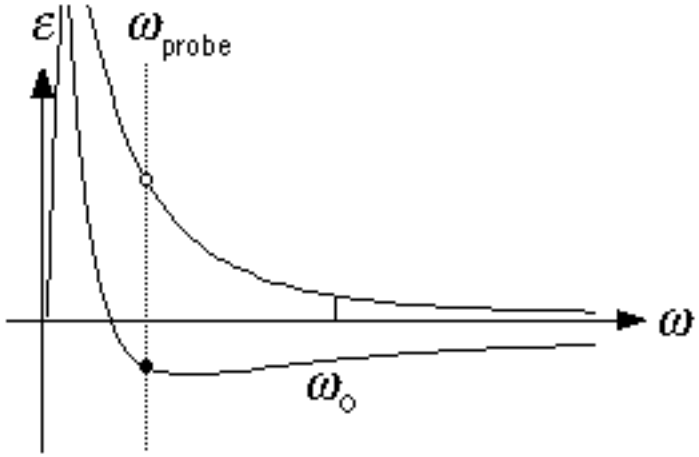

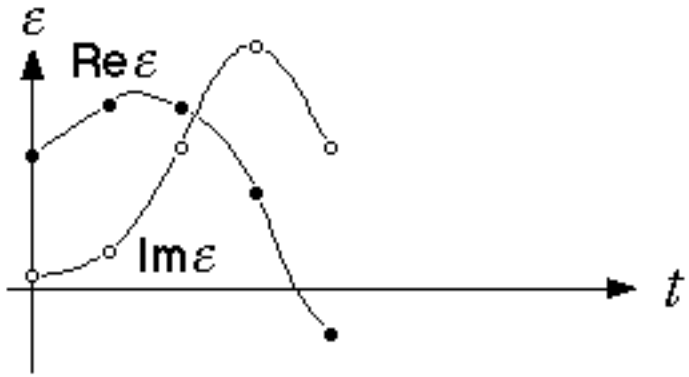

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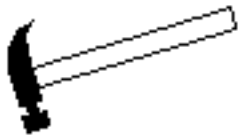
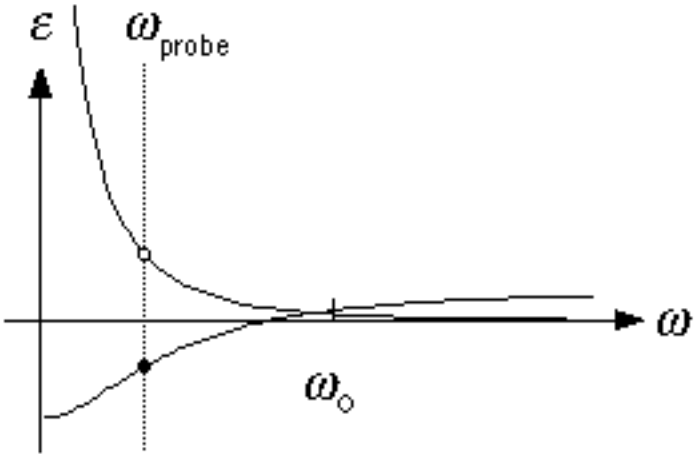
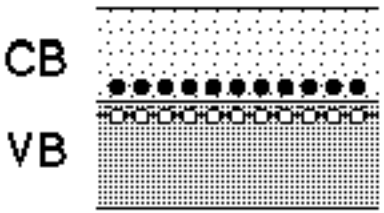
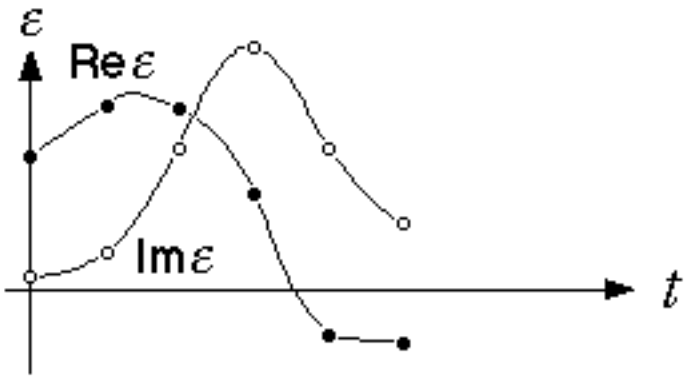






semimetal



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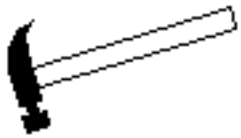
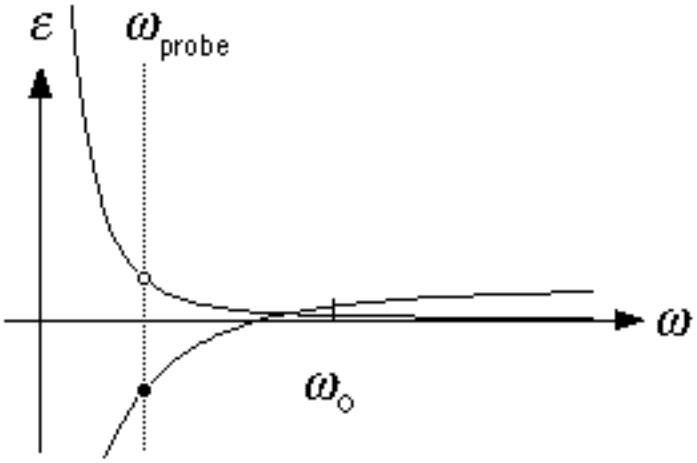
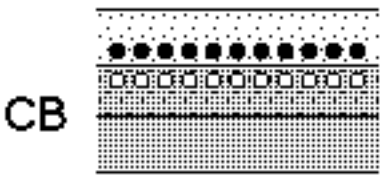
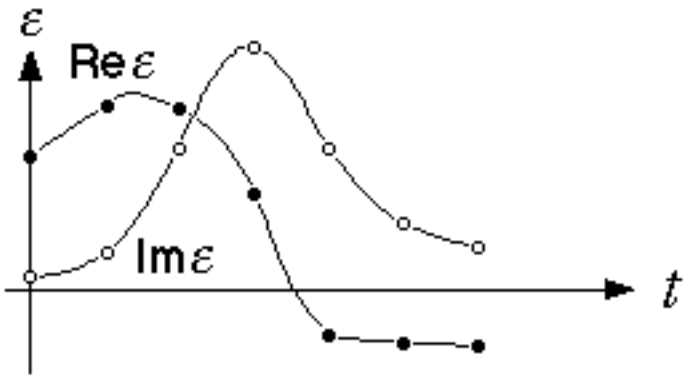


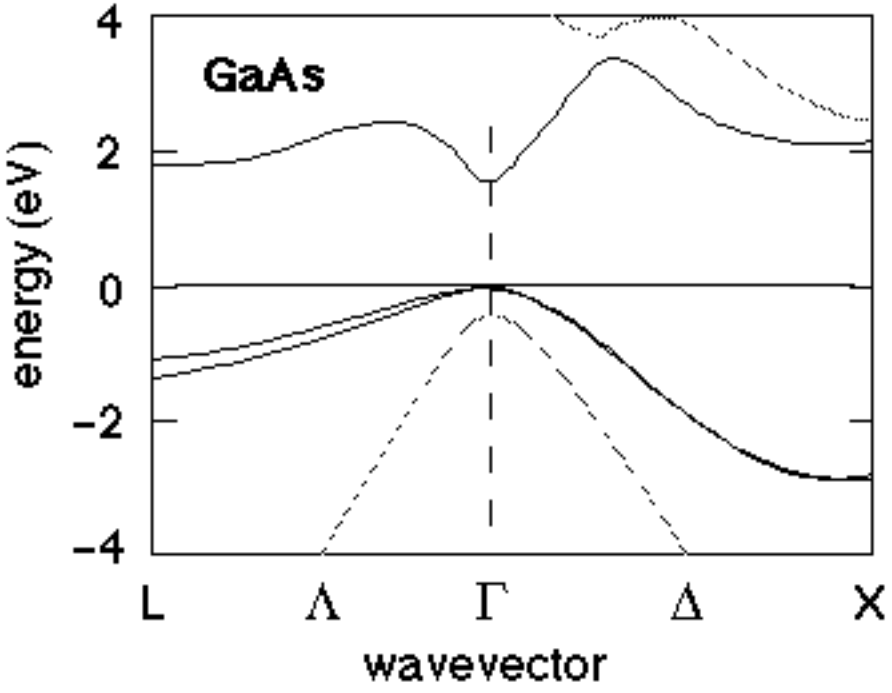




metal



GaAs BANDSTRUCTURE



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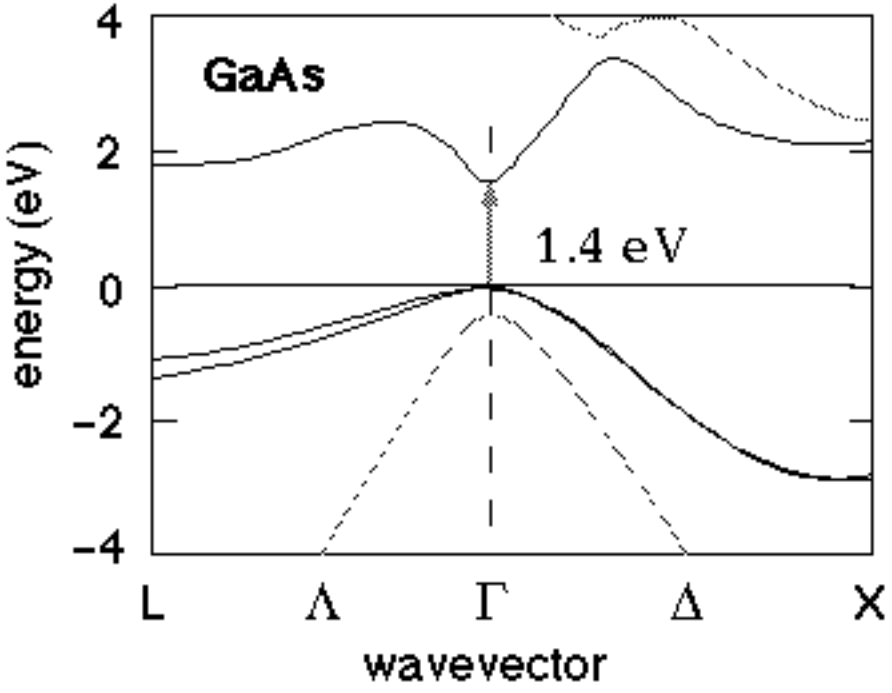

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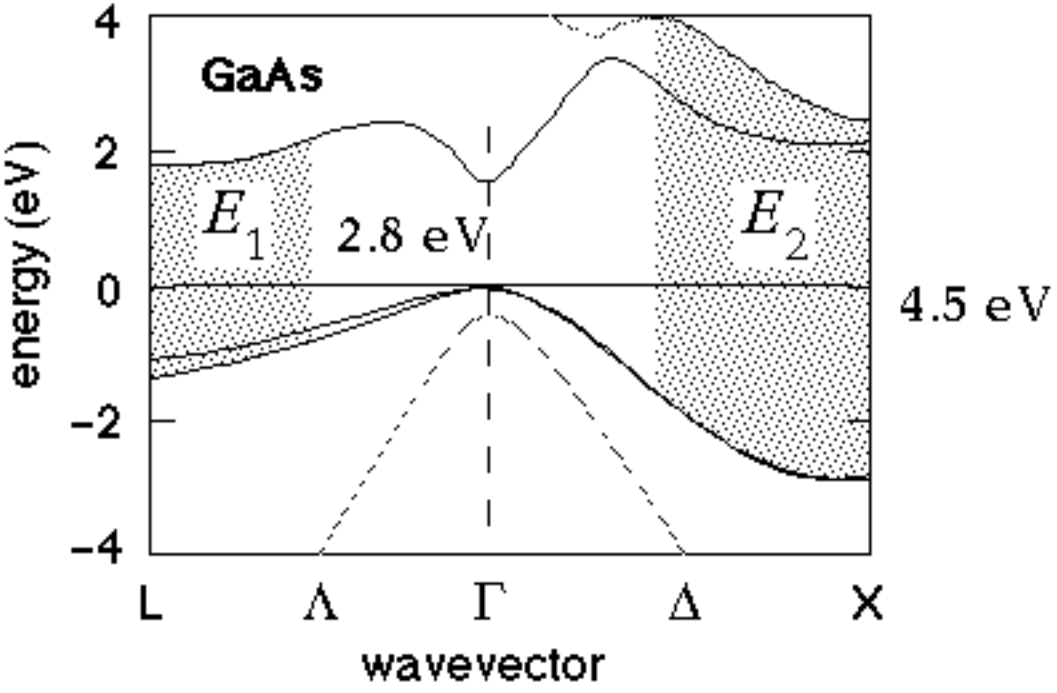

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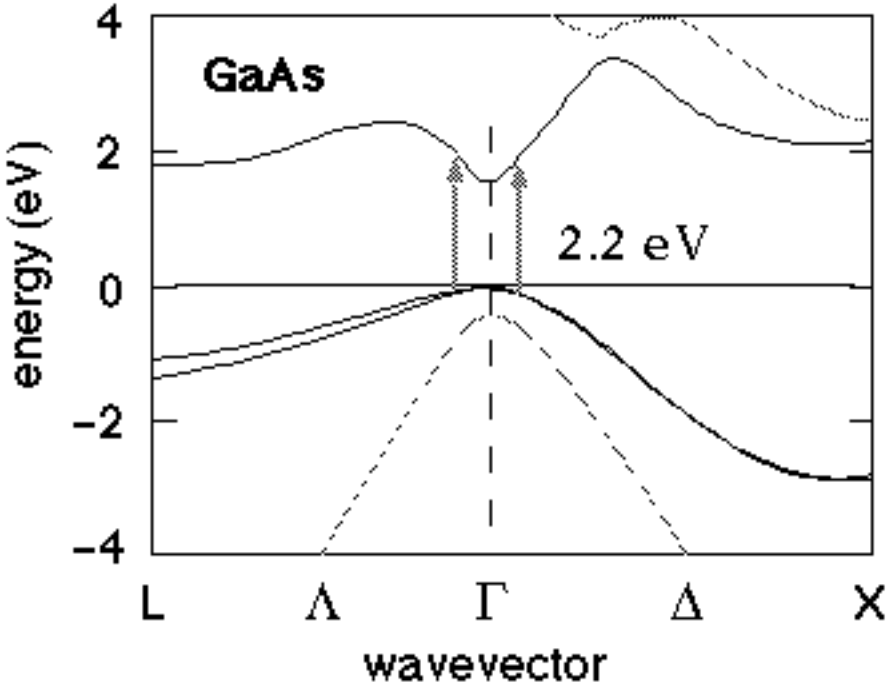

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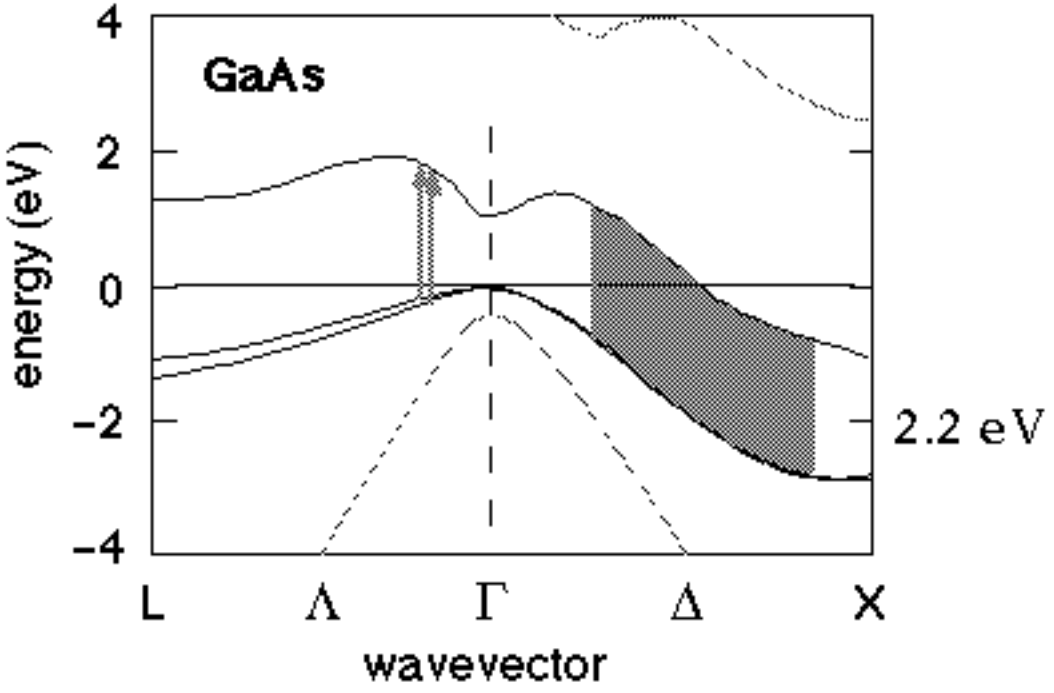
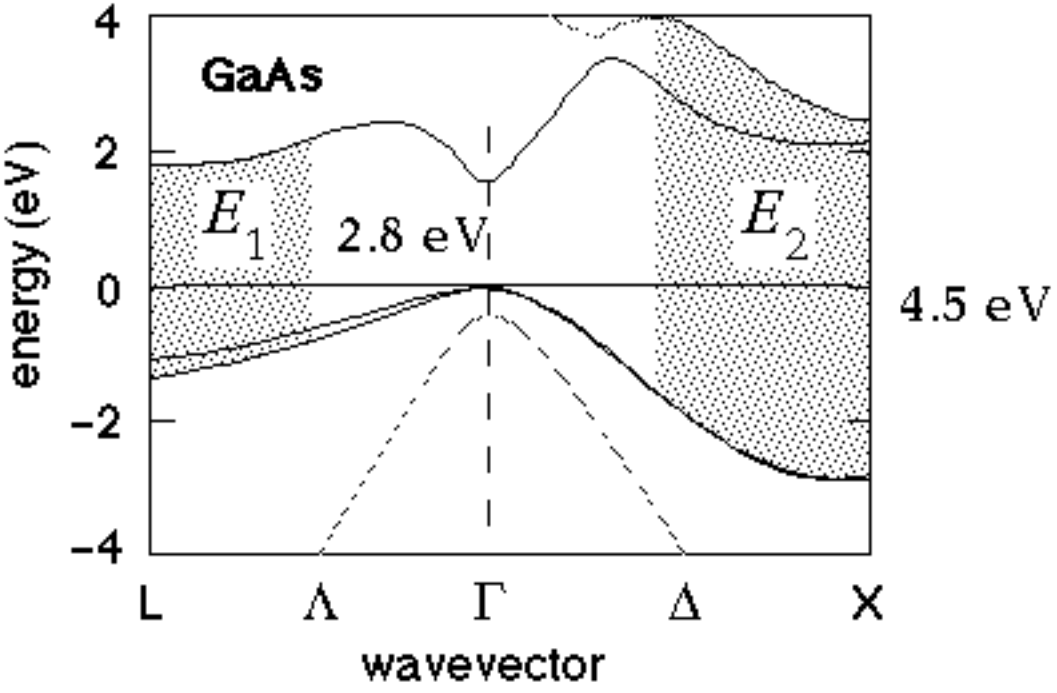
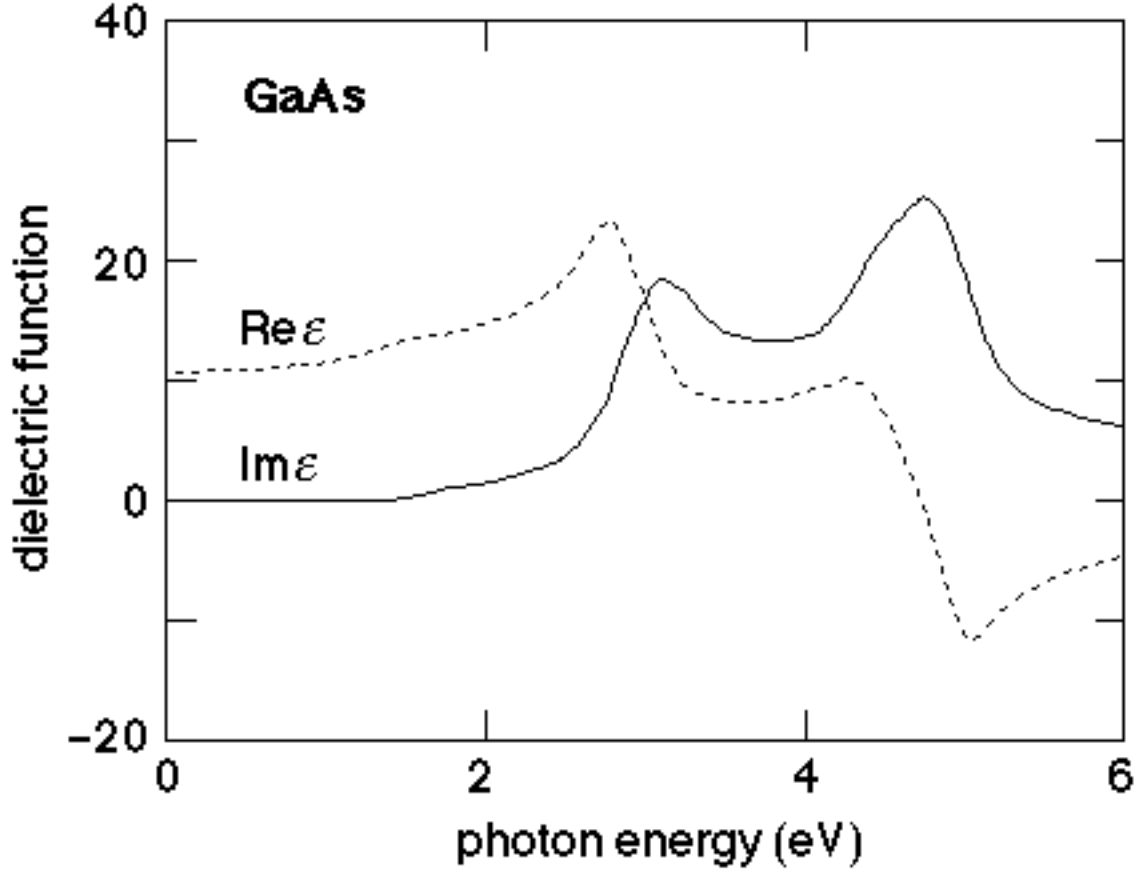


