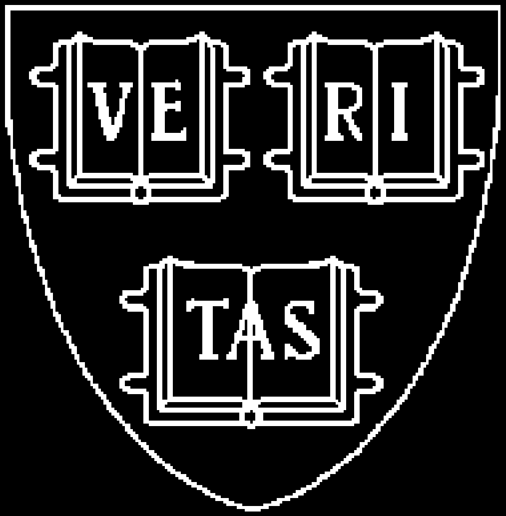

VE RI
TAS

PHASE TRANSITIONS INDUCED BY FEMTOSECOND LASER PULSES

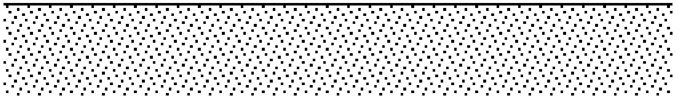
Eli Glezer
Li Huang
Paul Callan
Yakir Siegal

Texas A&M
17 April 1996

INTRODUCTION

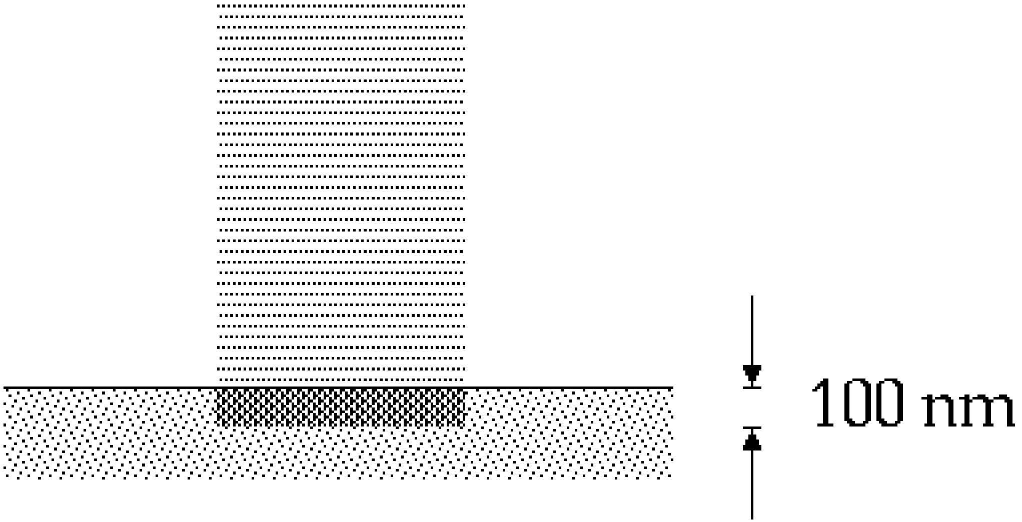
1976: (nanosecond) laser annealing of crystals damaged by ion bombardment

1985: femtosecond laser ‘melting’ of Si



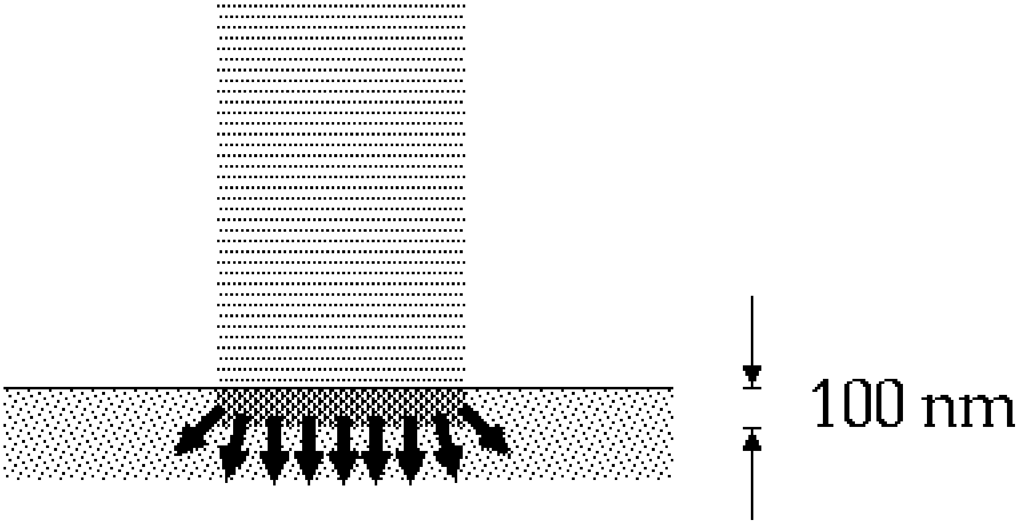
INTRODUCTION

laser deposits energy near surface...



INTRODUCTION

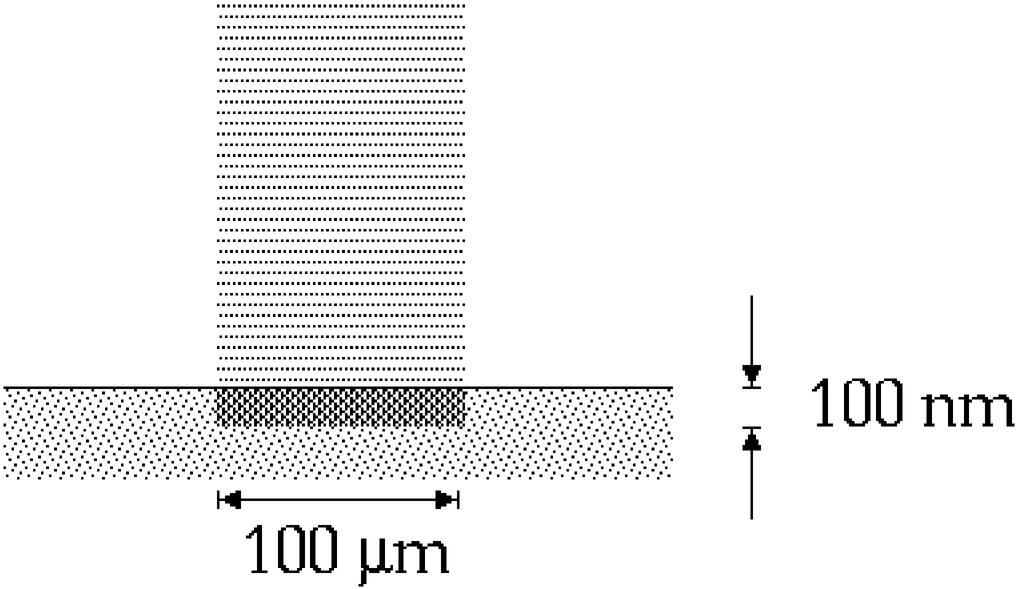
energy rapidly diffuses into bulk...



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!