S. Froyen *et al., Phys Rev. B* **28** (1983) 3258
D.H. Kim *et al., Sol. State. Comm.* **89** (1994) 119

GaAs BANDSTRUCTURE



GaAs DIELECTRIC FUNCTION



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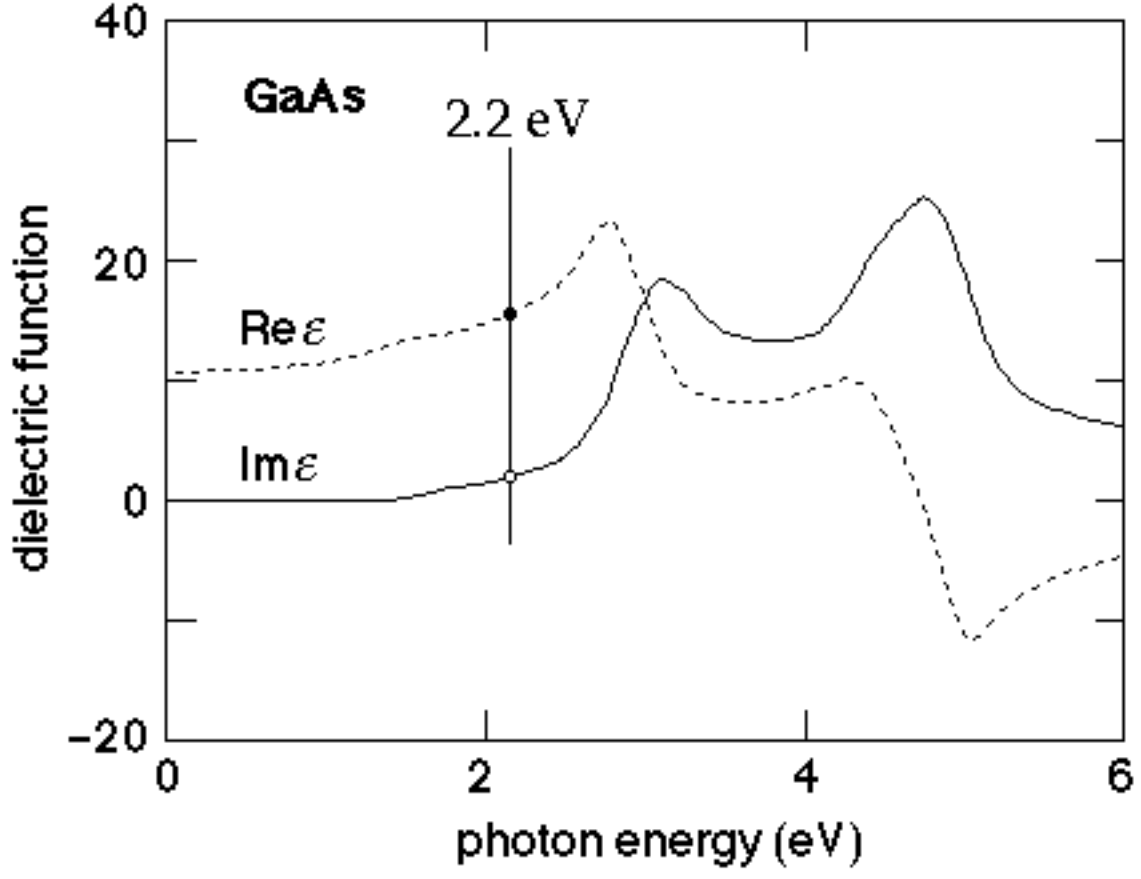

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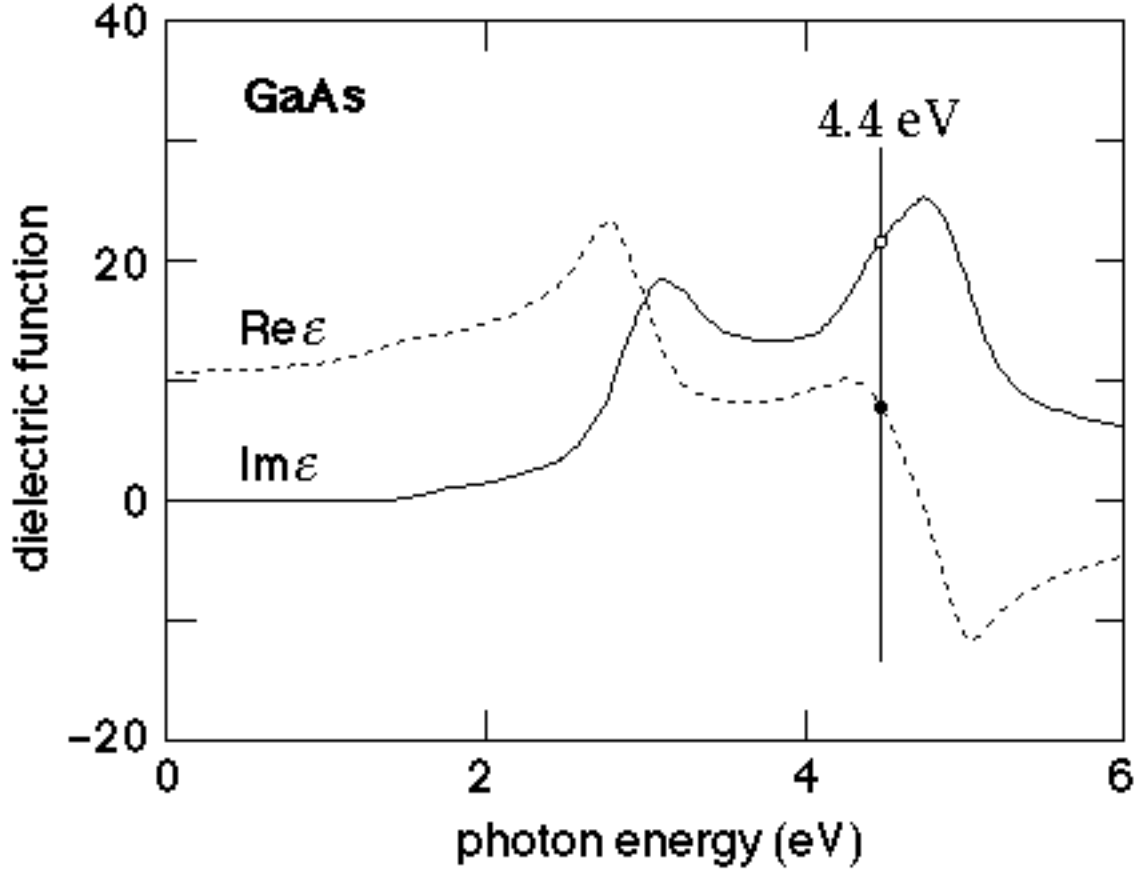
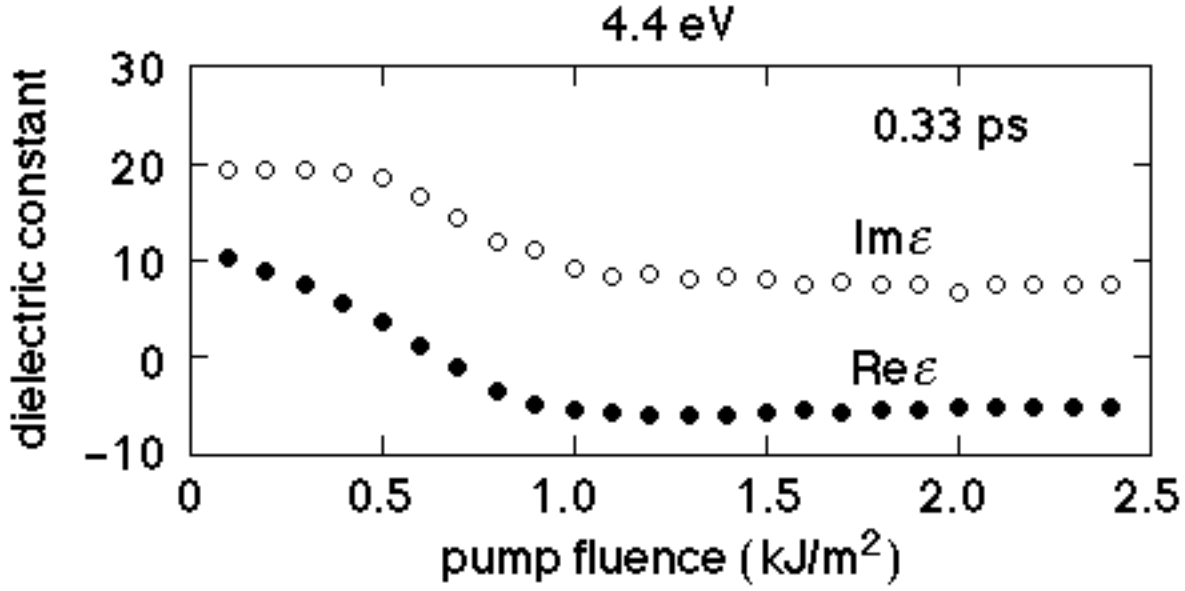


FLUENCE DEPENDENCE



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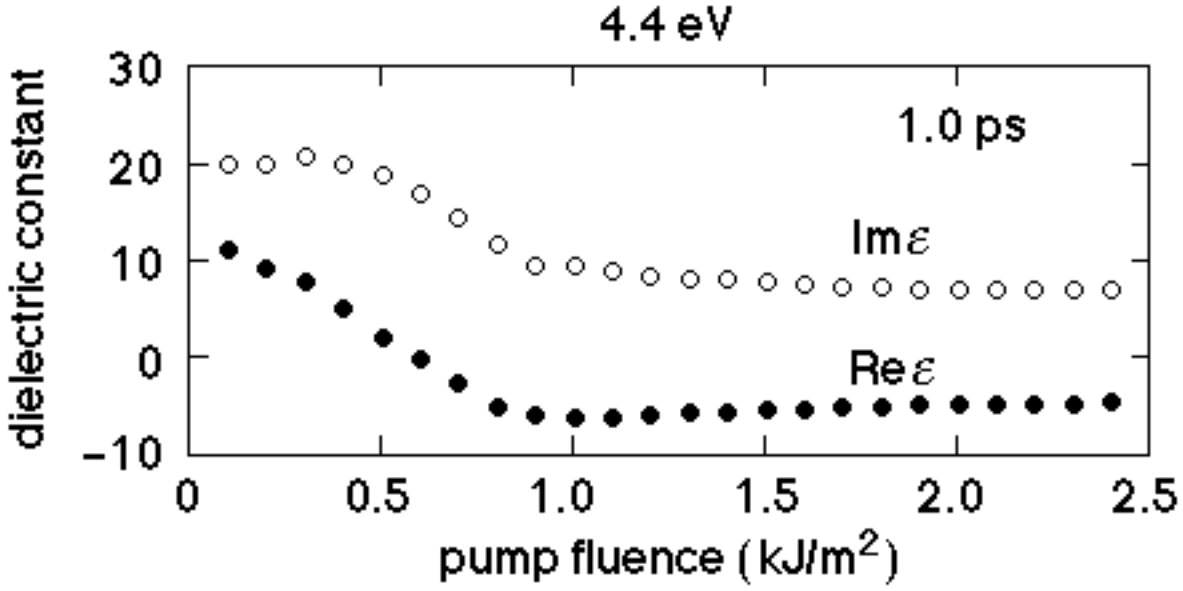

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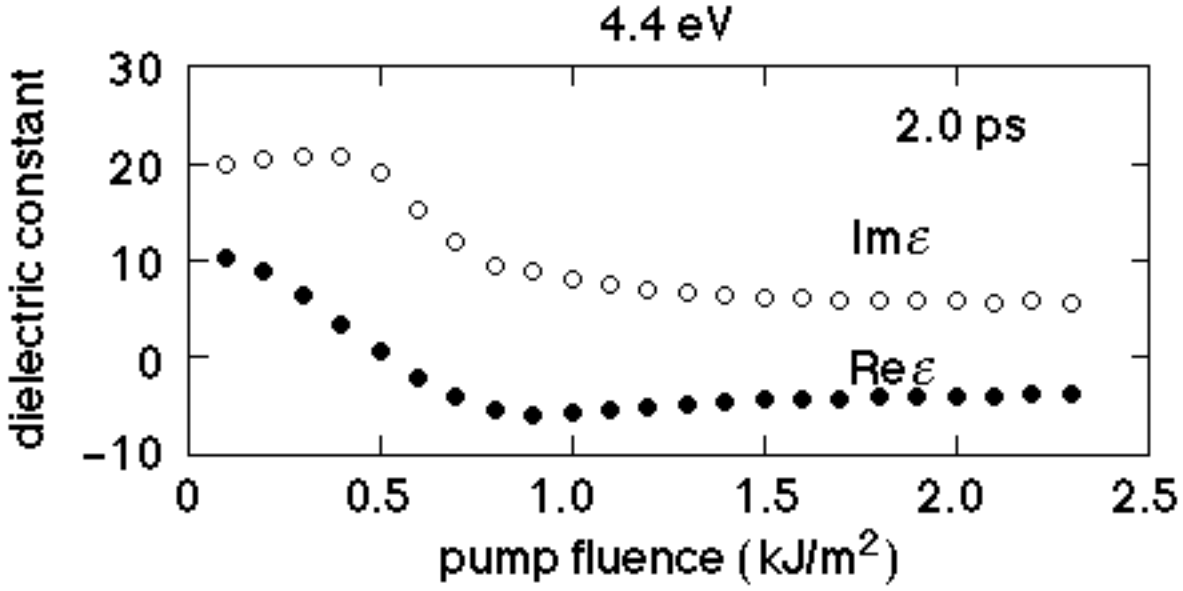

Phys. Rev. B **51** (1995) 6959

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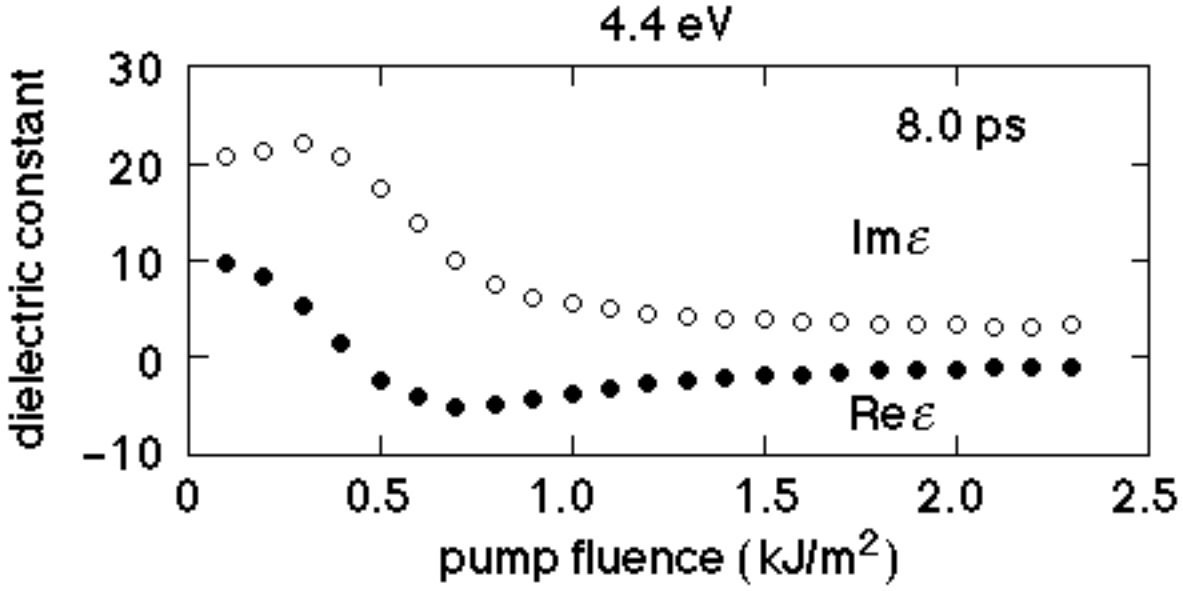

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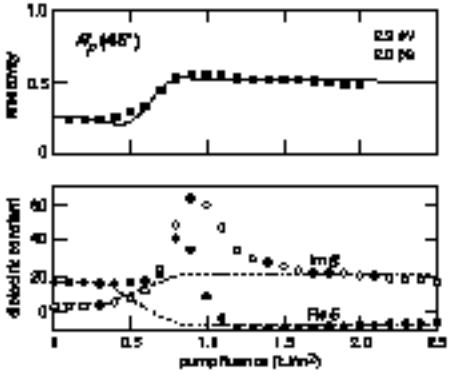
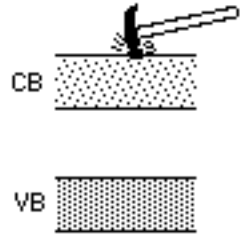
SUMMARY

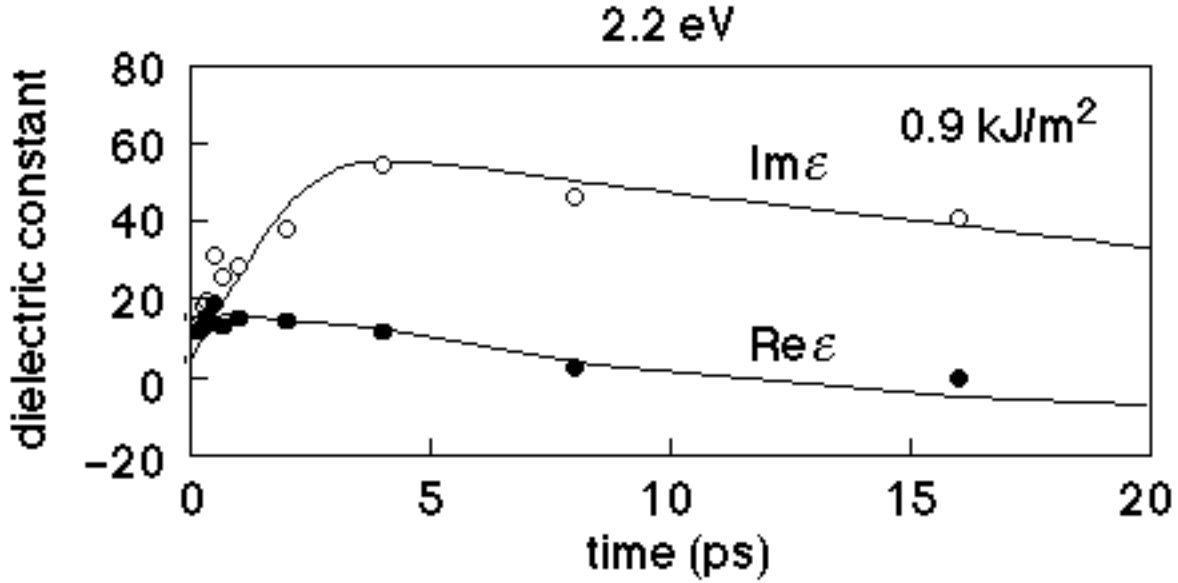


single-angle reflectivity not a good indicator of what happens



laser pulse causes bandgap collapse

CAUSE OF COLLAPSE



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

structural change?

Field strength at second harmonic:

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

Field strength at second harmonic:

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

where:

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

so:

Field strength at second harmonic:

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

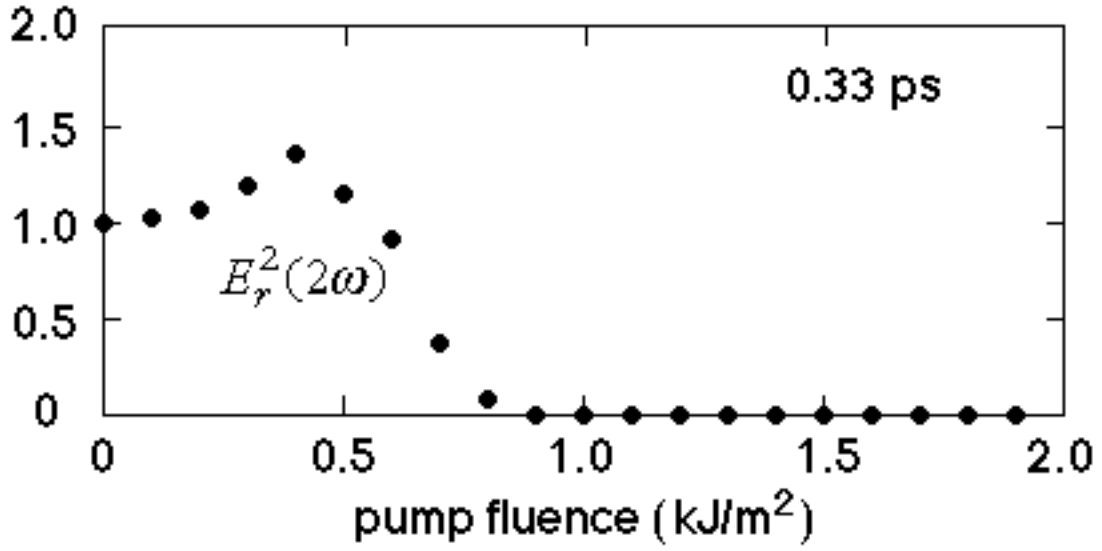
where:

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

so:

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

SECOND HARMONIC DATA



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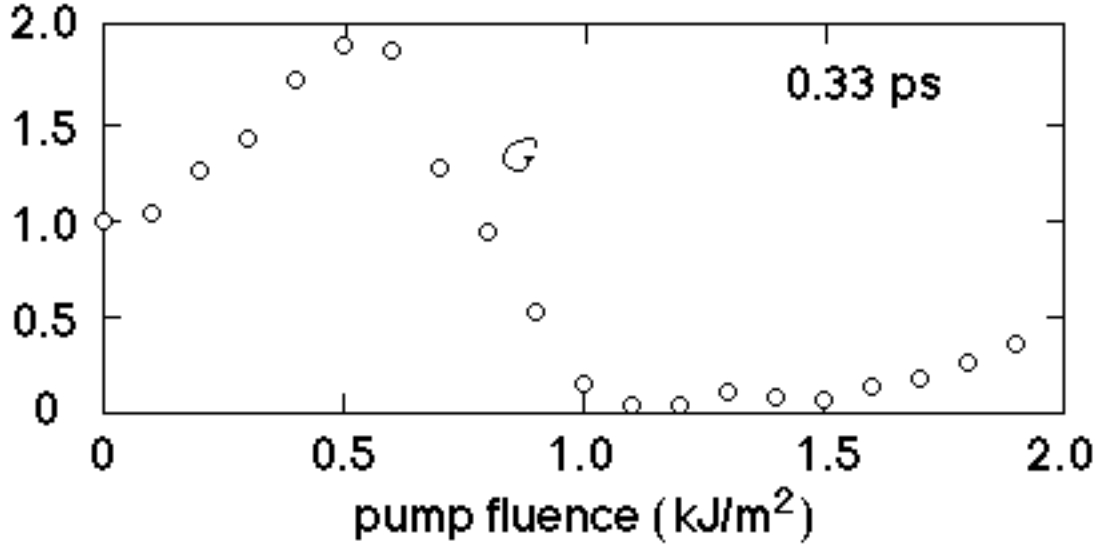

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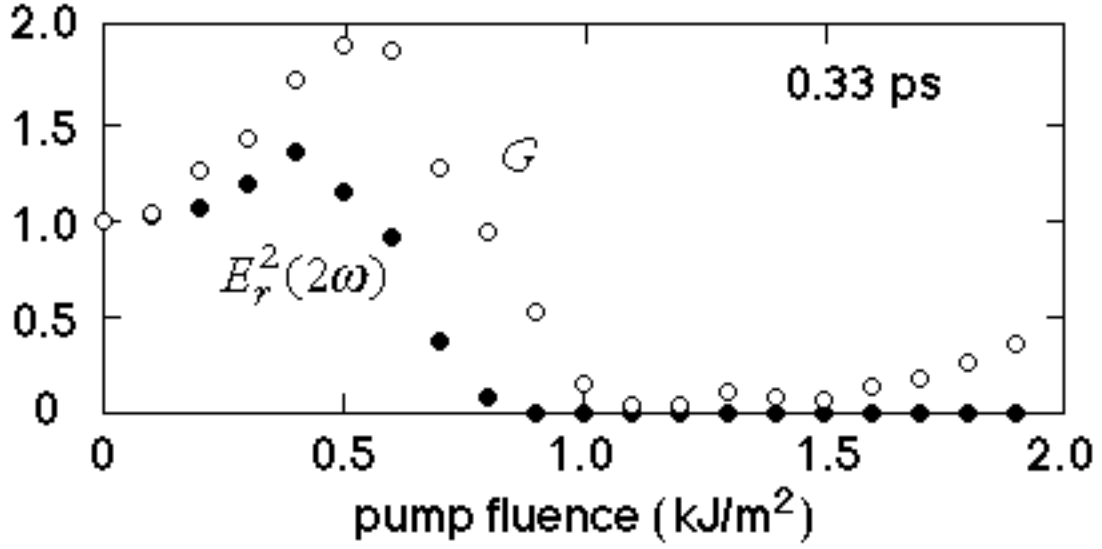

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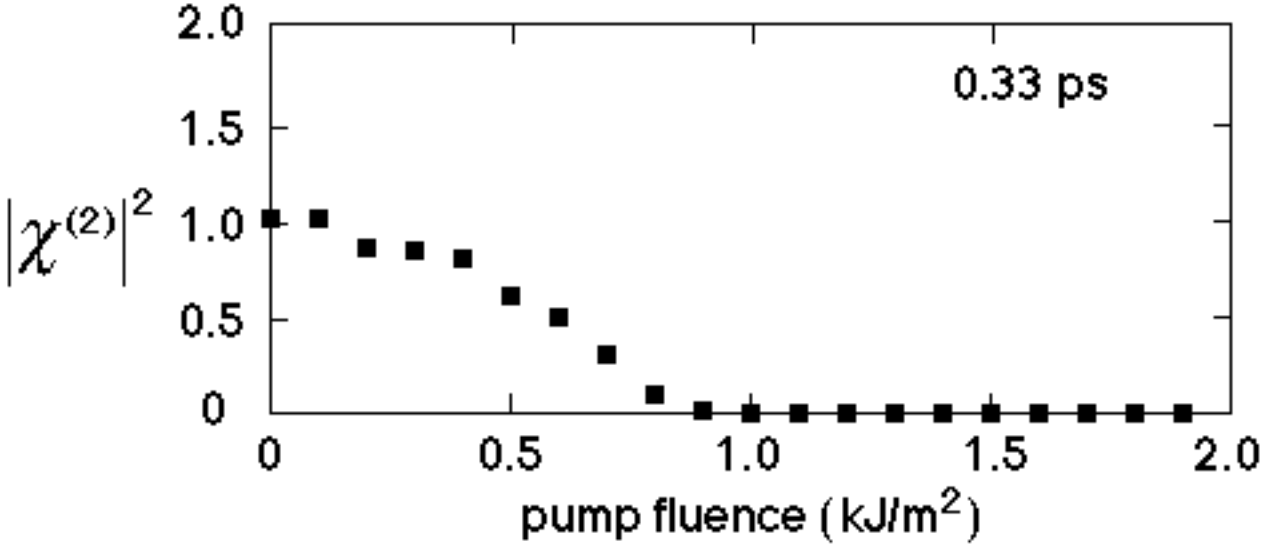

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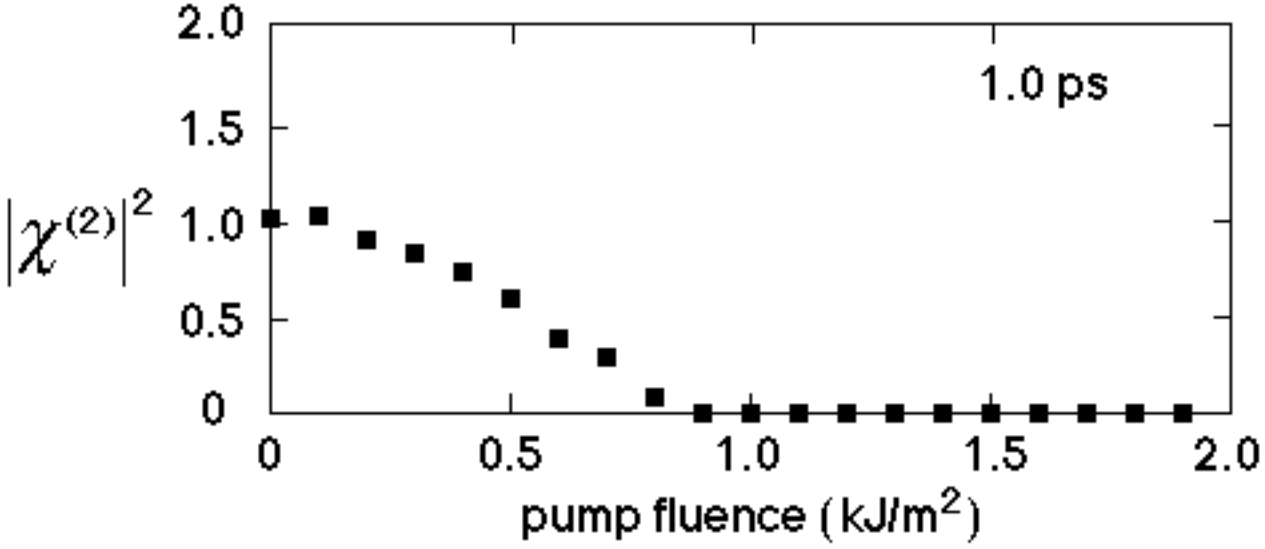

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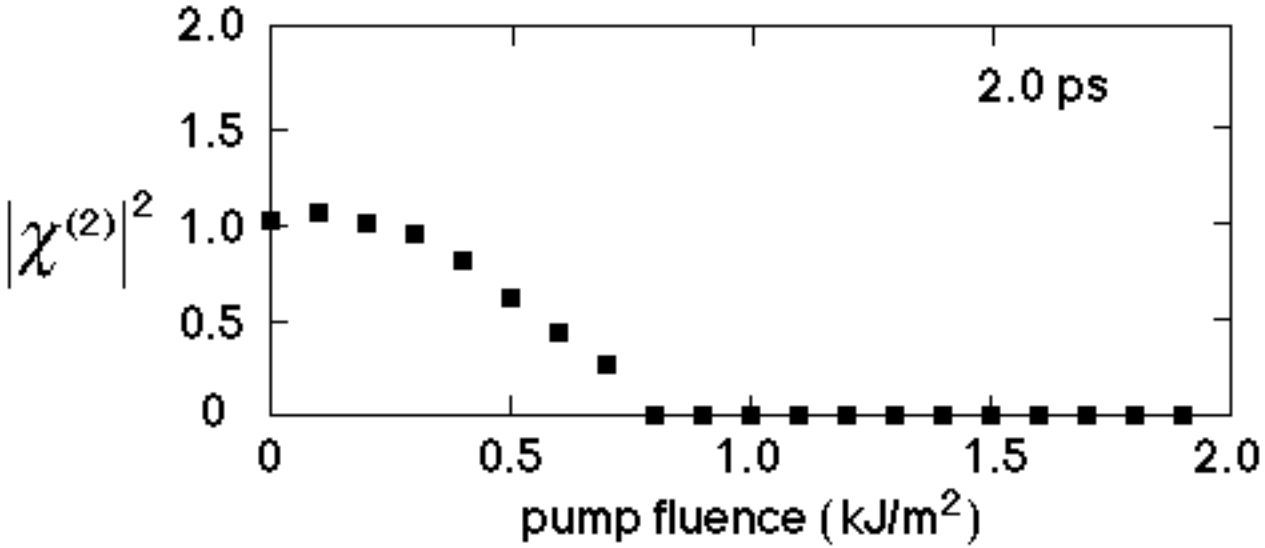

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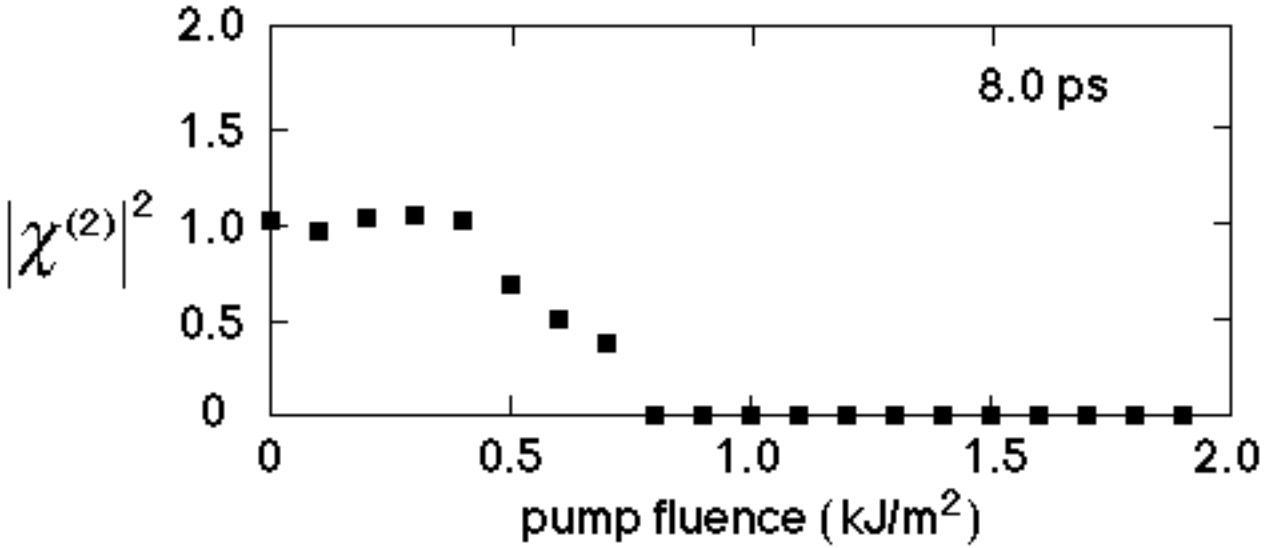

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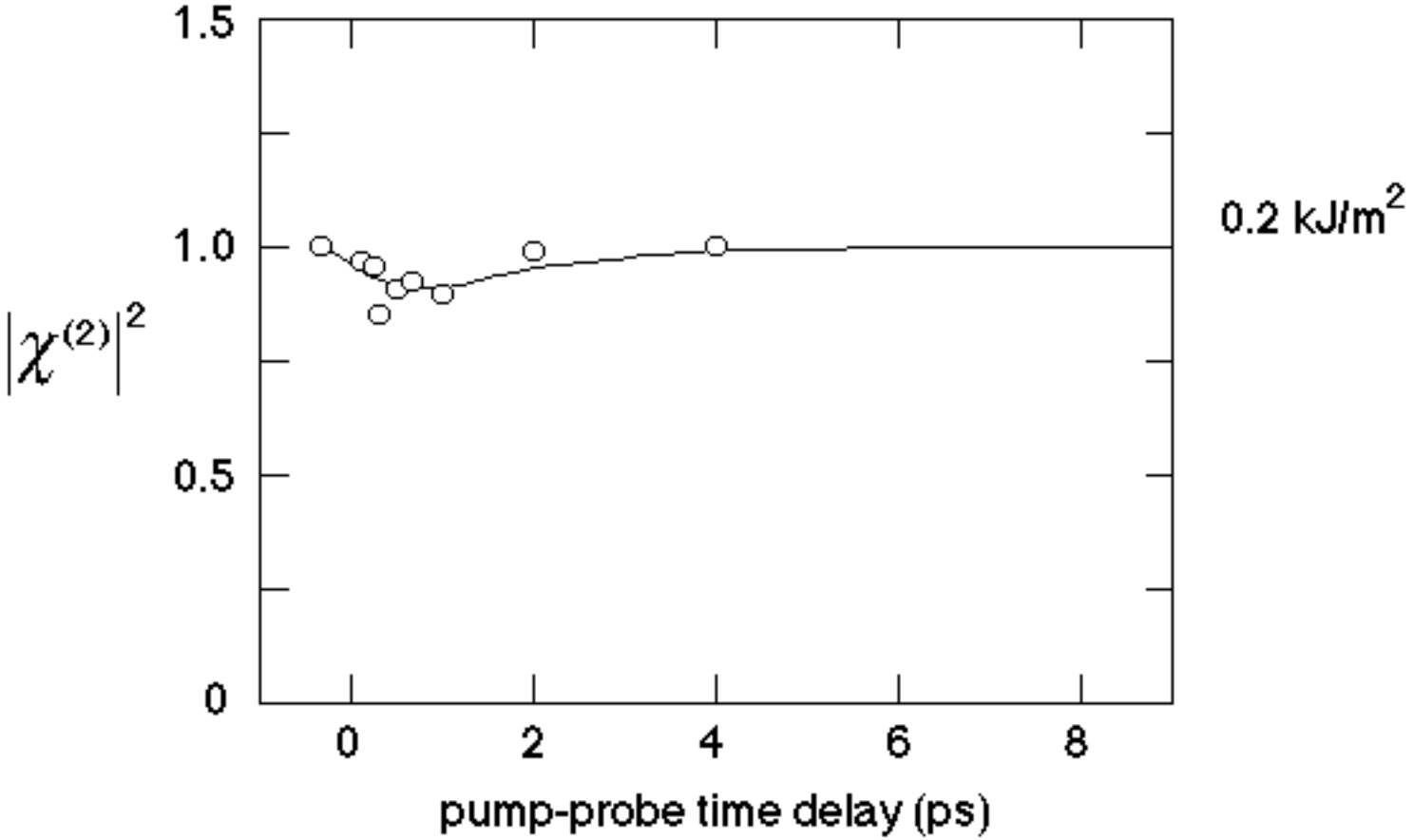

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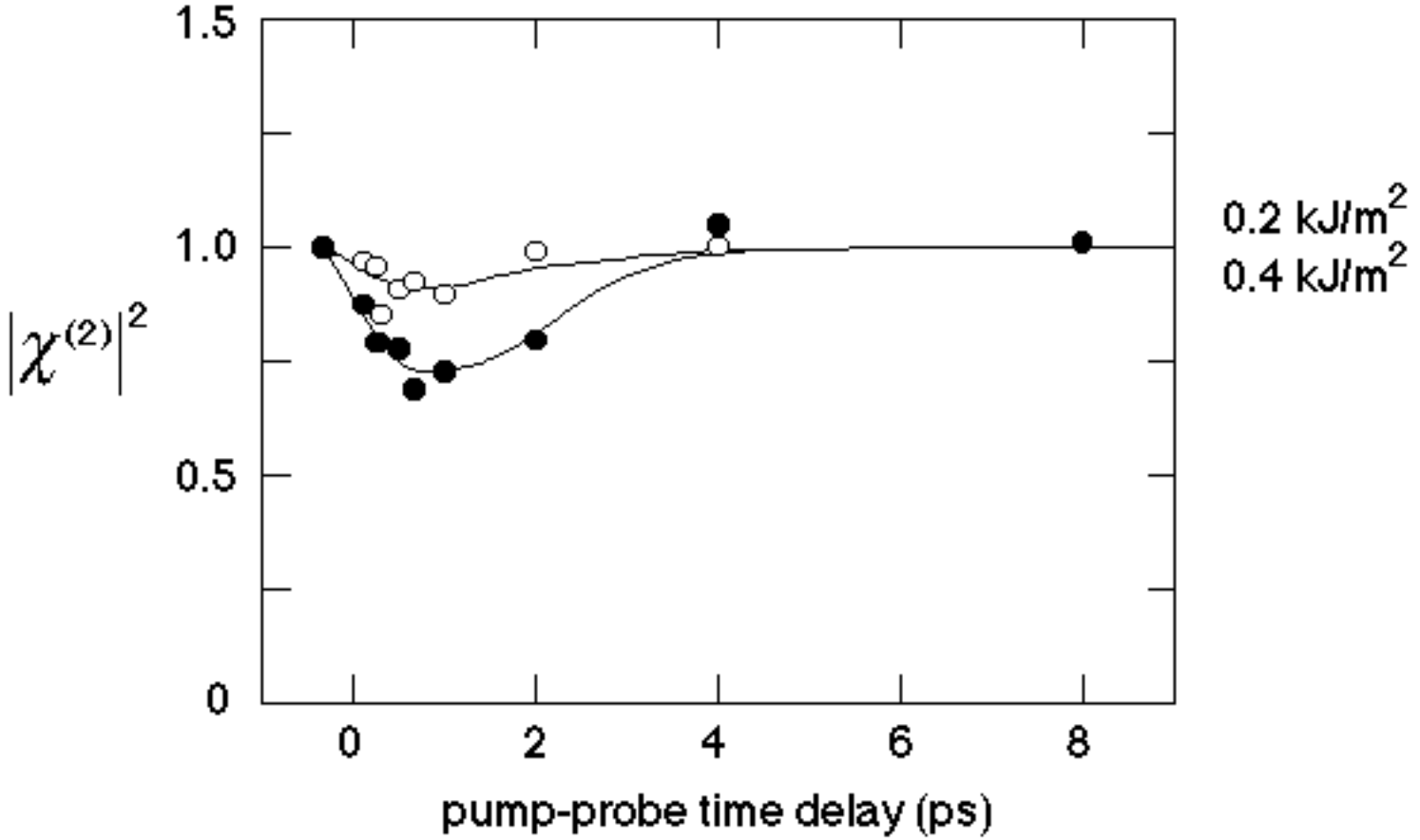