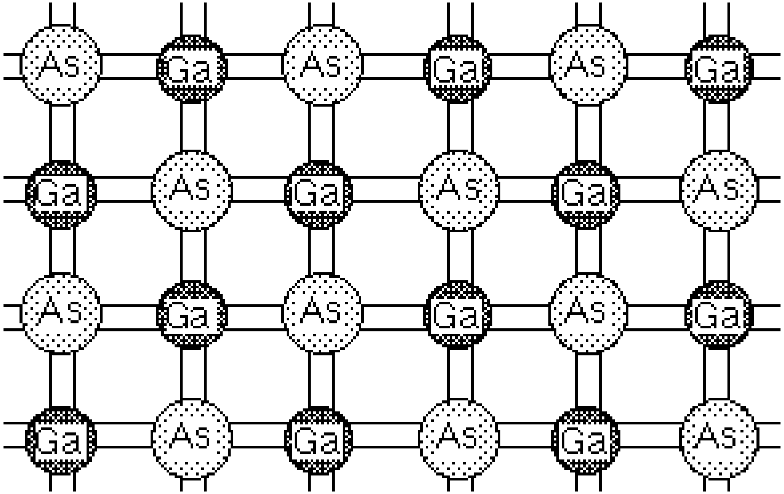
OUTLINE

❶ Background

❷ Femtosecond work

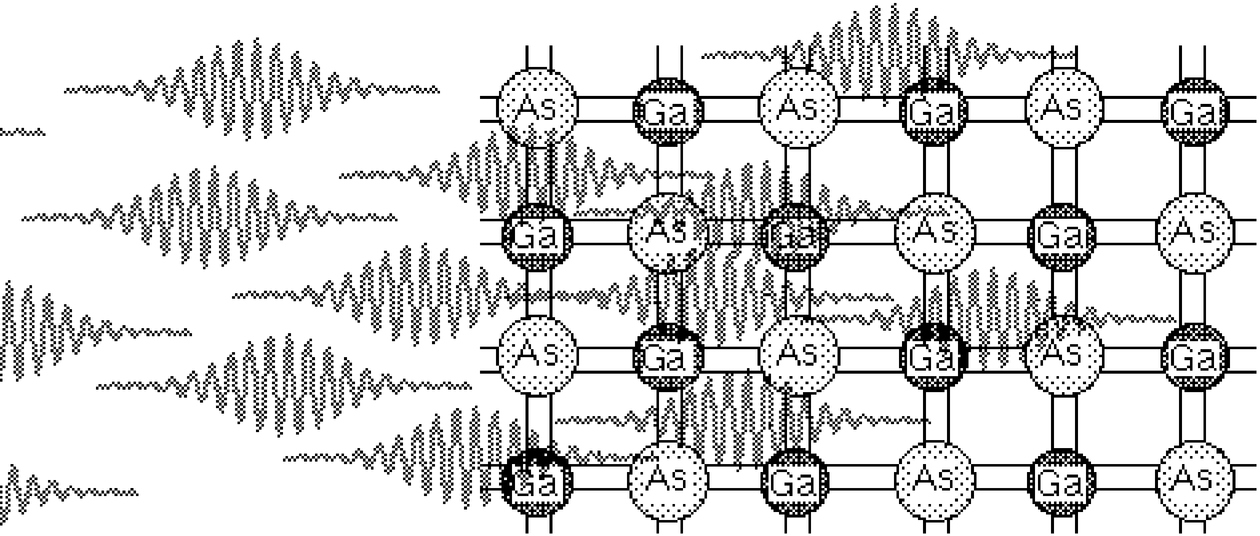
❸ Future

EARLY MODELS



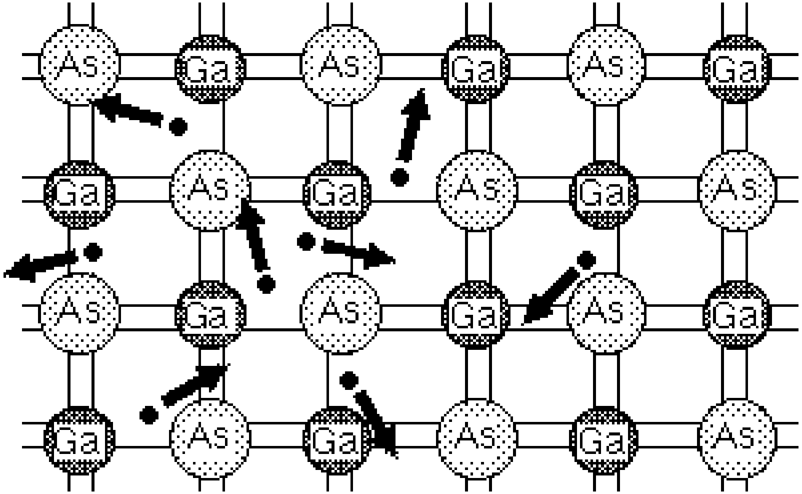
how does light melt a solid?

THERMAL MODEL



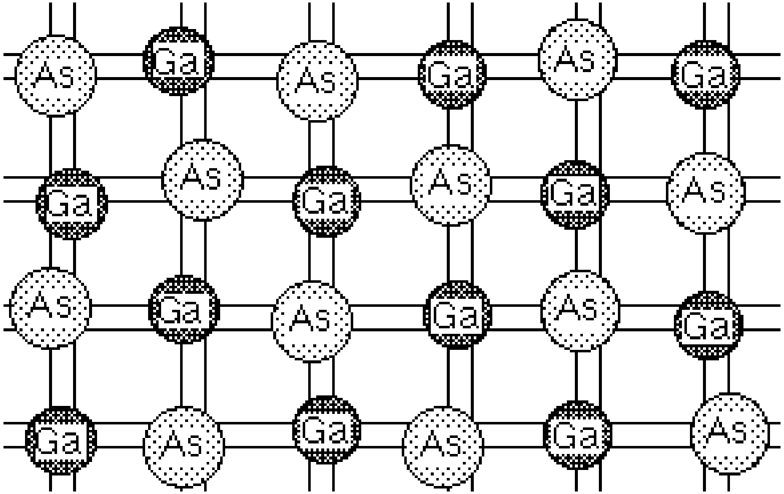
photons excite valence electrons...

THERMAL MODEL



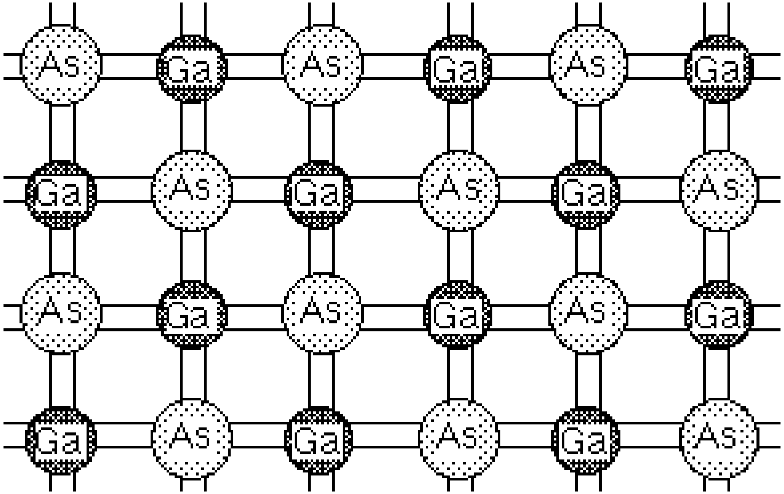
...and create hot electrons...

THERMAL MODEL



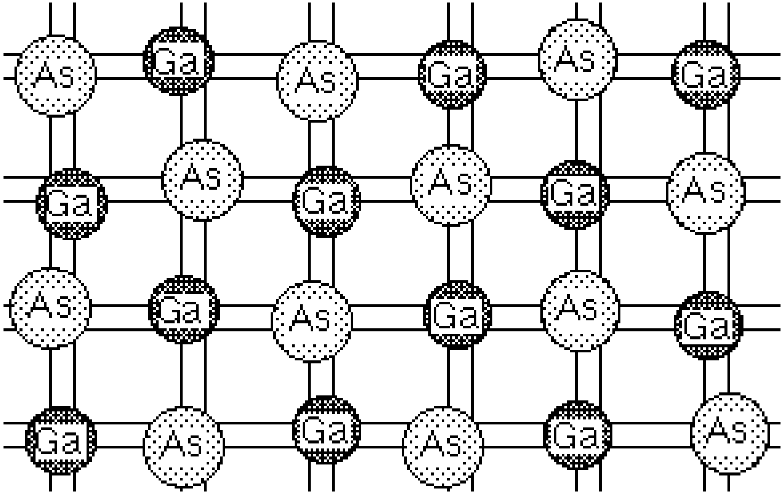
...which heat lattice

THERMAL MODEL



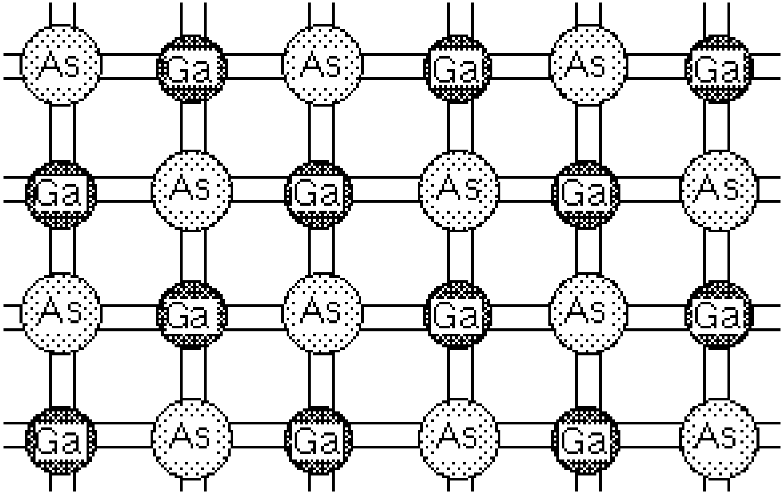
...which heat lattice

THERMAL MODEL



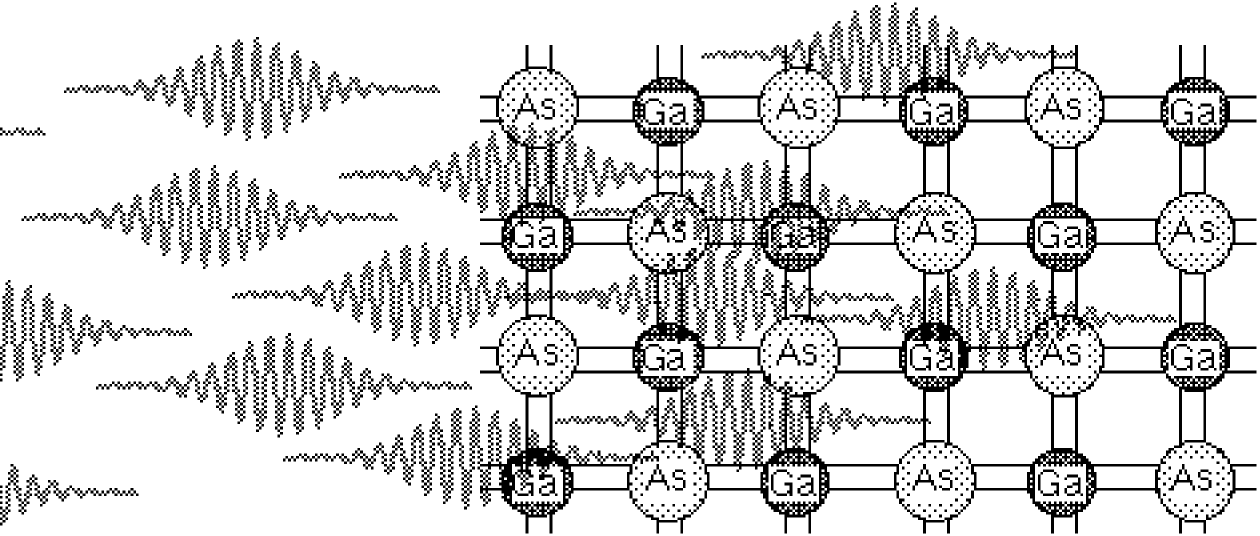
...which heat lattice

EARLY MODELS



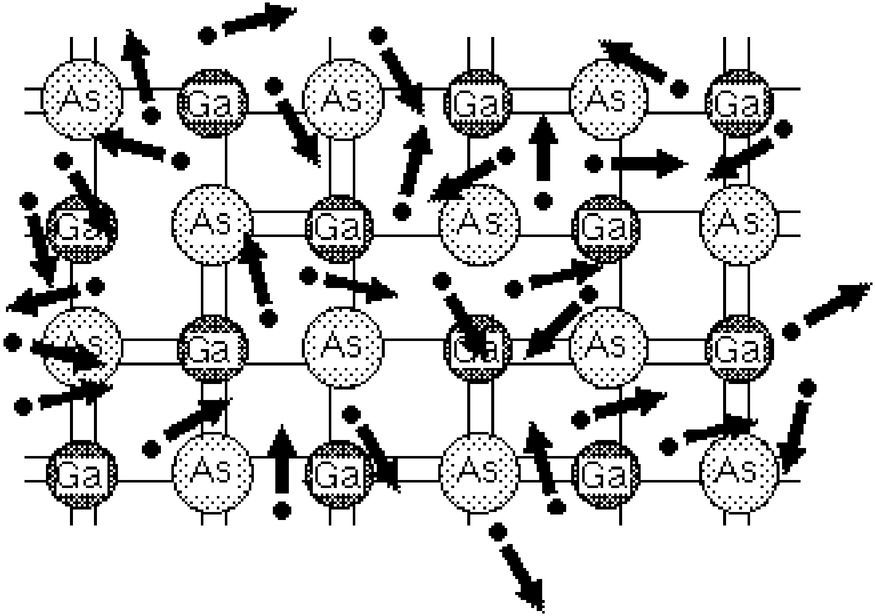
...or...

PLASMA ANNEALING



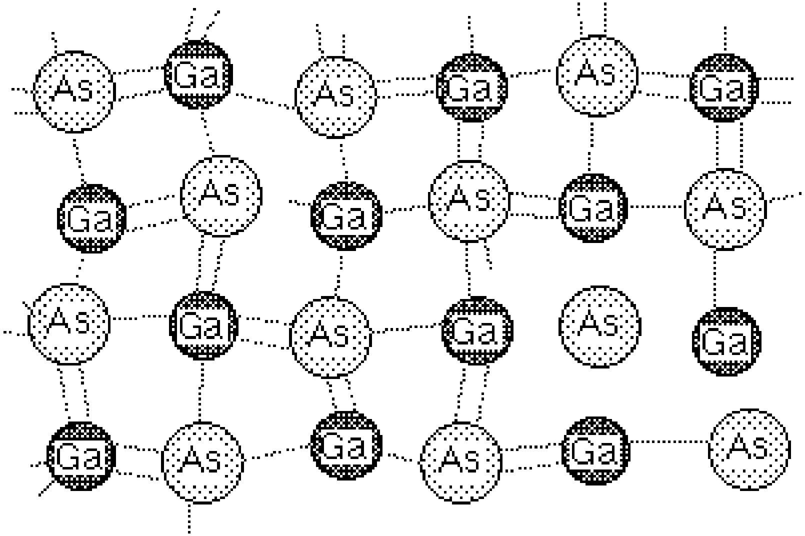
photons excite valence electrons...