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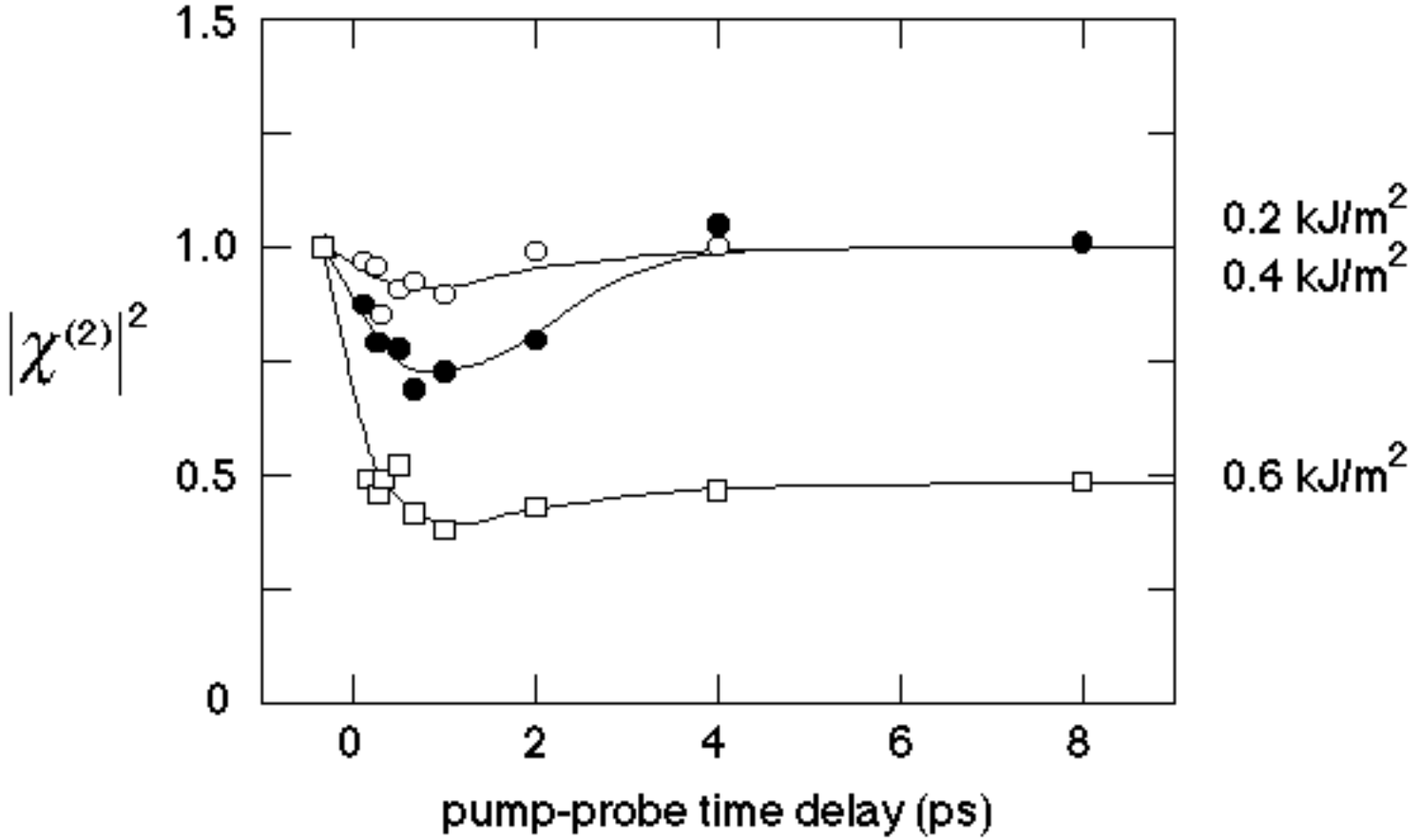

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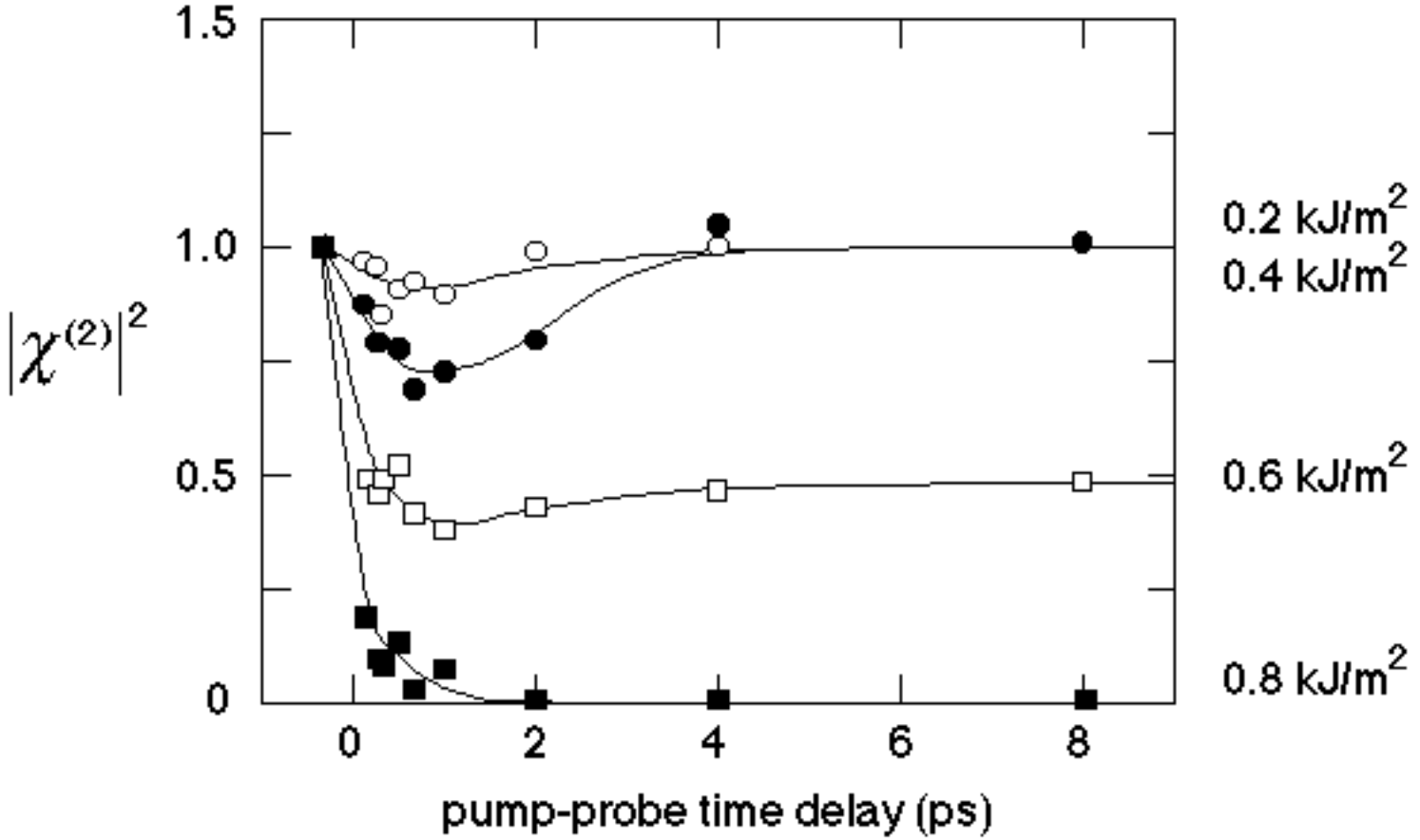

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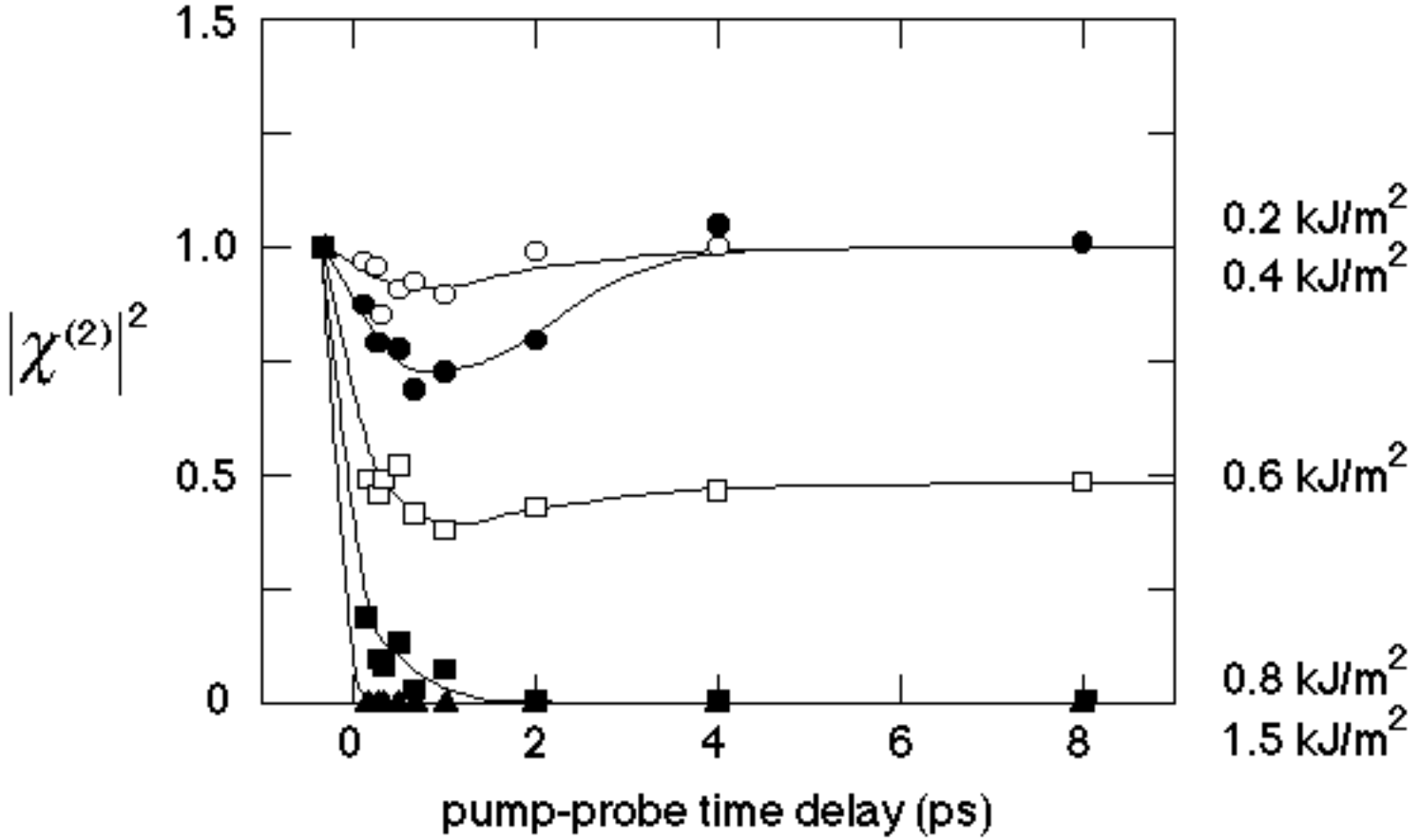

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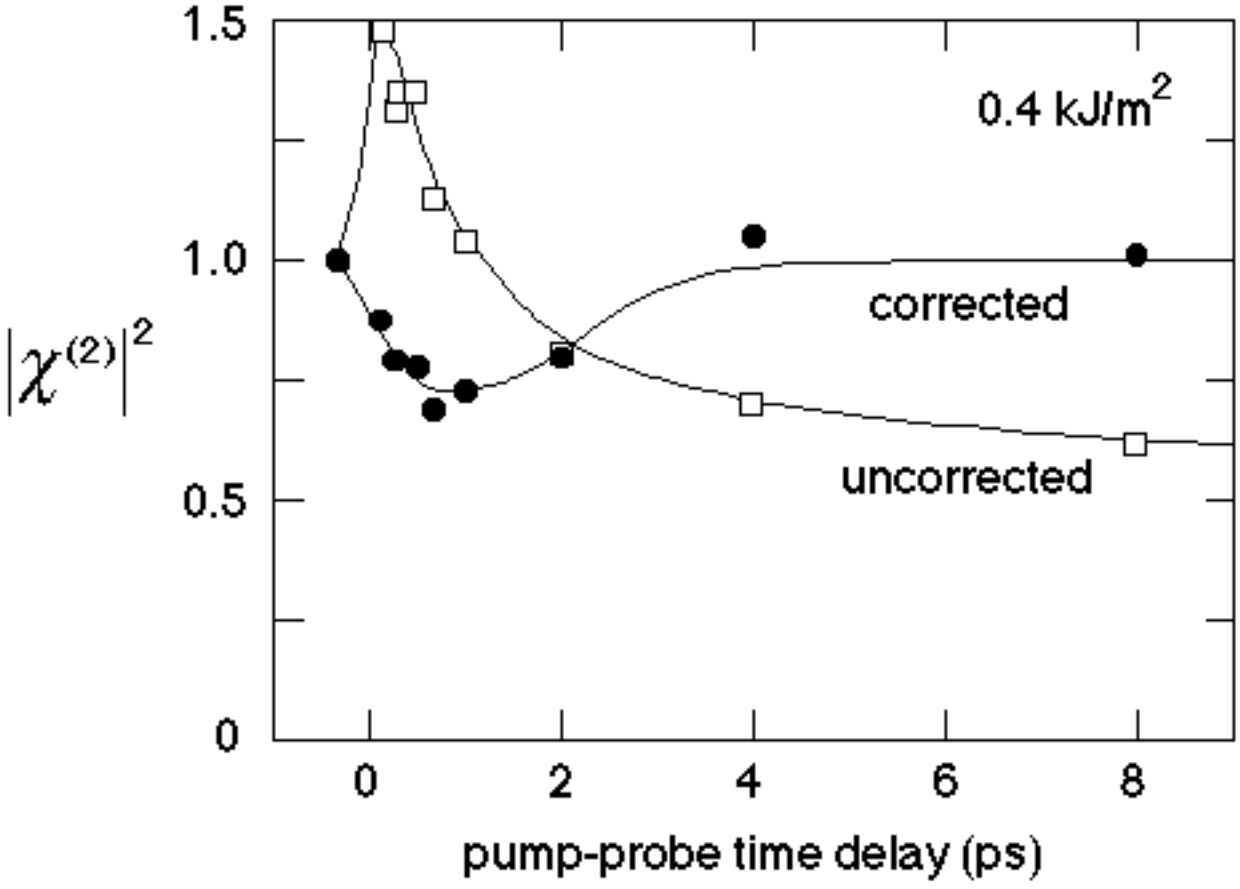
EFFECT OF CHANGES IN ε



FEMTOSECOND WORK

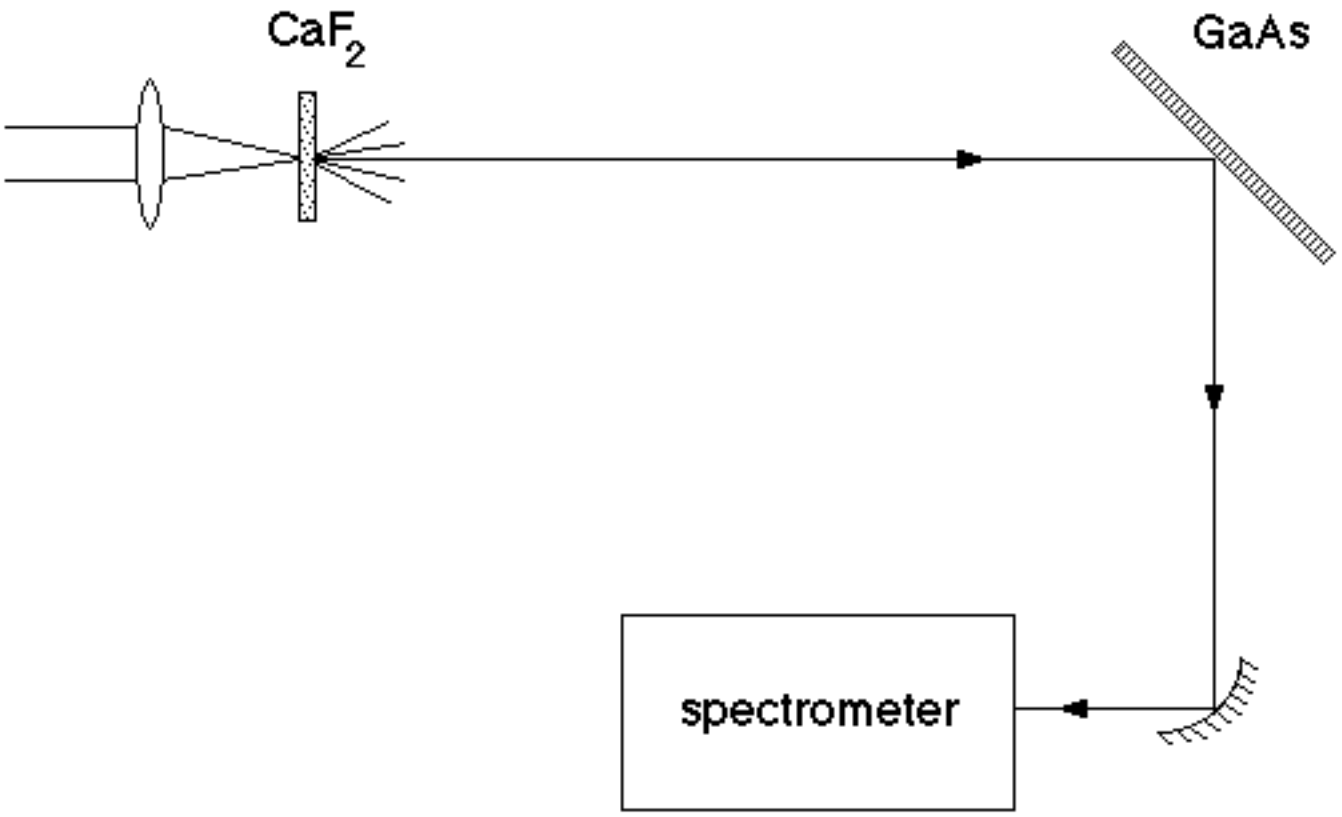
- $\chi^{(2)}$ sensitive to loss of long-range order
- long-range order disappears even below threshold
- structural change due to destabilization of covalent bonds
- time-scale for structural change reasonable (10% of bond length in 1 ps requires 25 m/s)