PLASMA ANNEALING



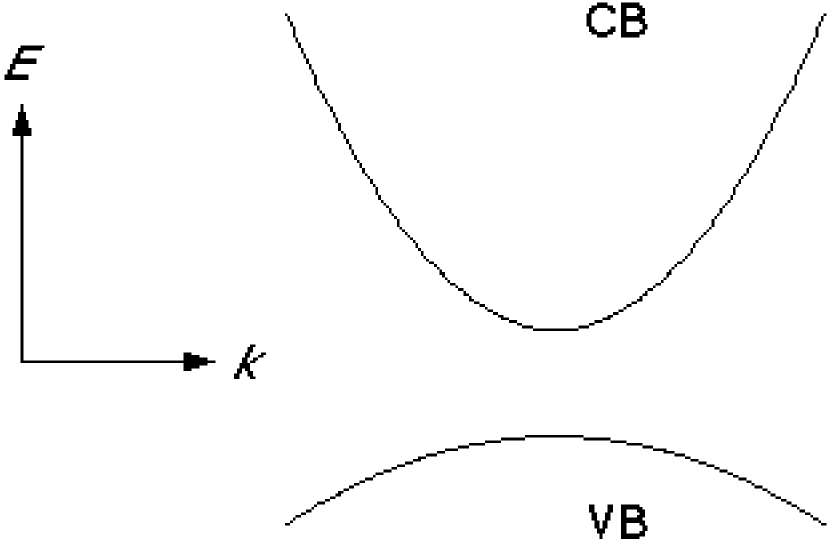
...creating a dense plasma...

PLASMA ANNEALING

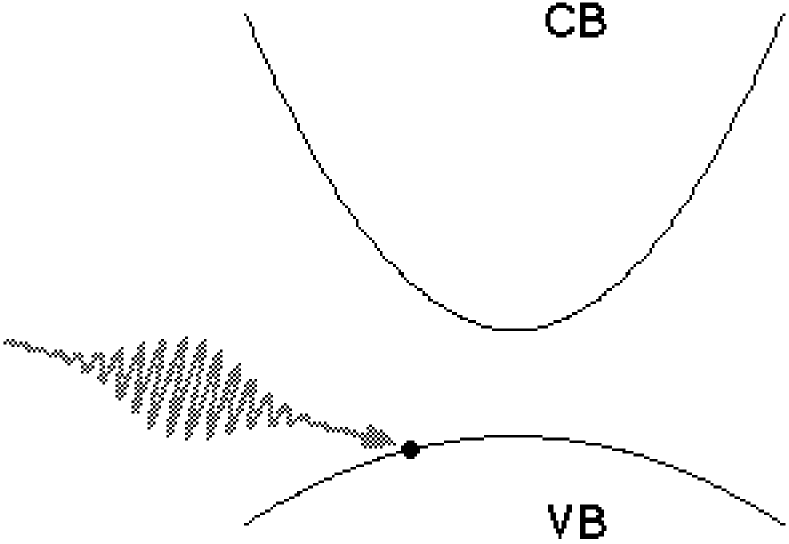


...and breaking bonds

EXCITATION

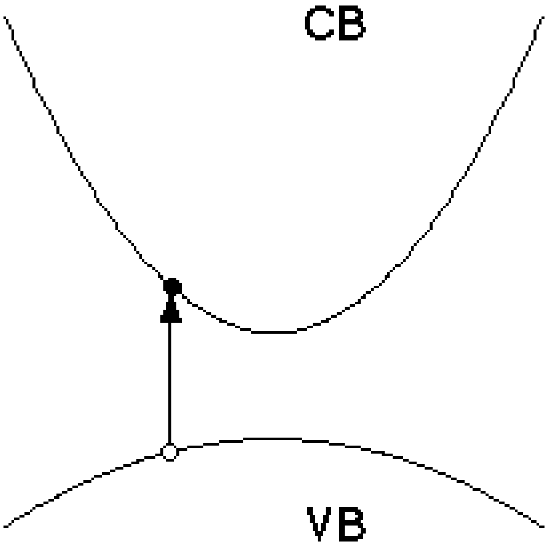


EXCITATION



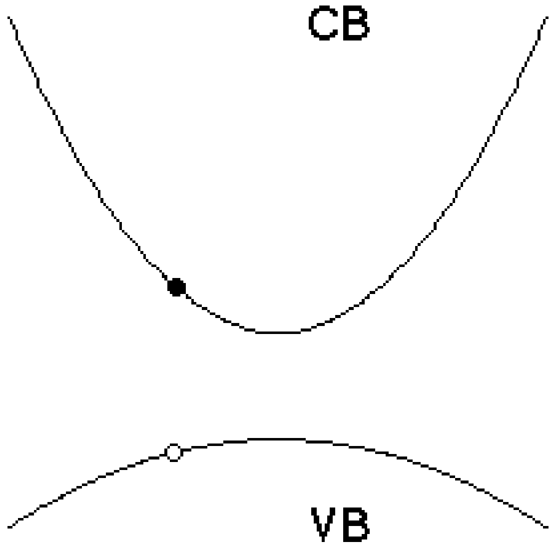
photon excites valence electron

EXCITATION

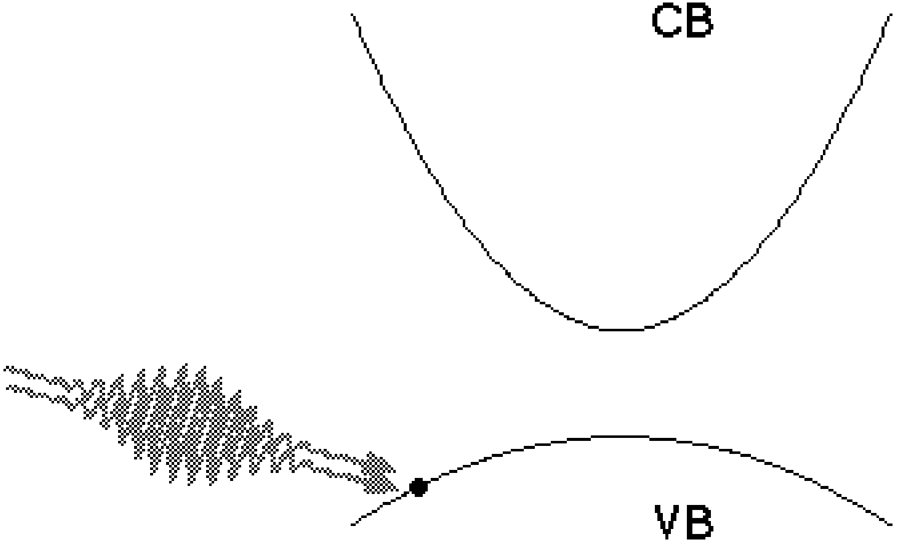


creating an electron-hole pair

EXCITATION

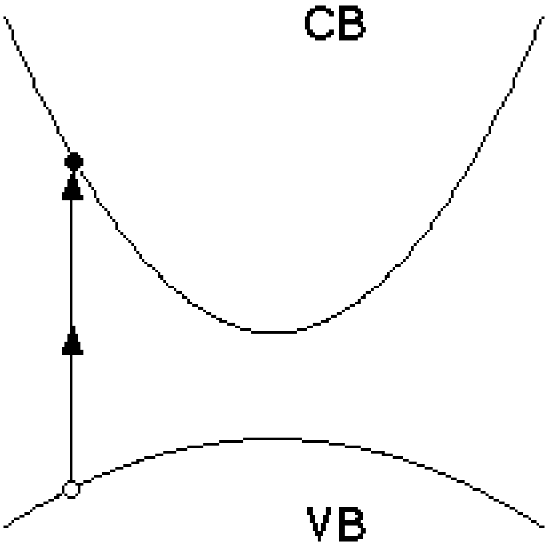


EXCITATION

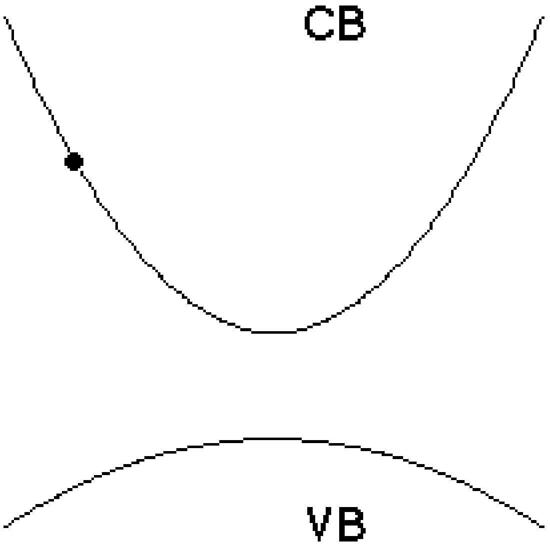


multiphoton excitation

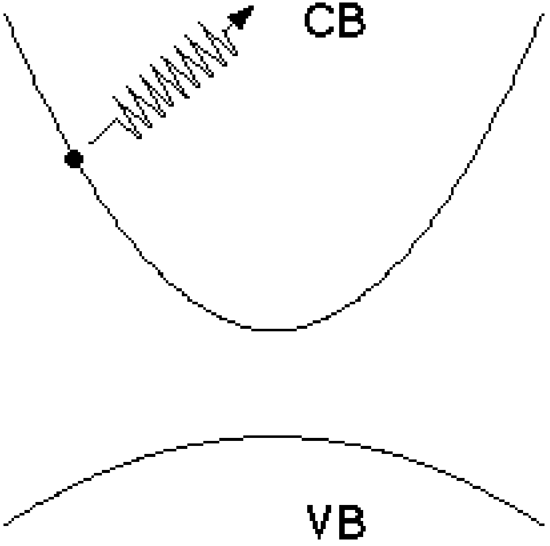
EXCITATION



EXCITATION

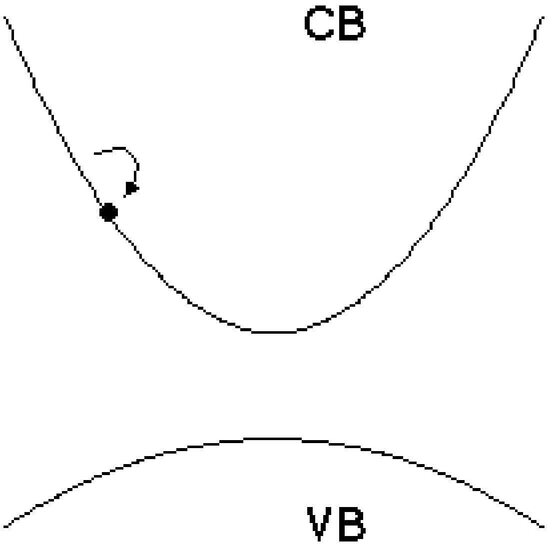


RELAXATION



emission of LO phonons

RELAXATION



emission of LO phonons

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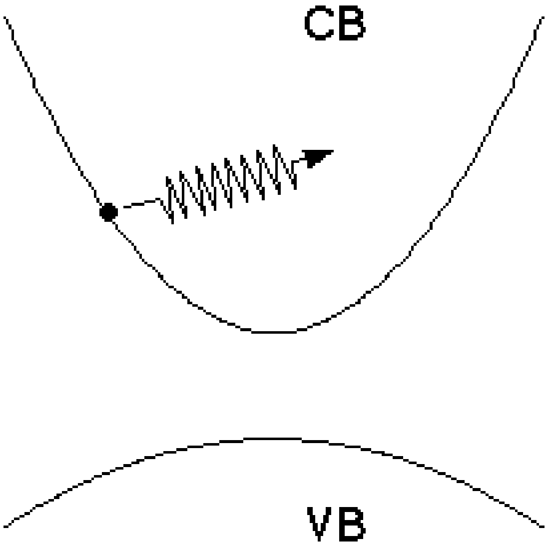
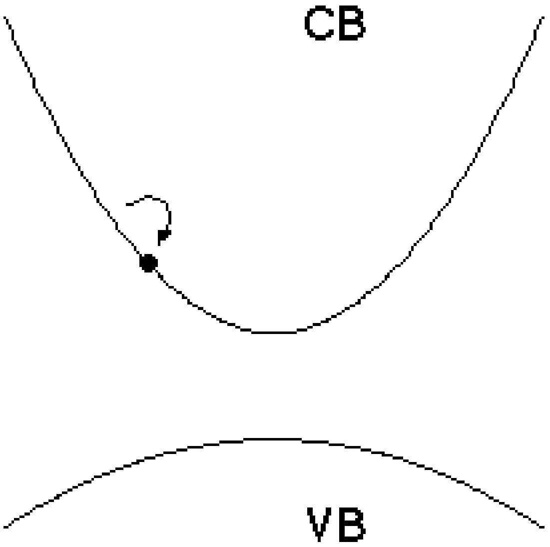


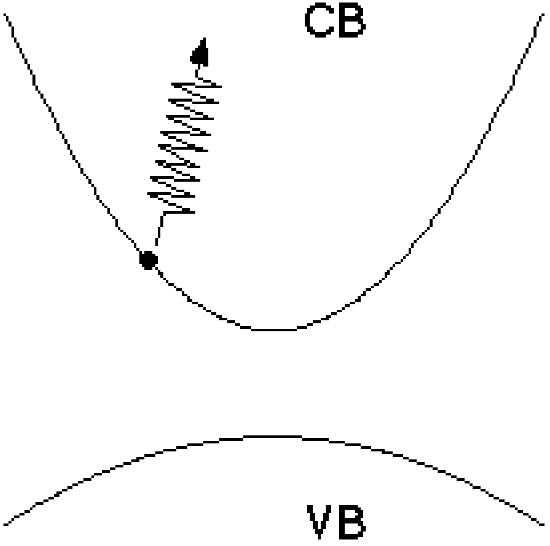
emission of LO phonons

RELAXATION



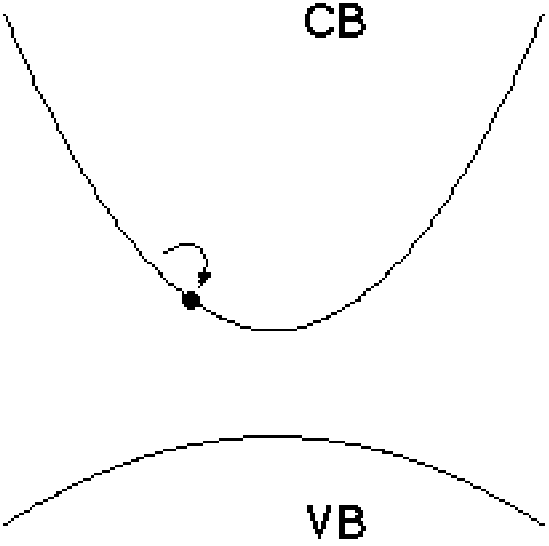
emission of LO phonons

RELAXATION



emission of LO phonons

RELAXATION



emission of LO phonons

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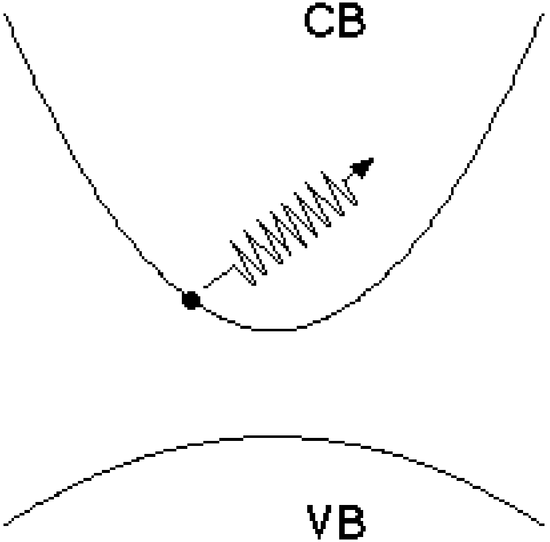
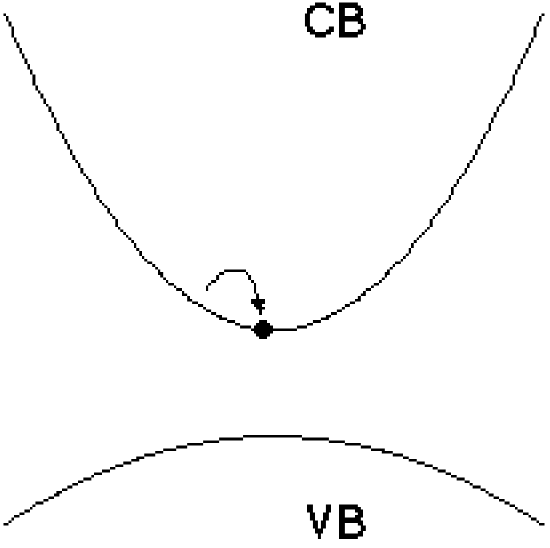