❶ Background

❷ Single frequency work

❸ Broadband work

BROADBAND PROBE



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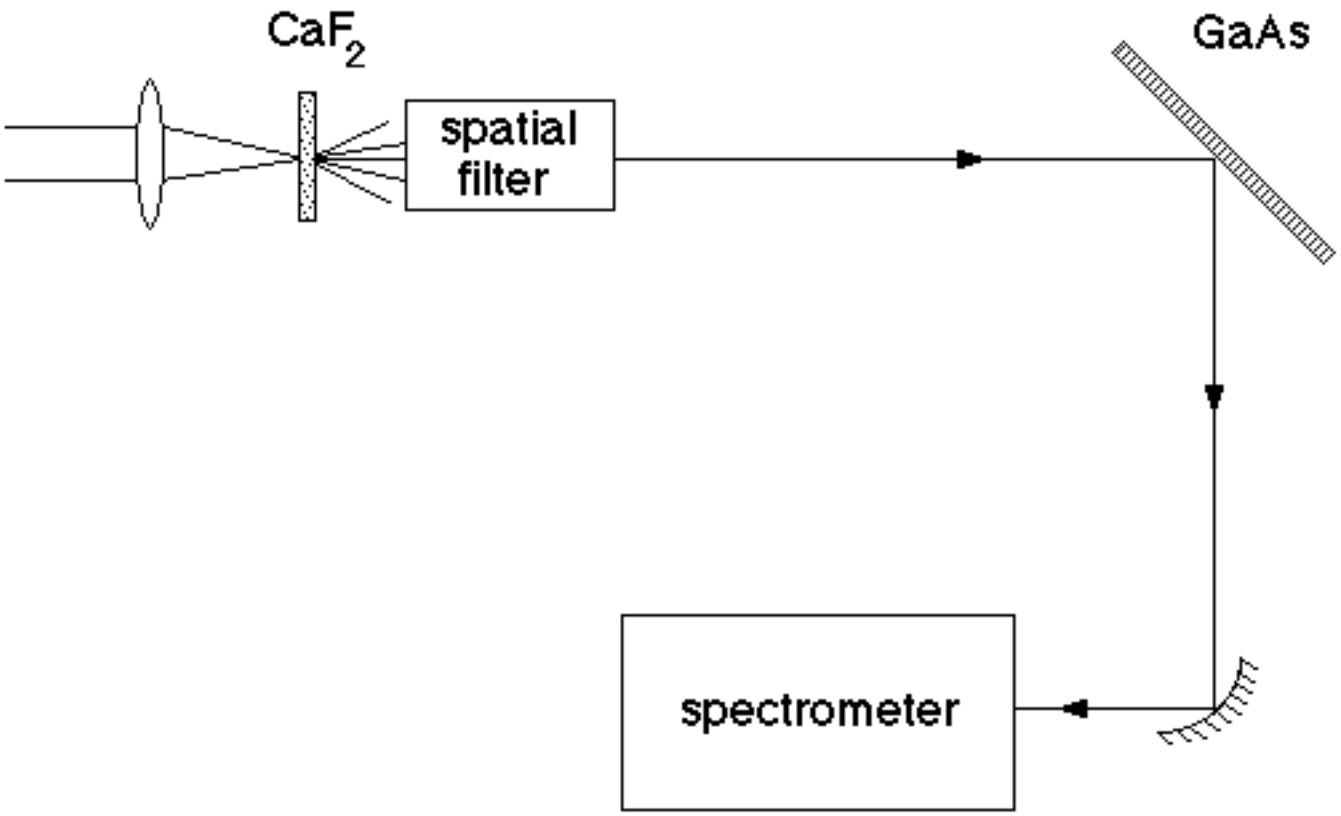

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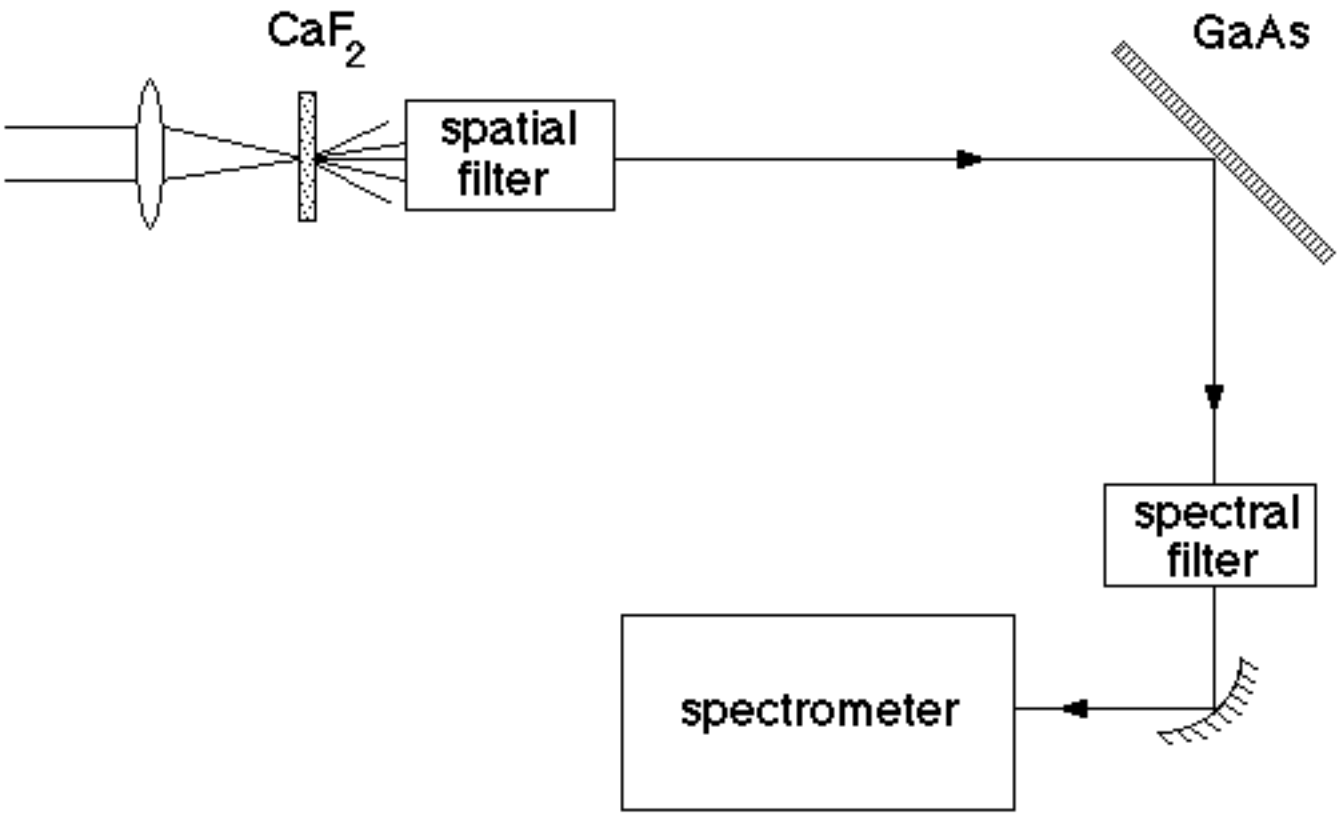

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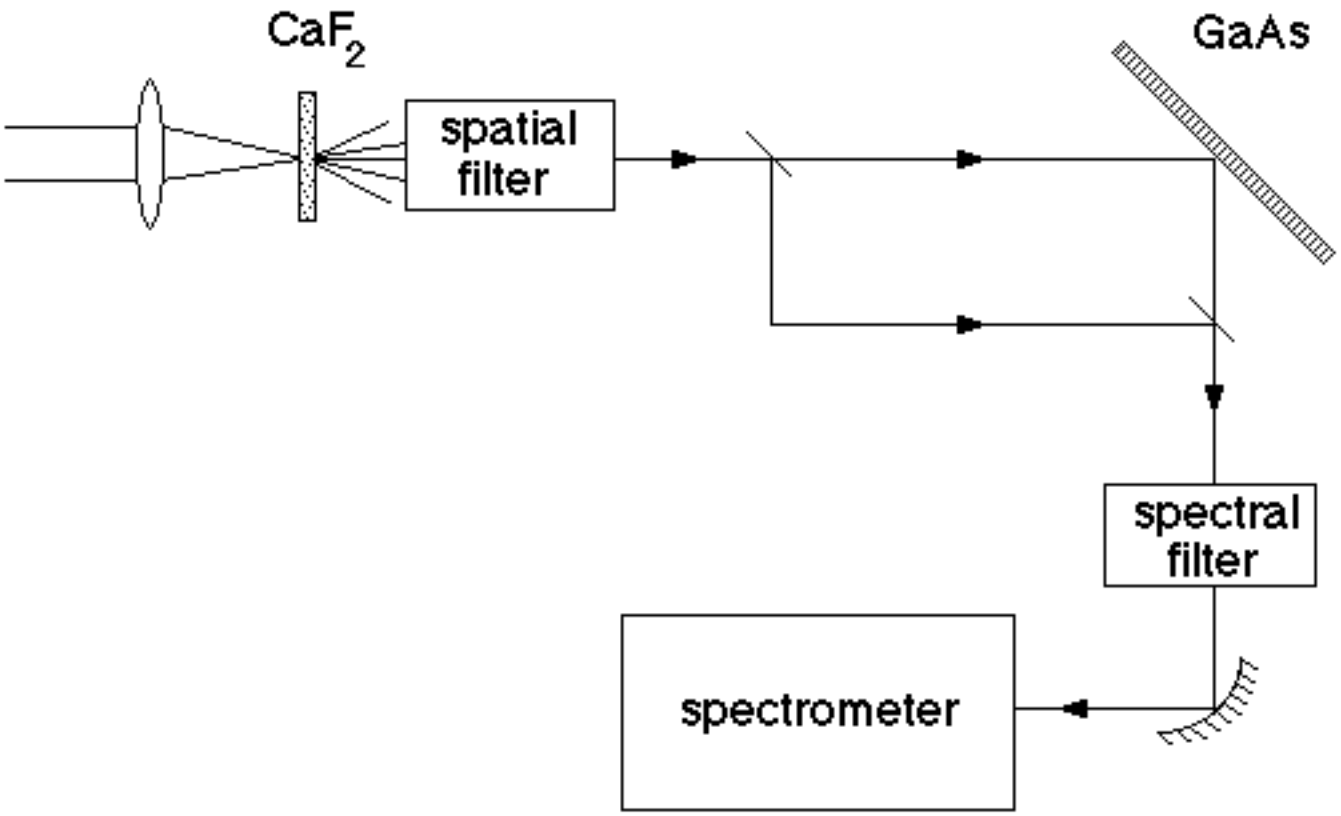
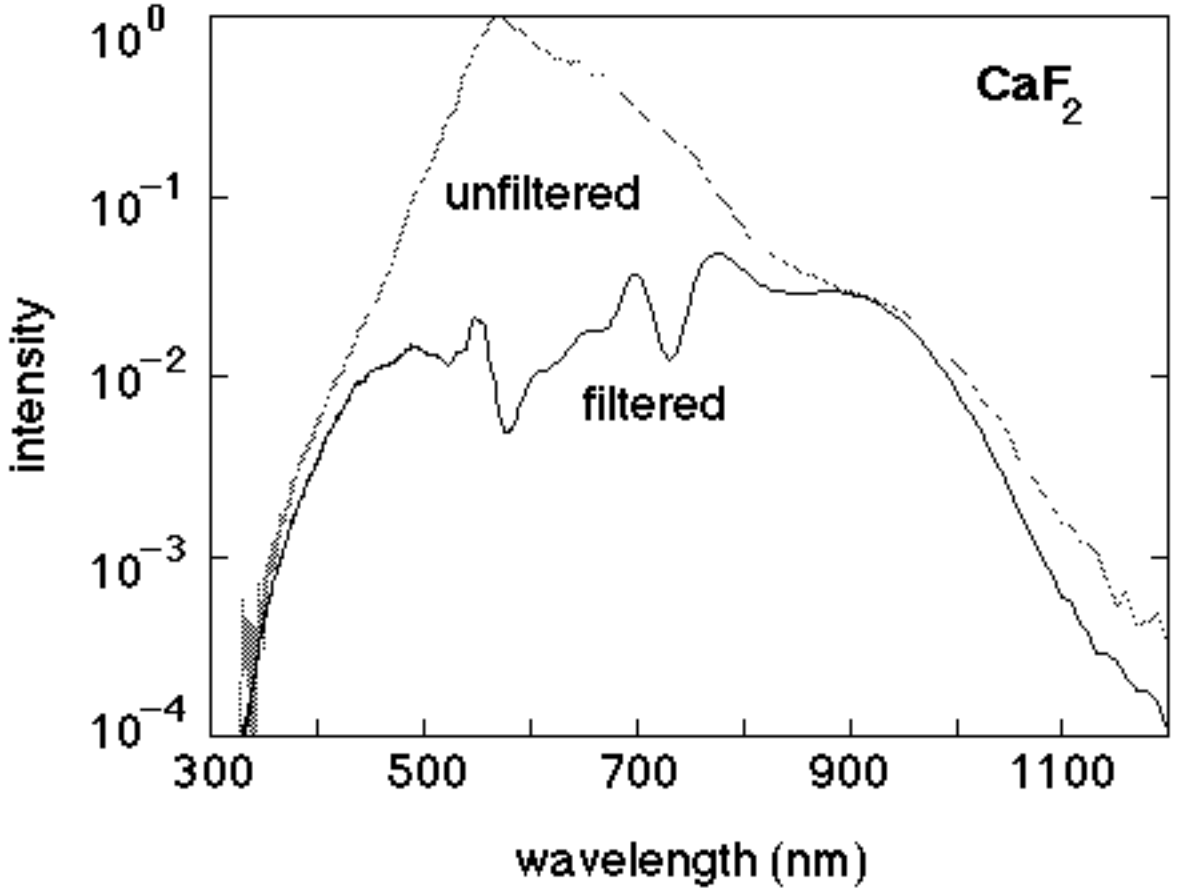


WHITE LIGHT SPECTRUM



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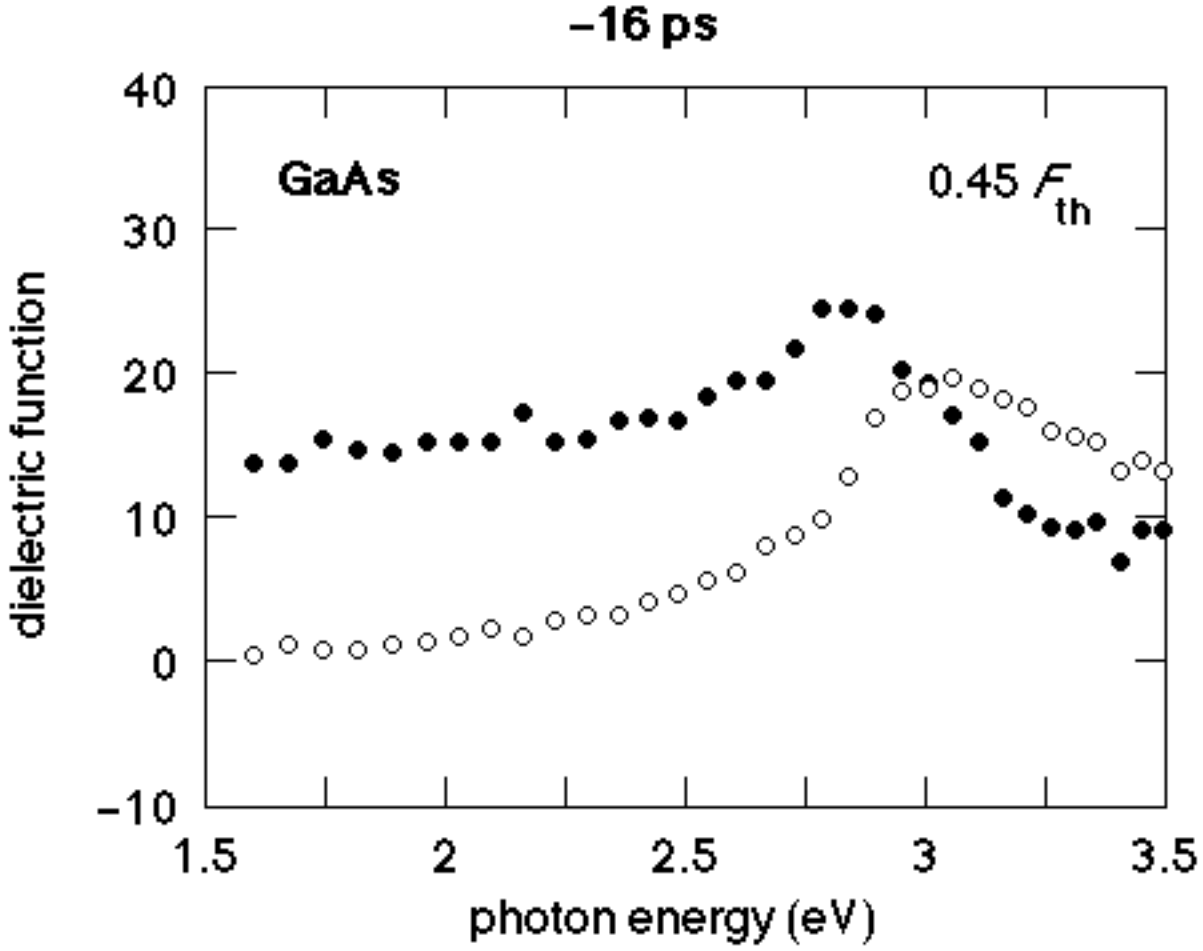

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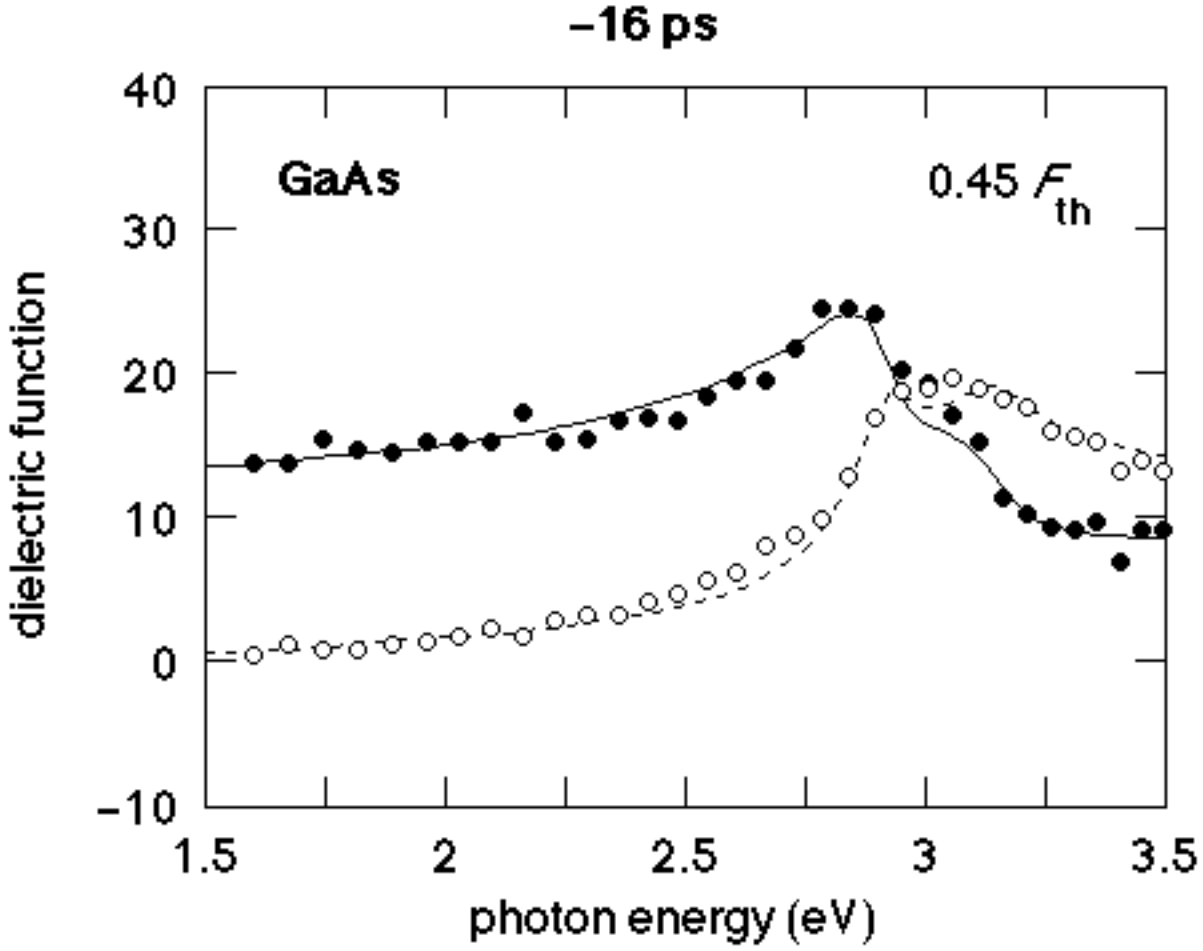