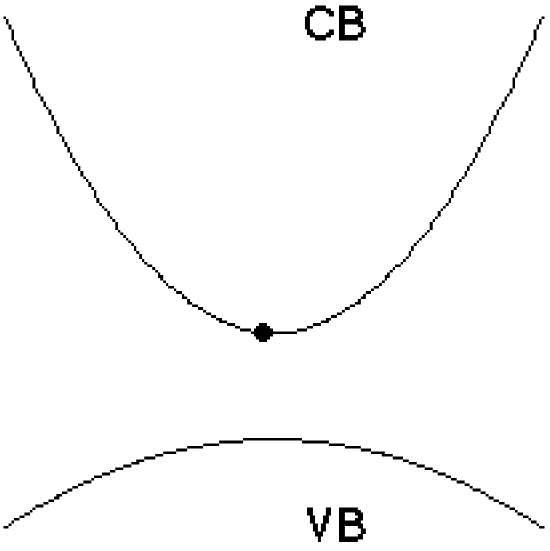
emission of LO phonons

RELAXATION



emission of LO phonons

RELAXATION



TIME SCALE

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

photons

↓

hot free electrons

↓

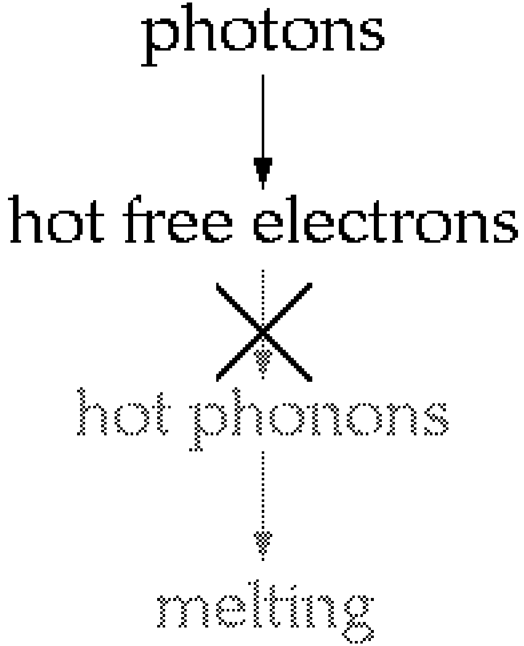
hot phonons

↓

melting

TIME SCALE

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



photons

hot free electrons

hot phonons

melting

OUTLINE

❶ Background

❷ Femtosecond work

❸ Future

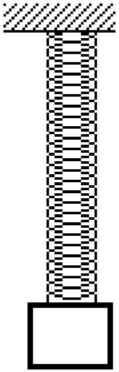
FEMTOSECOND WORK

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

EXCITATION OF PHONONS

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

IMPULSIVE RAMAN SCATTERING



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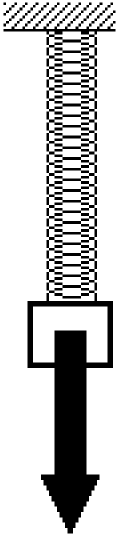

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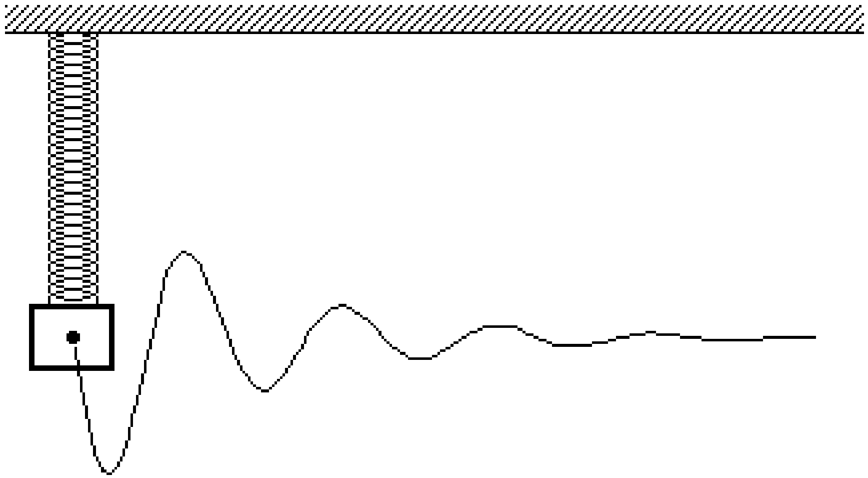

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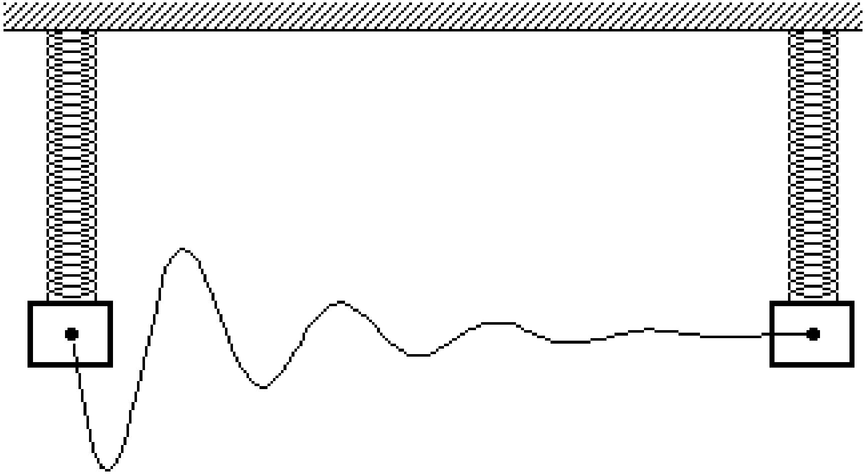

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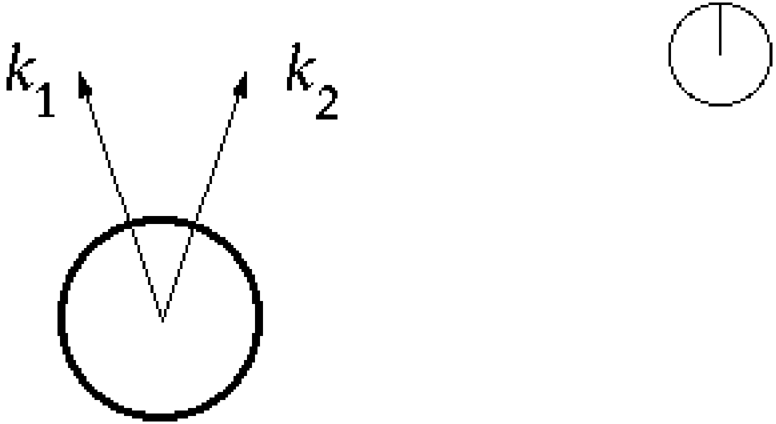

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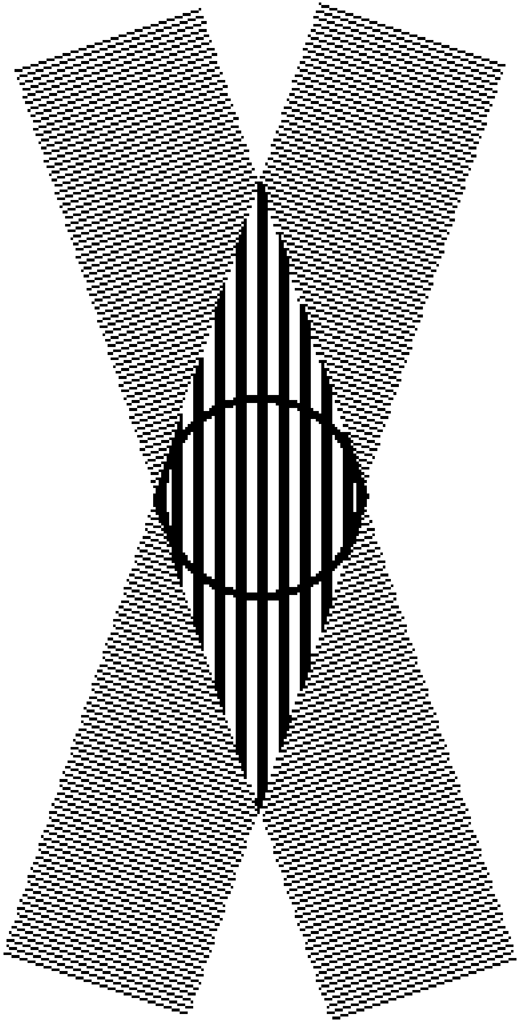
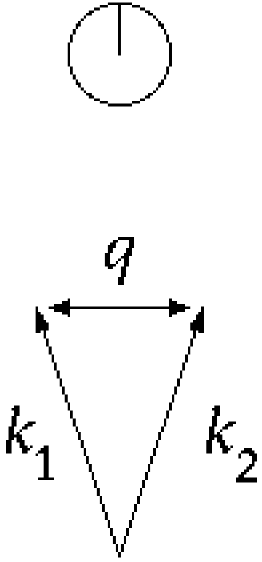
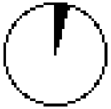
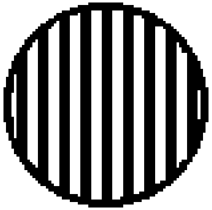




$$k_1 - k_2 = \pm q$$

IMPULSIVE RAMAN SCATTERING



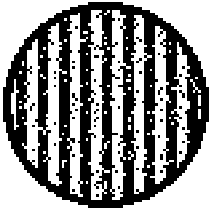


create decaying standing wave...

IMPULSIVE RAMAN SCATTERING

create decaying standing wave...

IMPULSIVE RAMAN SCATTERING



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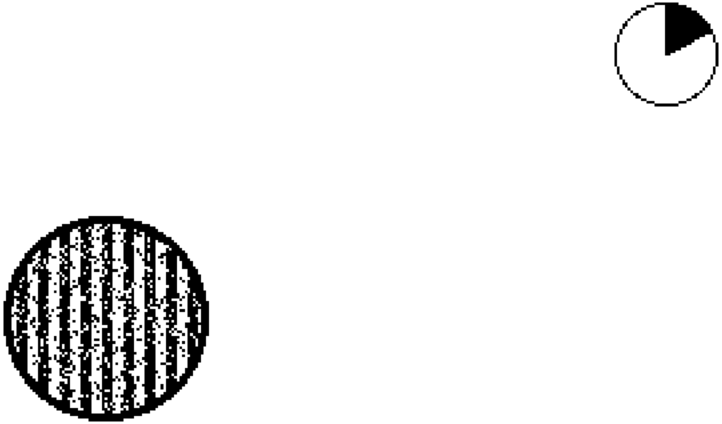

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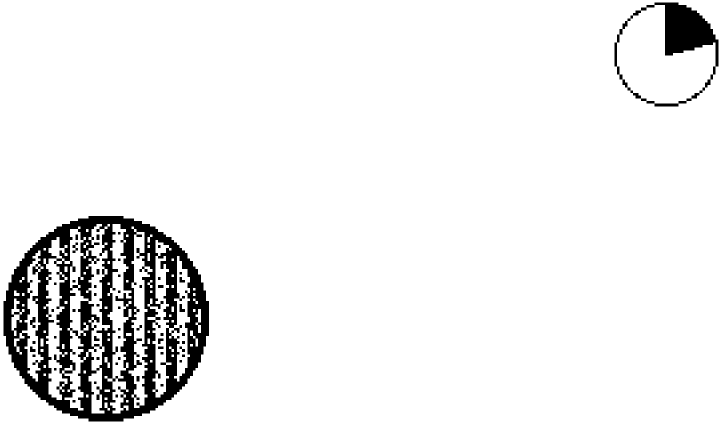

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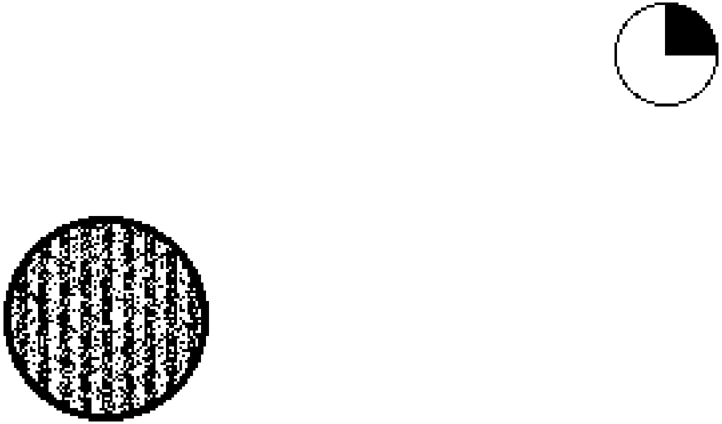

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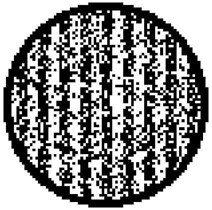

create decaying standing wave...

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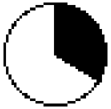

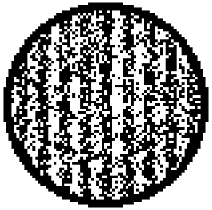

create decaying standing wave...

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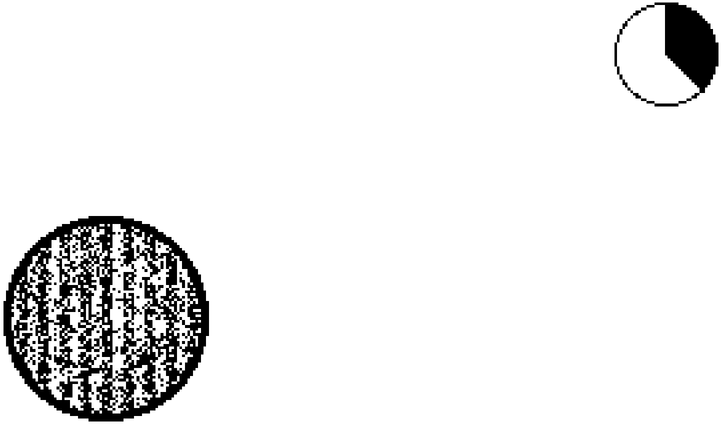

create decaying standing wave...