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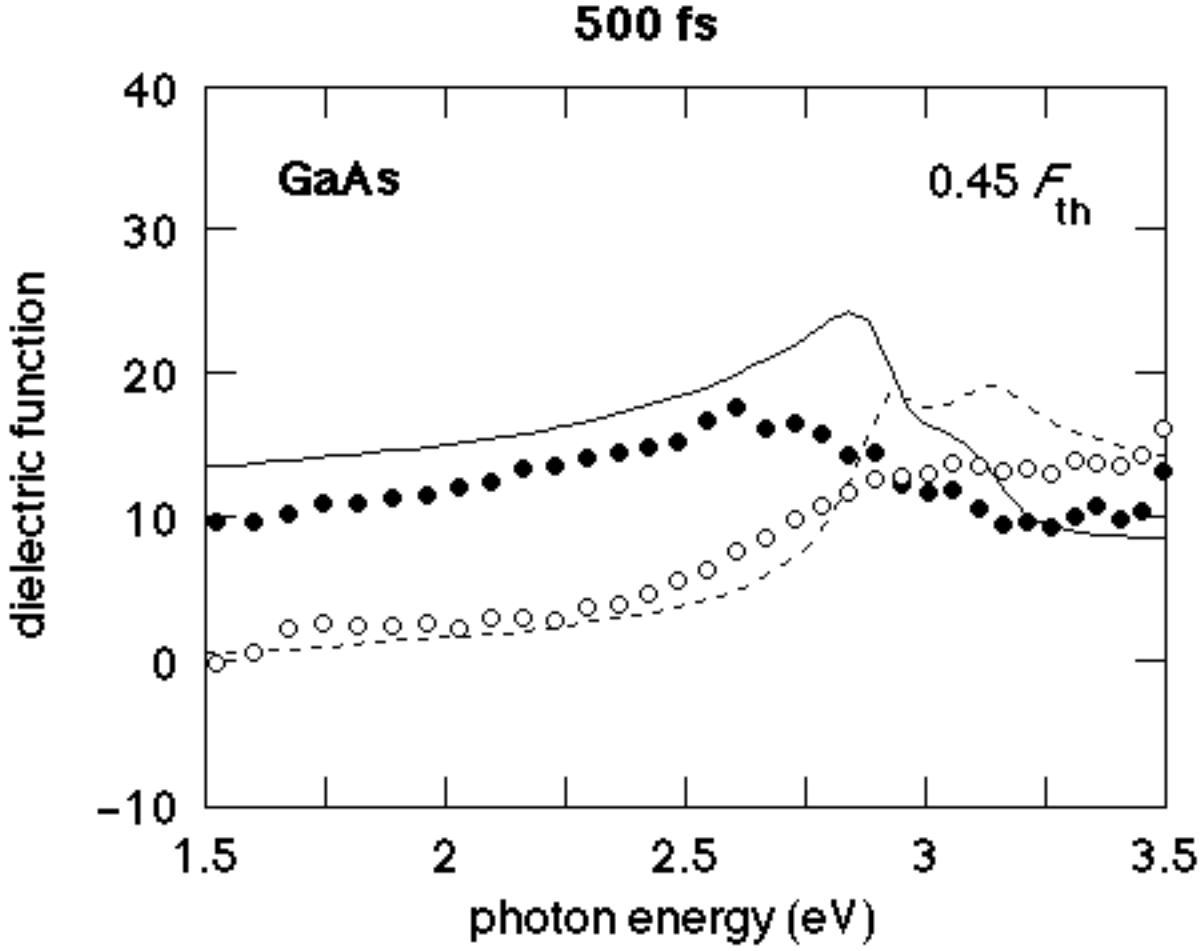

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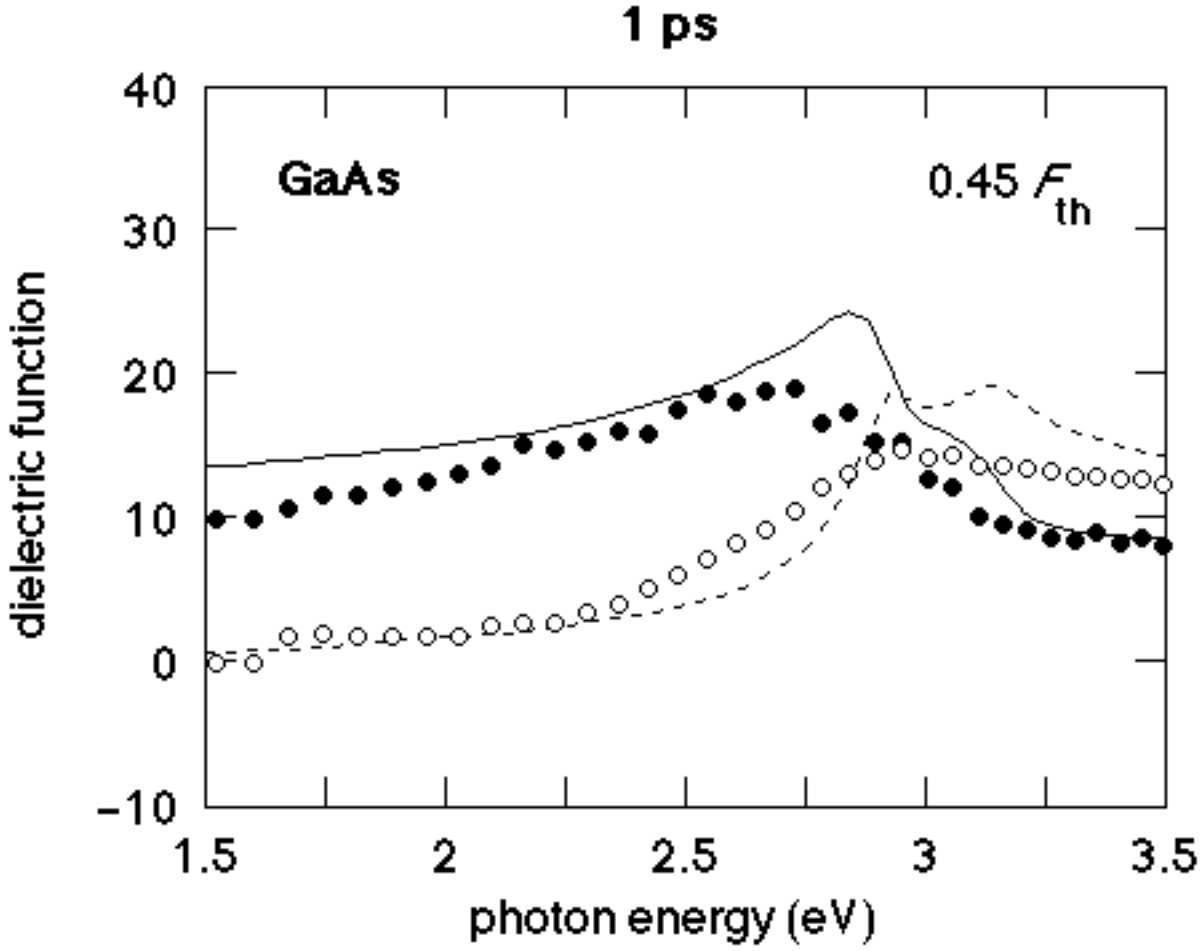

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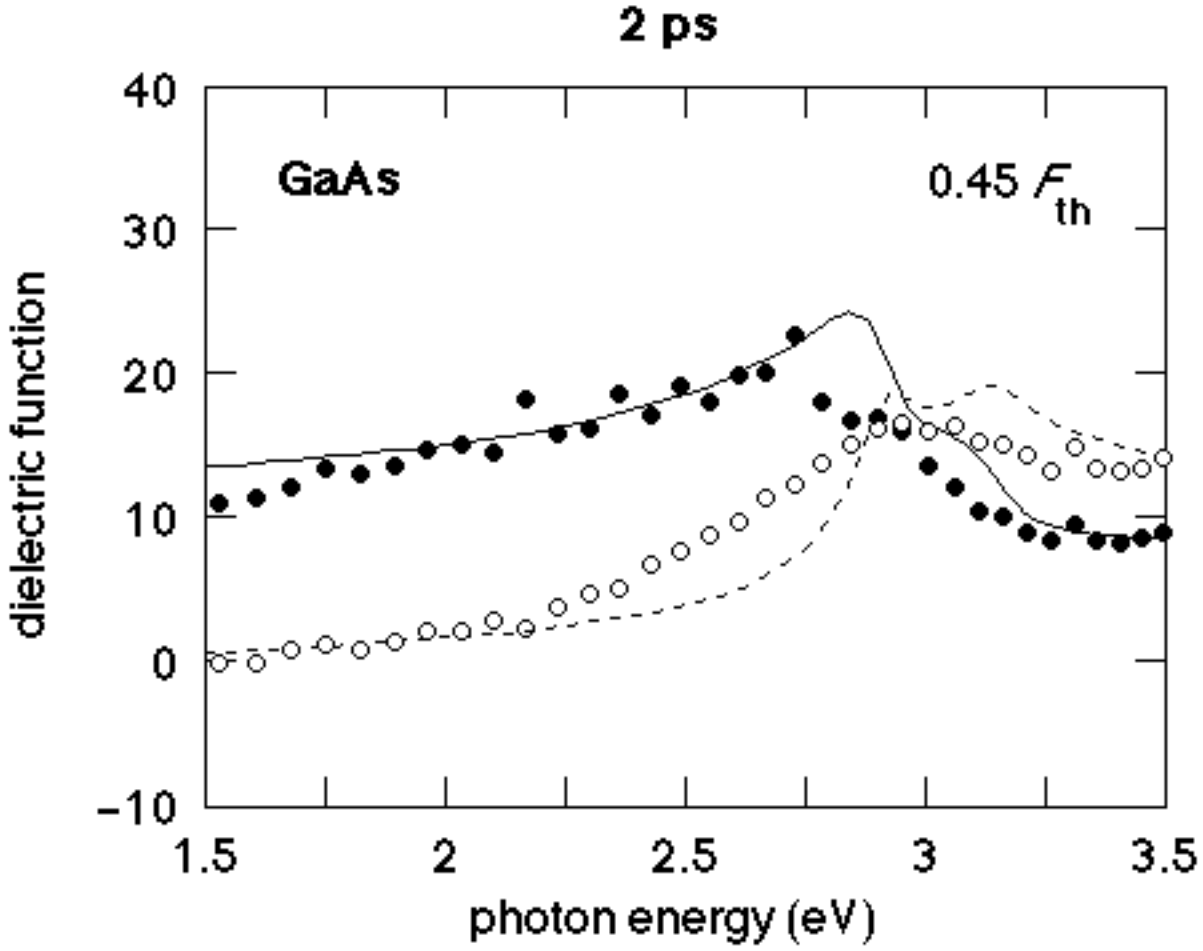

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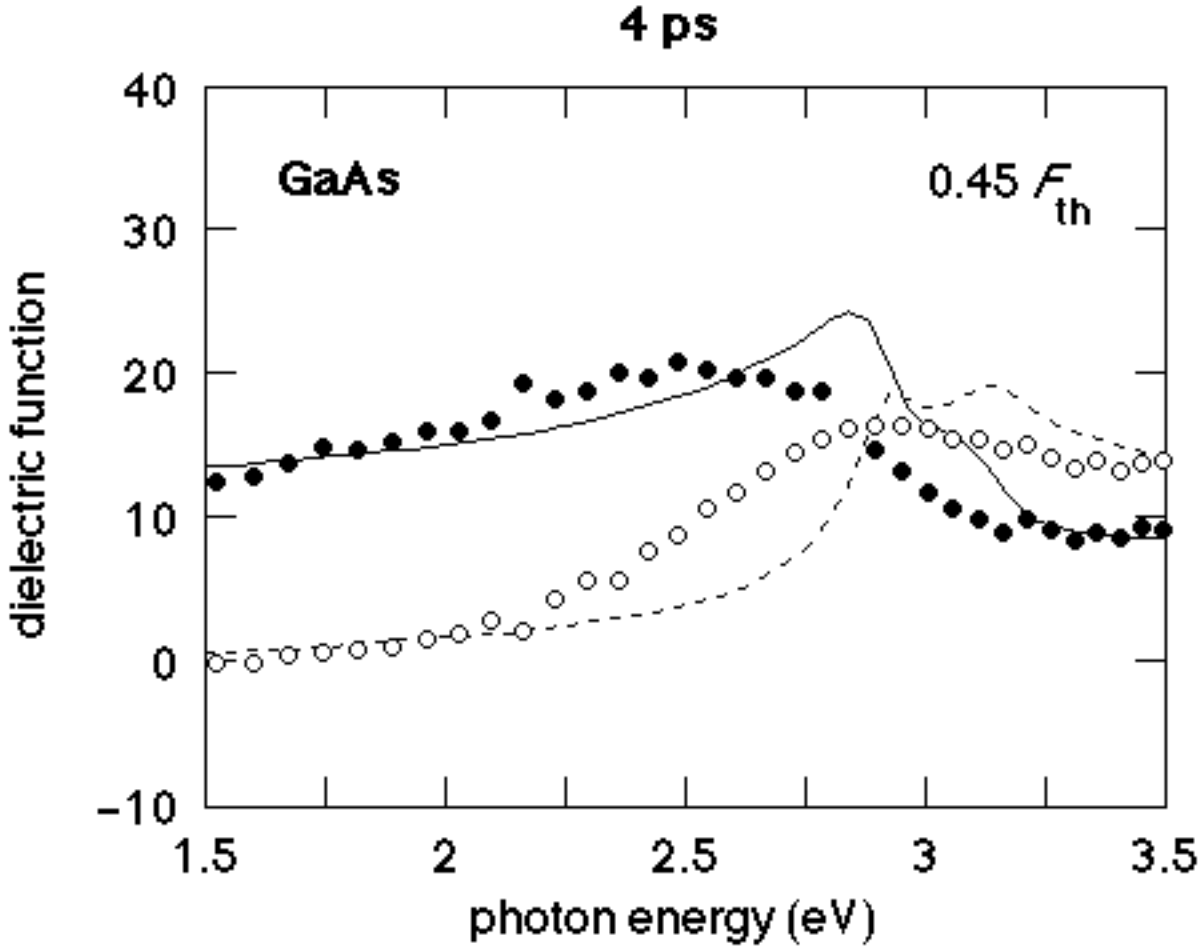

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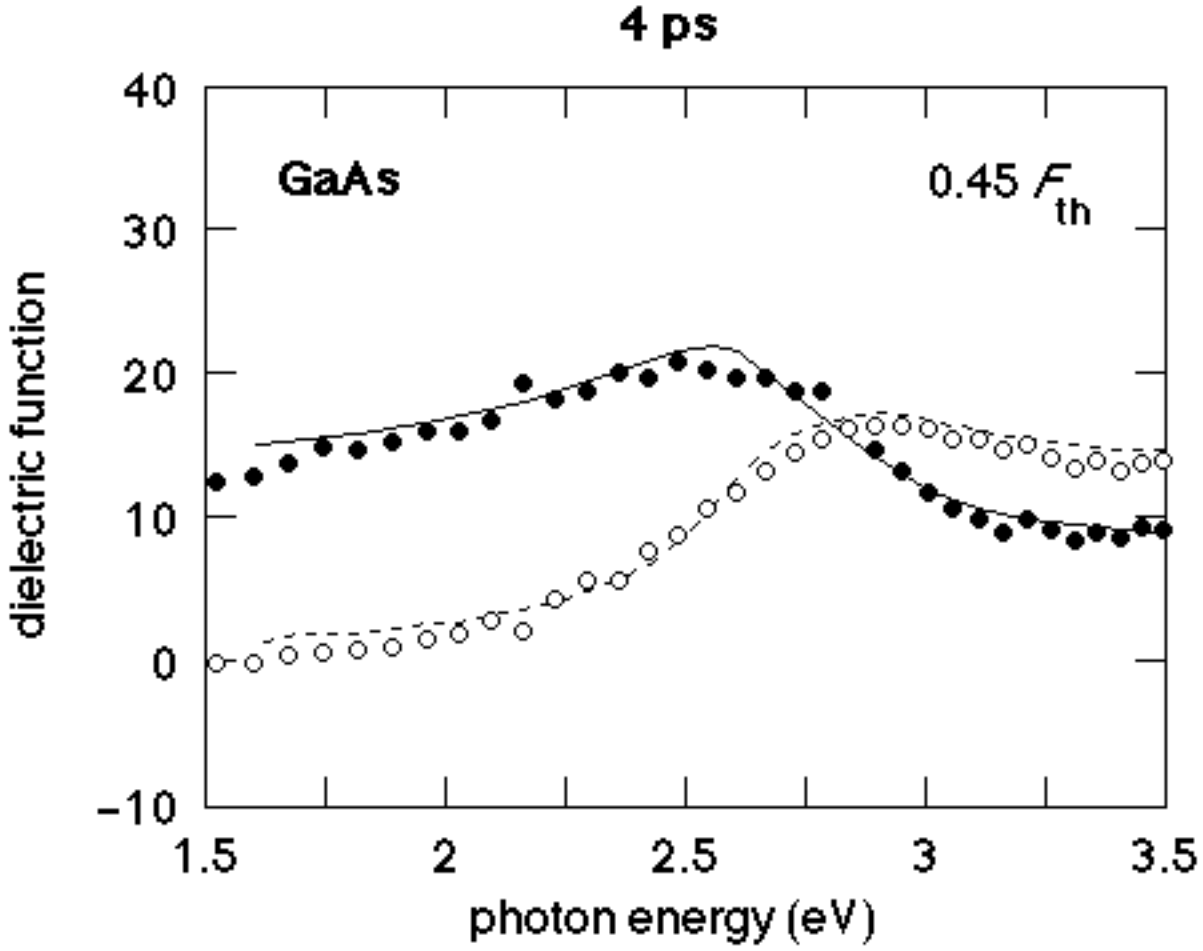

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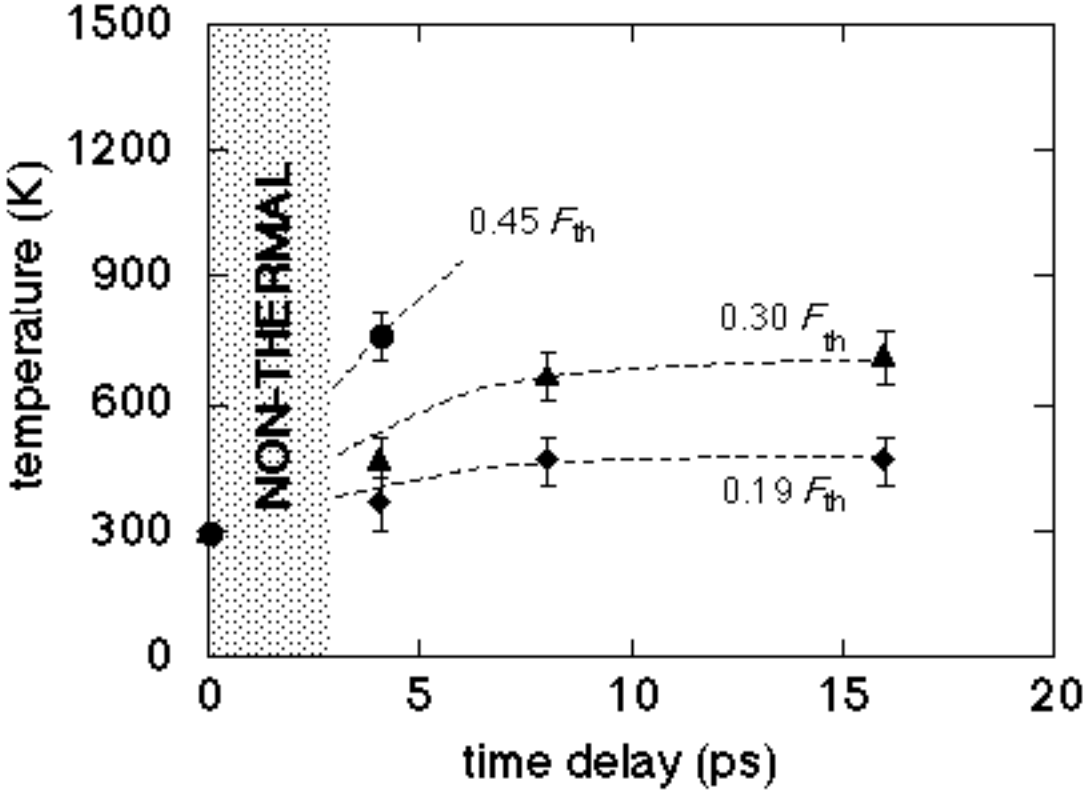

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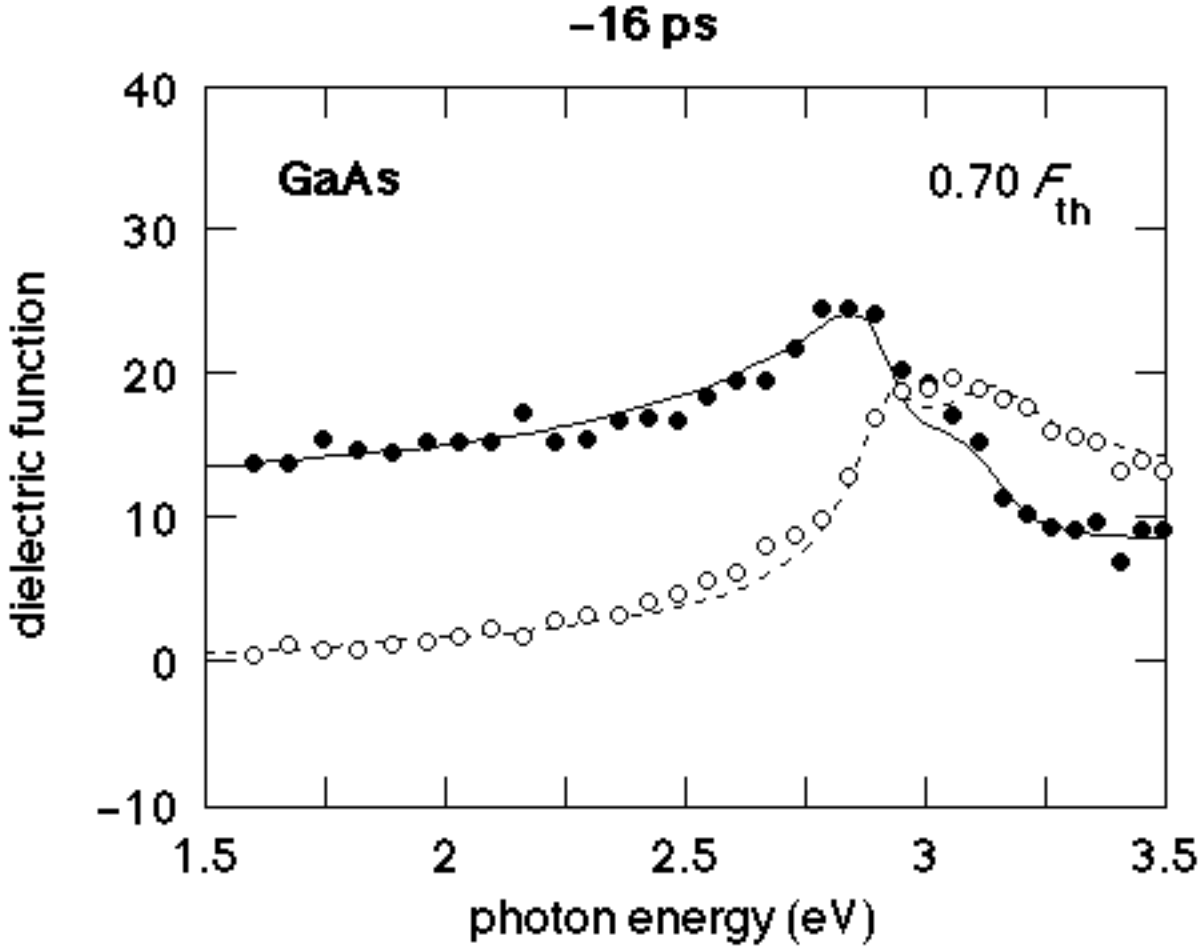

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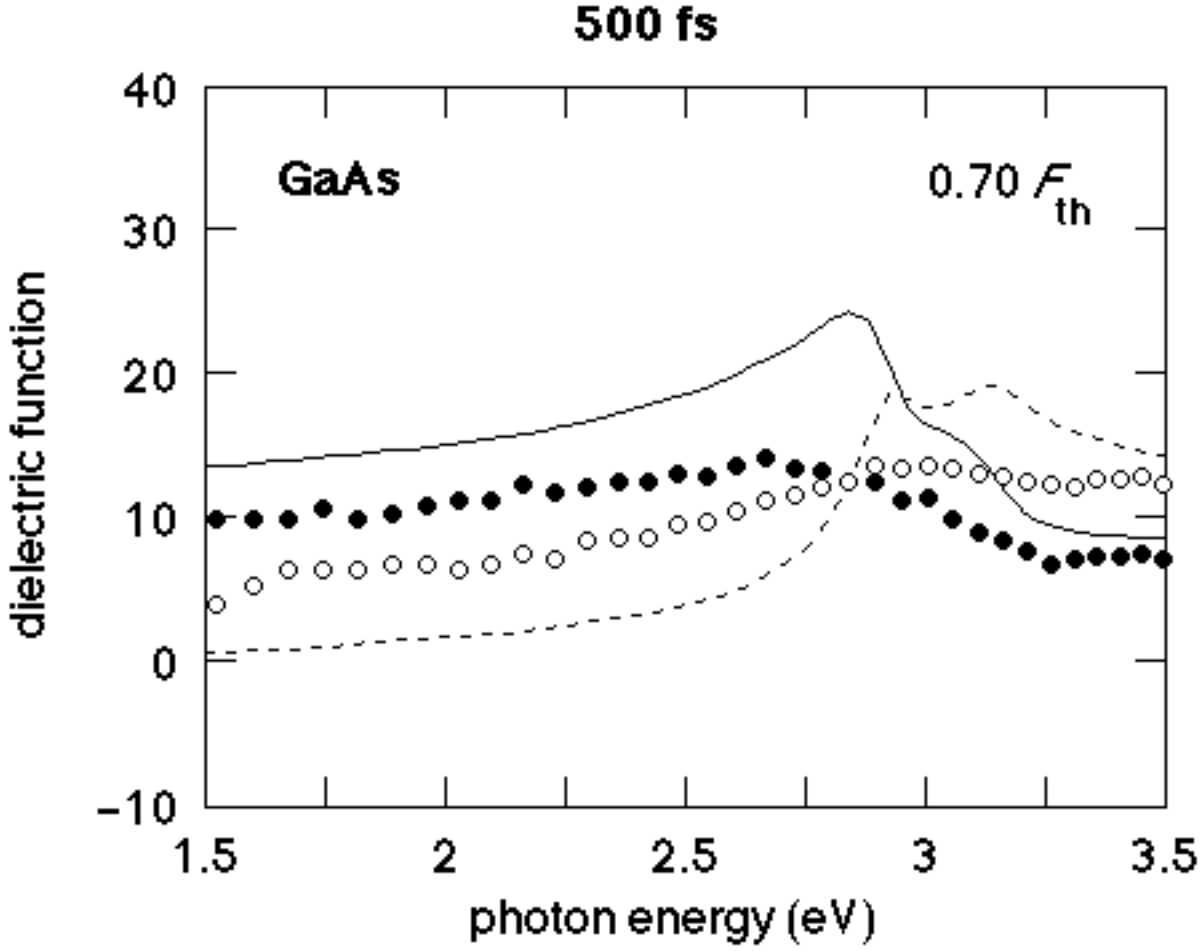

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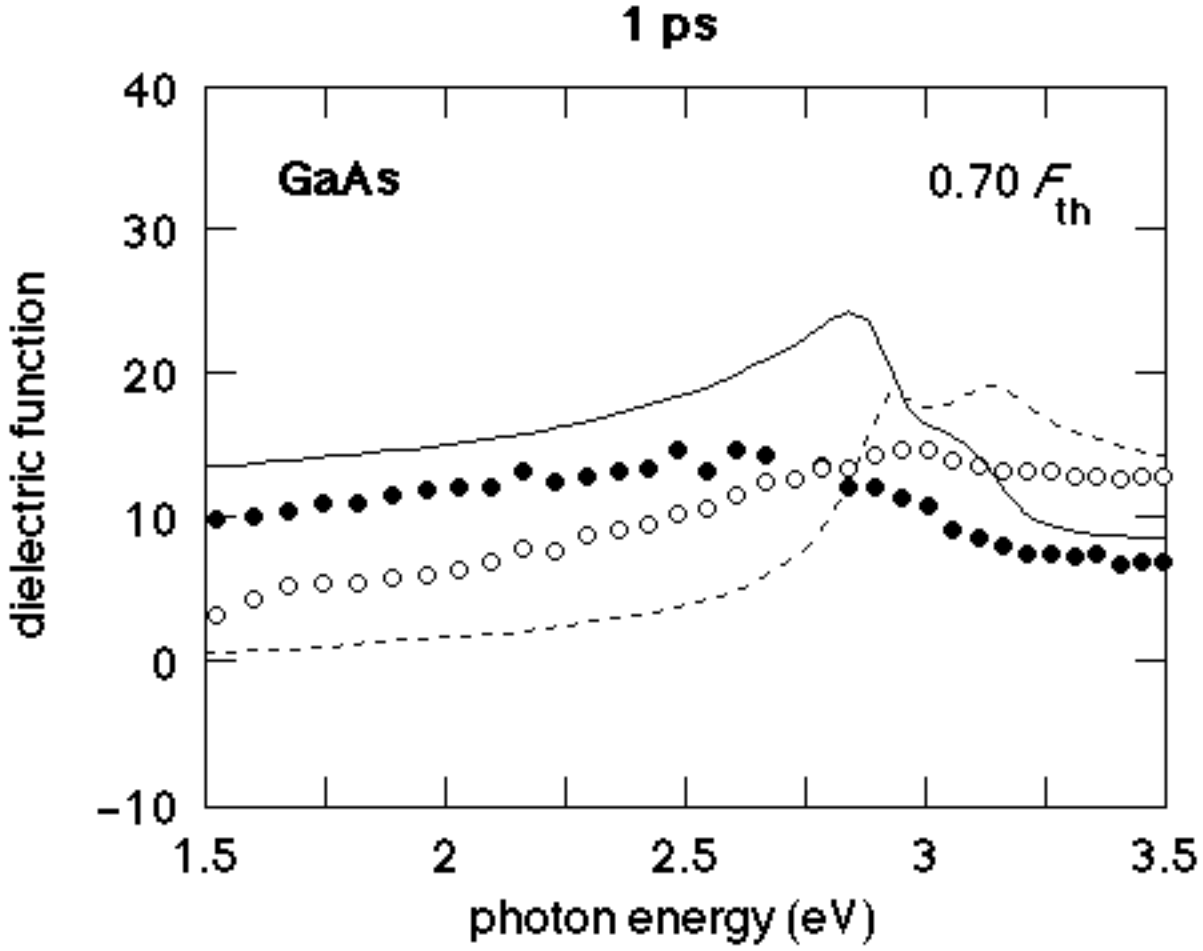

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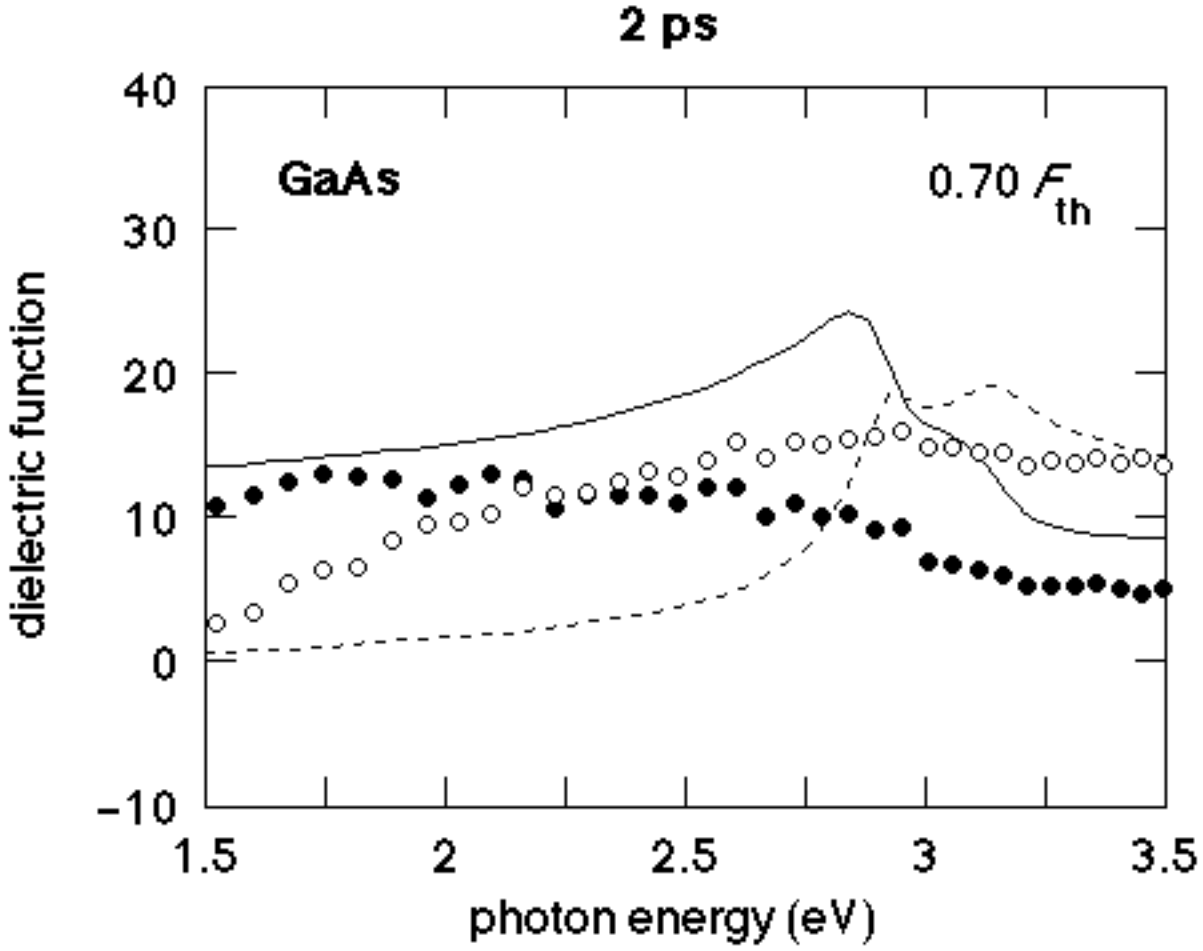

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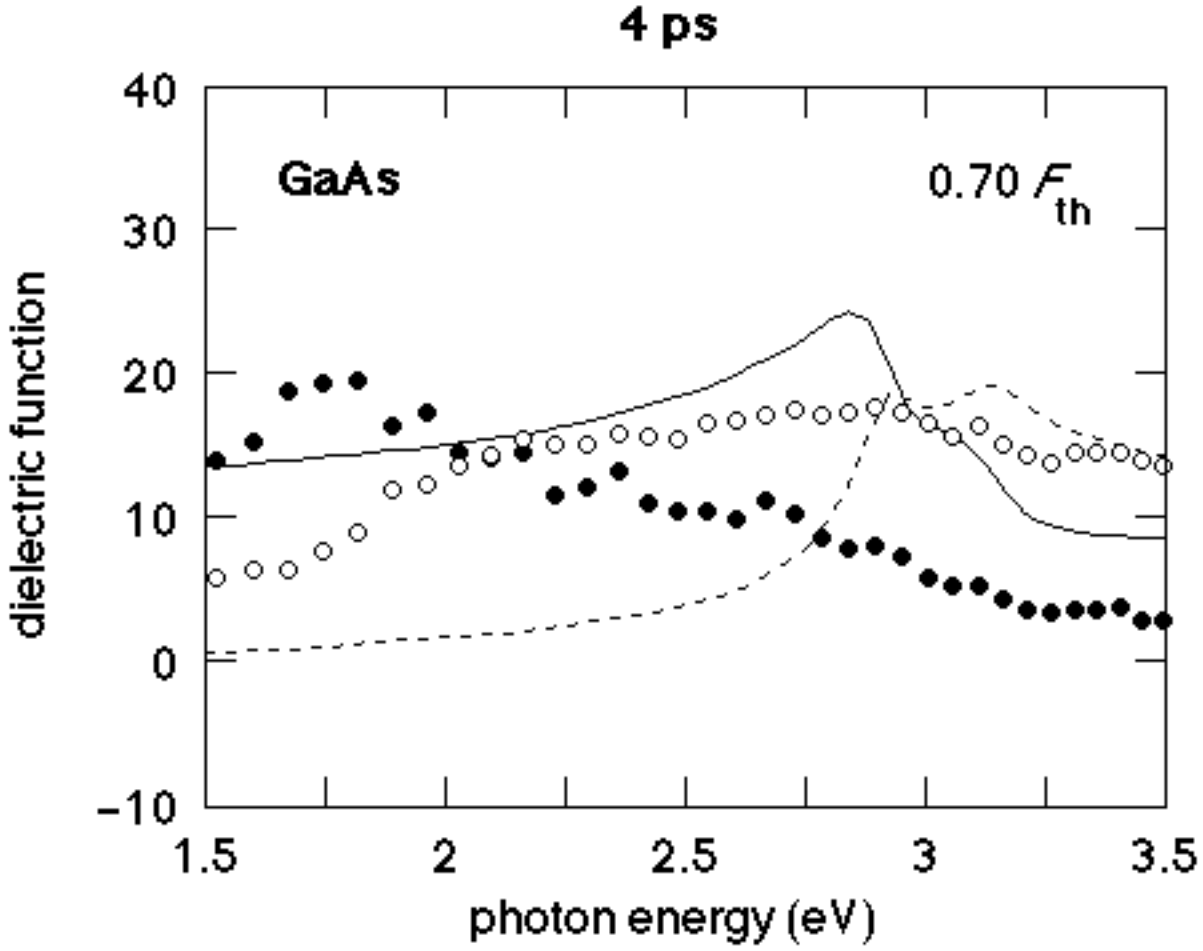

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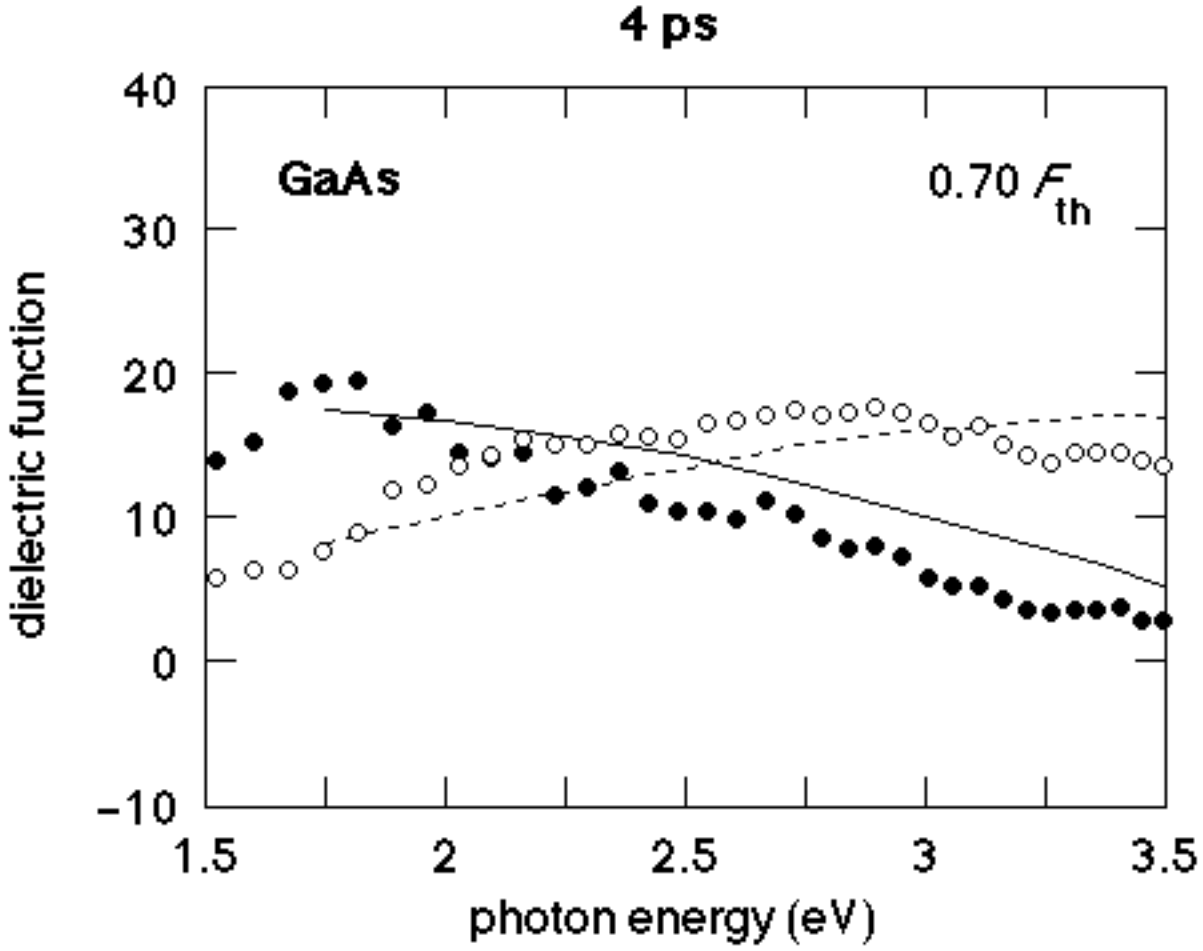

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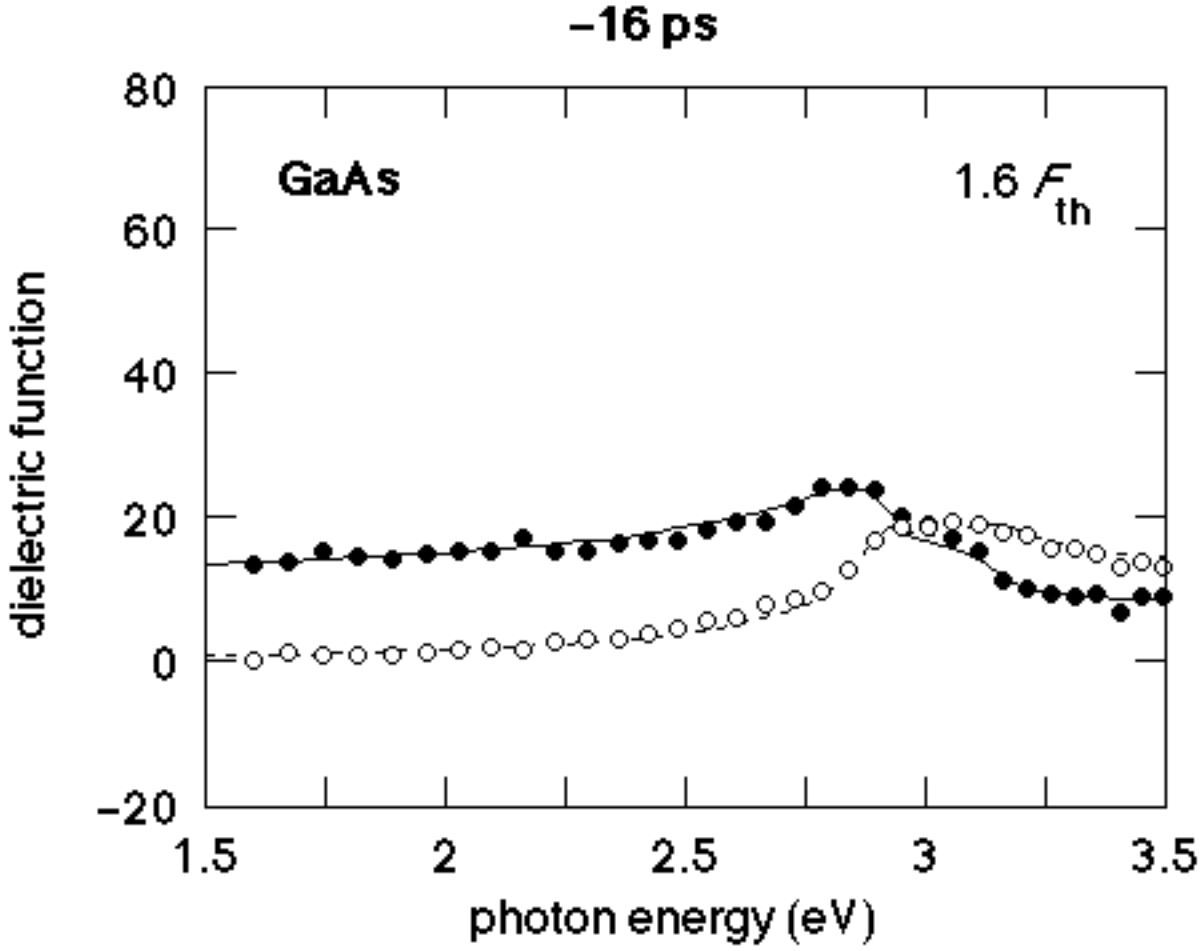

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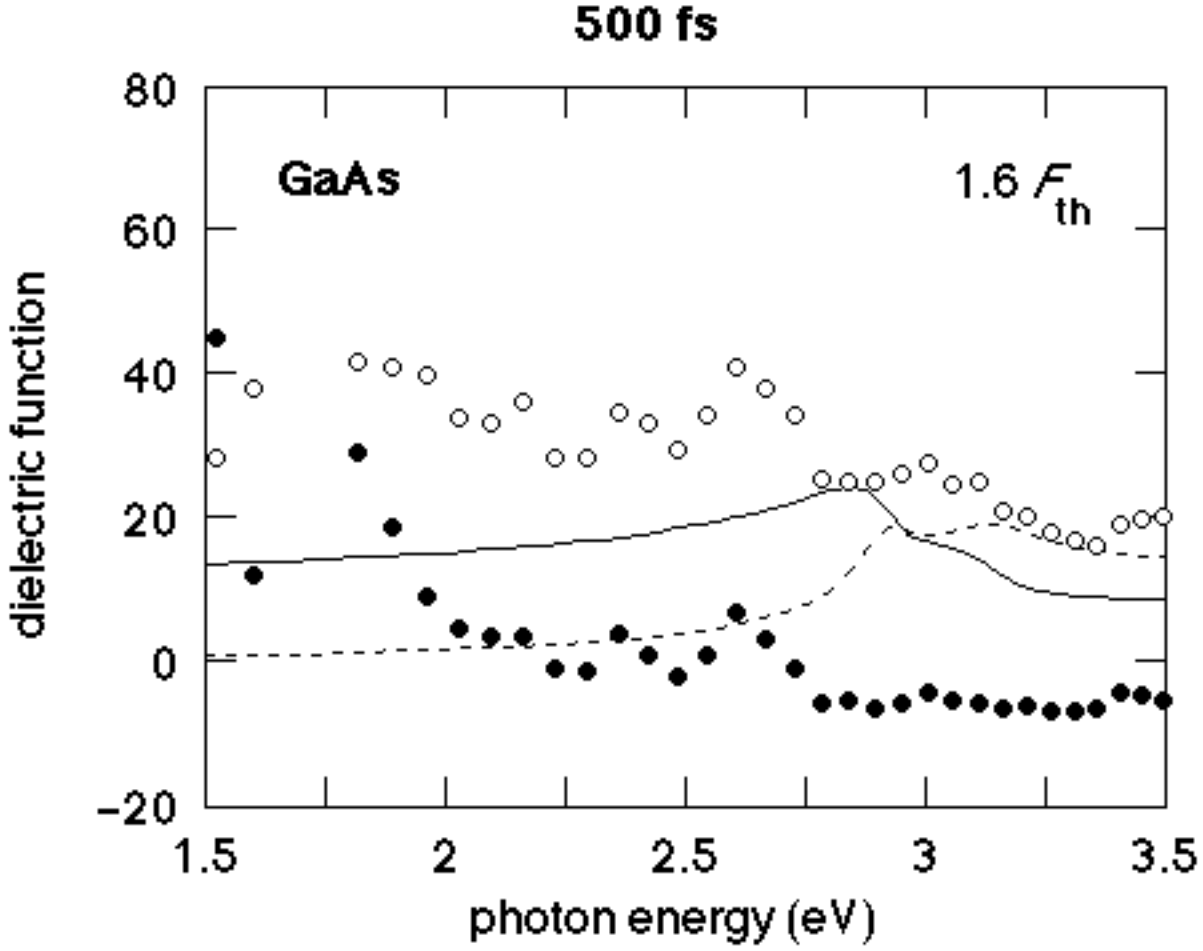

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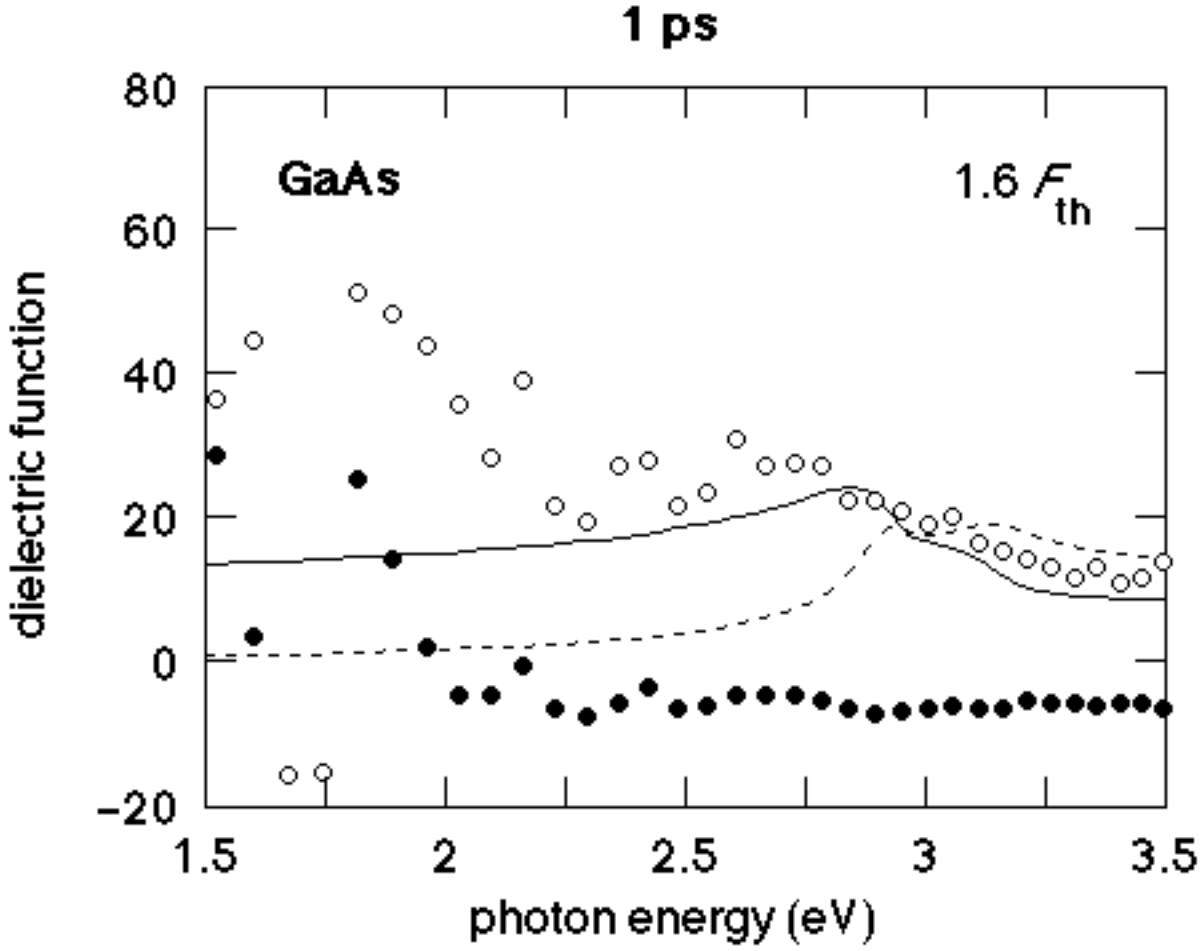

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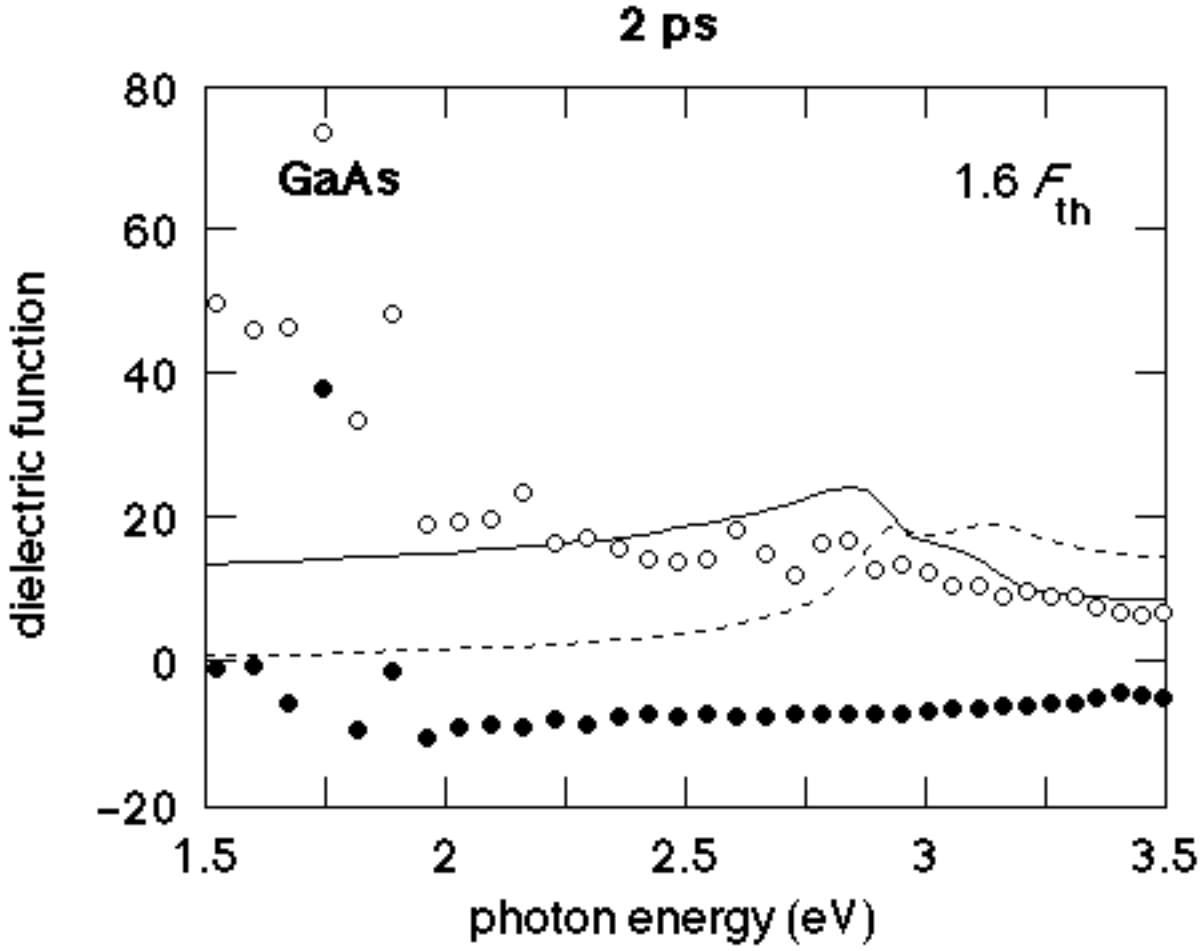

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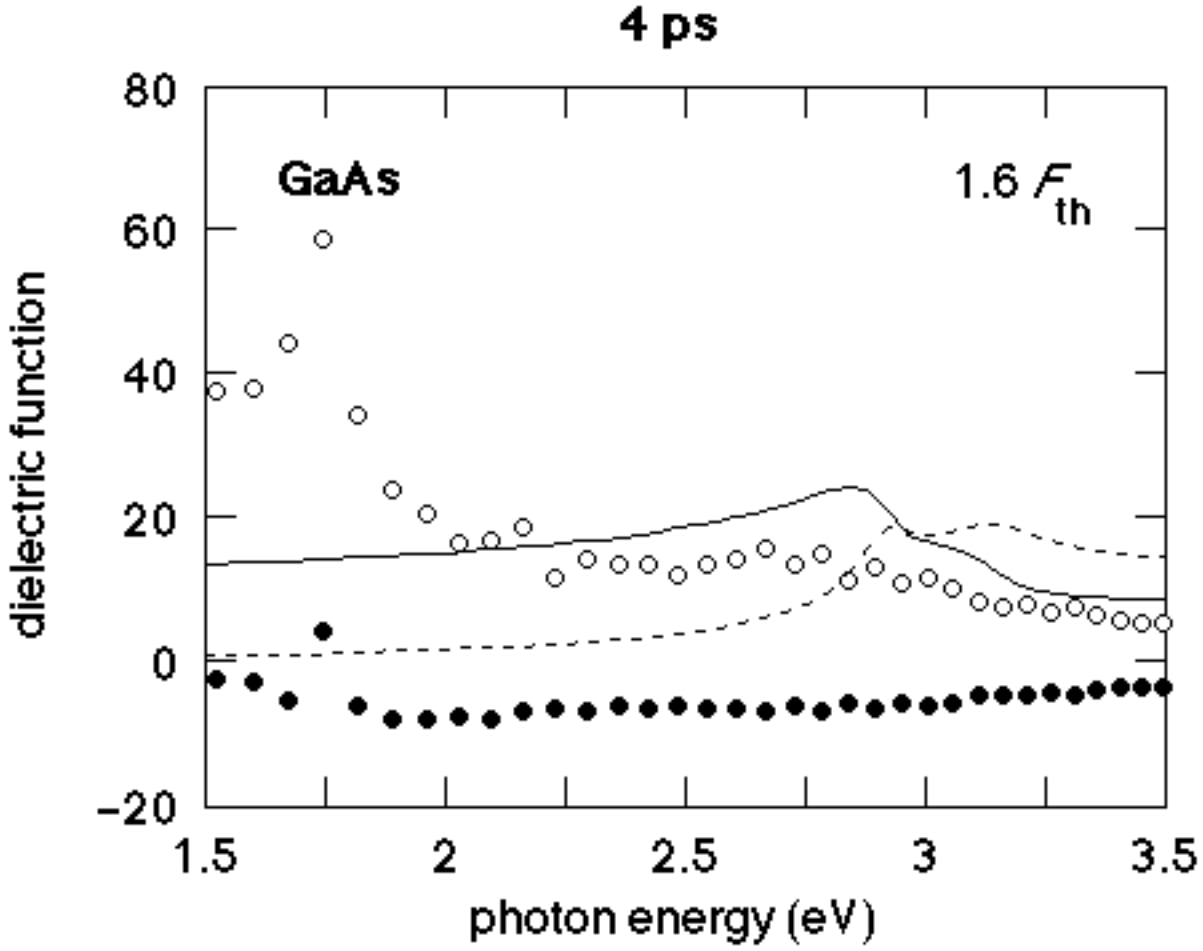

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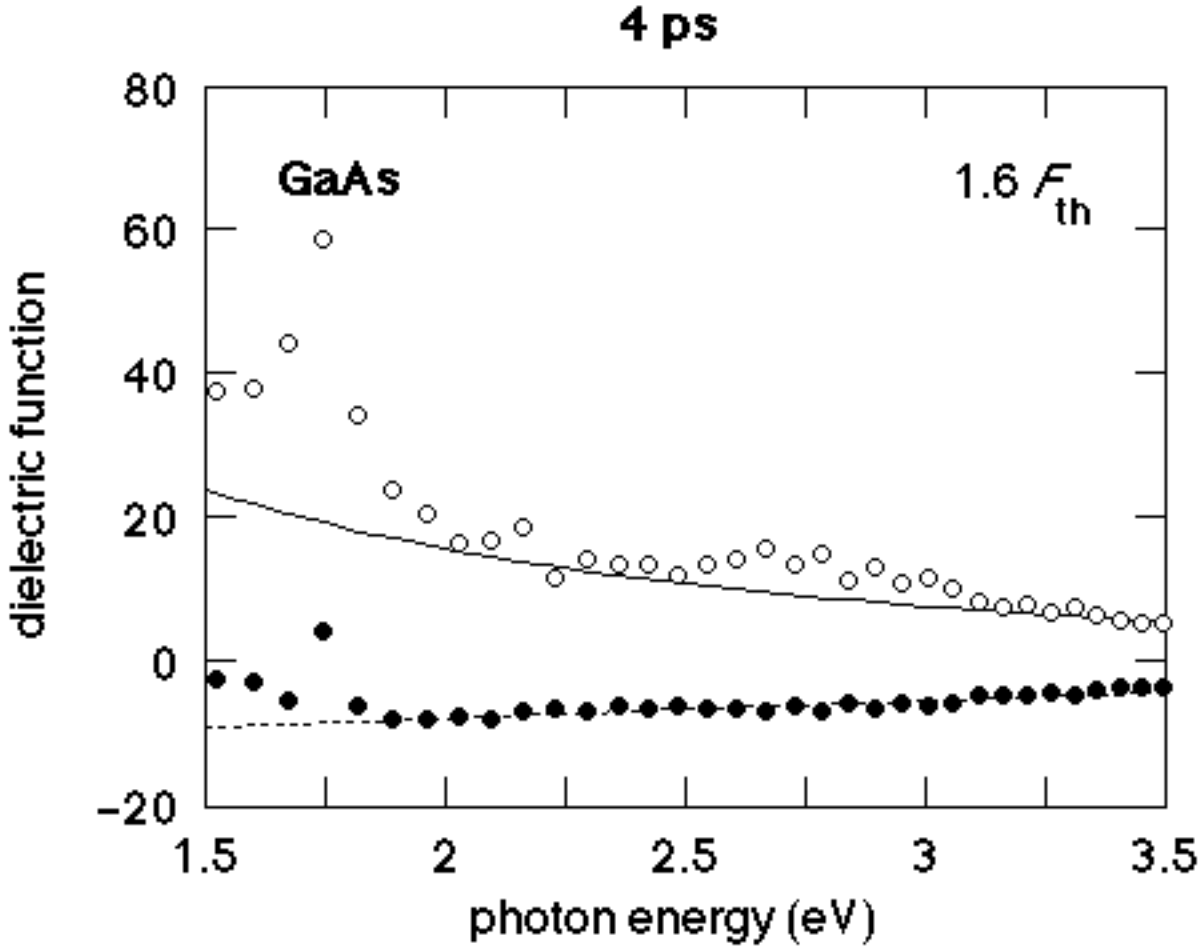

BROADBAND DATA

	short time	long time
low fluence		
medium fluence		
high fluence		

BROADBAND DATA

	short time	long time
low fluence	electronic many-particle effects	
medium fluence		
high fluence		

BROADBAND DATA

	short time	long time
low fluence	electronic many-particle effects	
medium fluence		
high fluence	structural effect?	

BROADBAND DATA

	short time	long time
low fluence	electronic many-particle effects	hot lattice
medium fluence		
high fluence	structural effect?	

BROADBAND DATA

	short time	long time
low fluence	electronic many-particle effects	hot lattice
medium fluence		lattice disordering
high fluence	structural effect?	

BROADBAND DATA

	short time	long time
low fluence	electronic many-particle effects	hot lattice
medium fluence		lattice disordering
high fluence	structural effect?	metallic behavior

electronic excitation can drive phonons incoherently, coherently, or impulsively

electronic excitation can drive a structural transition

femtosecond lasers allow us to see the dynamics of the transition

Acknowledgments

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

ONR N00014-89-J1023
NSF DMR 89-20490

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