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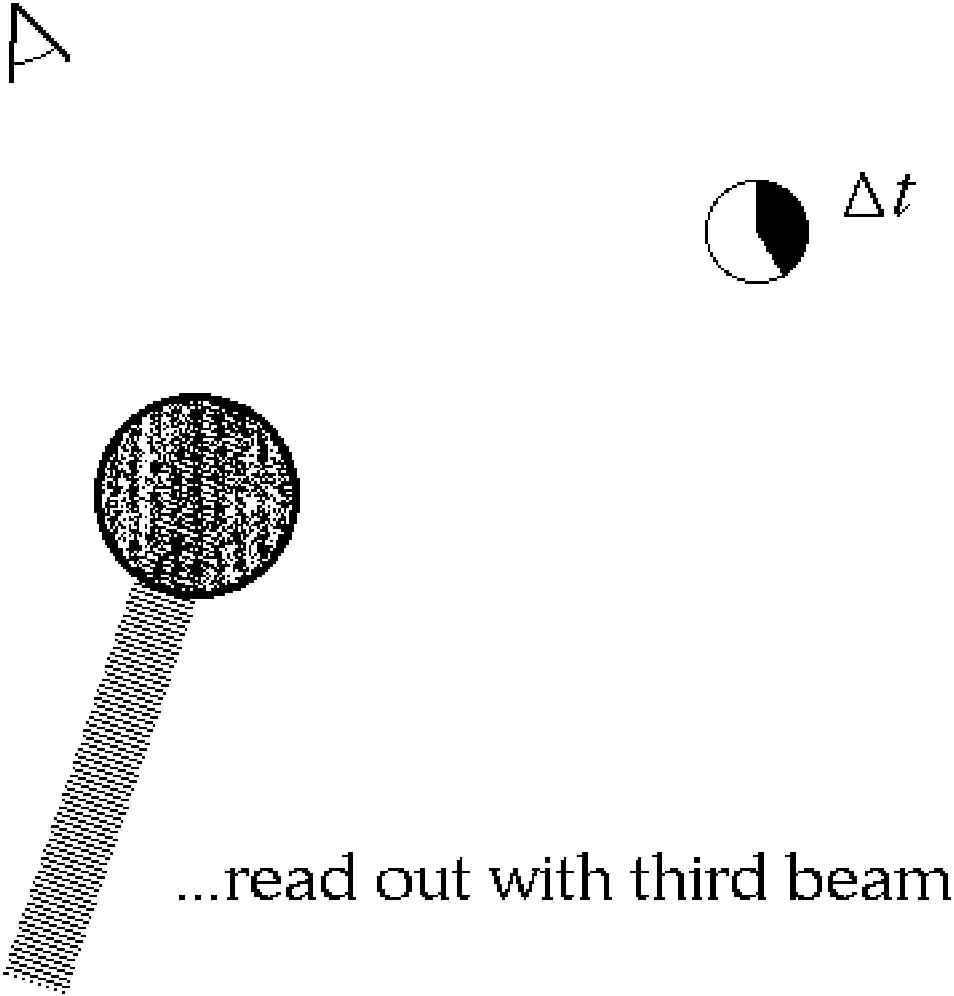
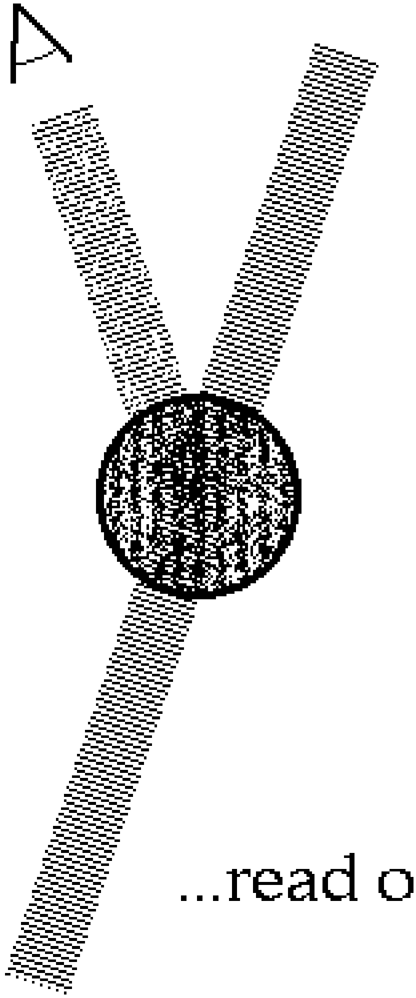
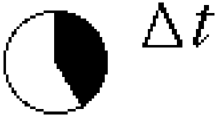


...read out with third beam

IMPULSIVE RAMAN SCATTERING





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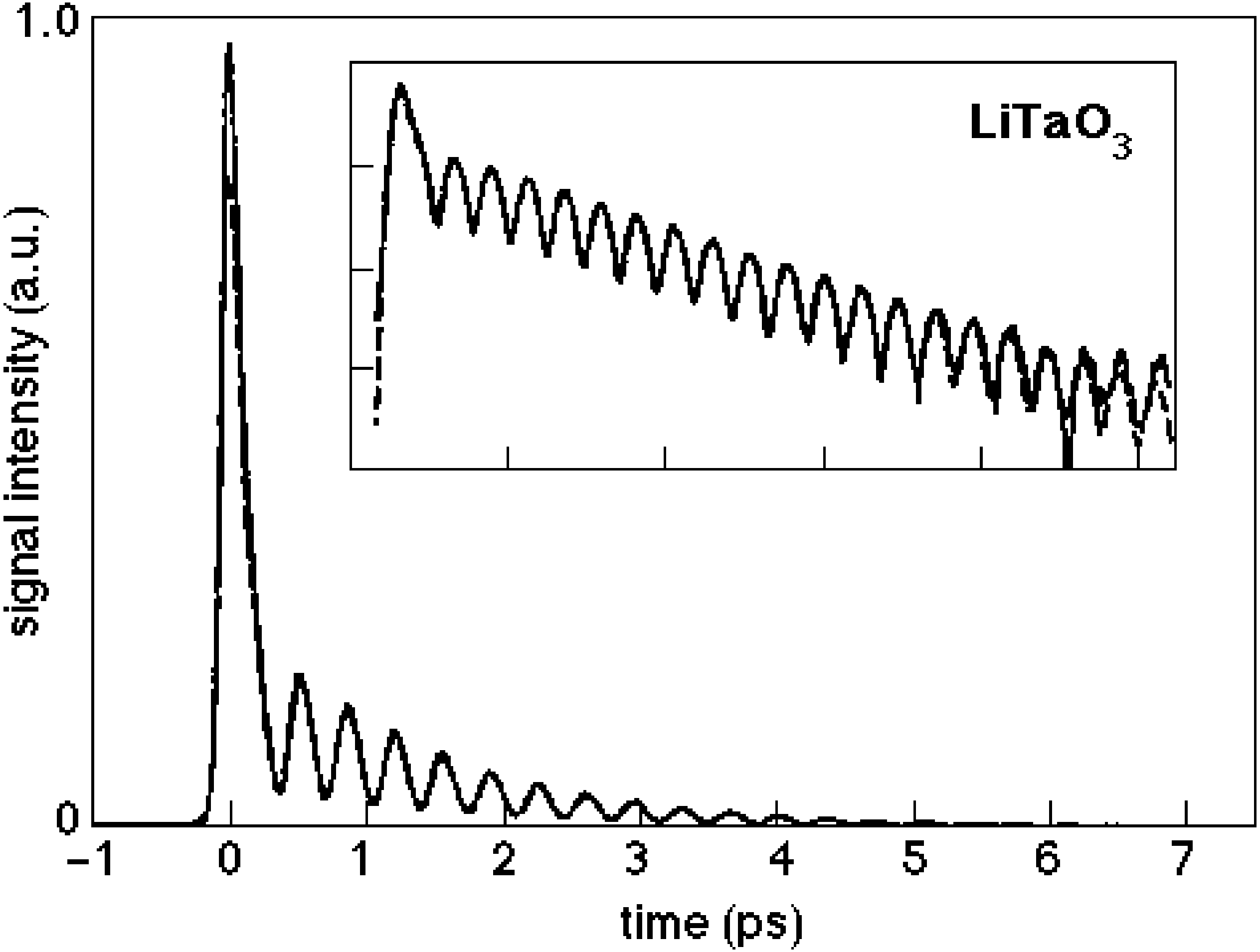
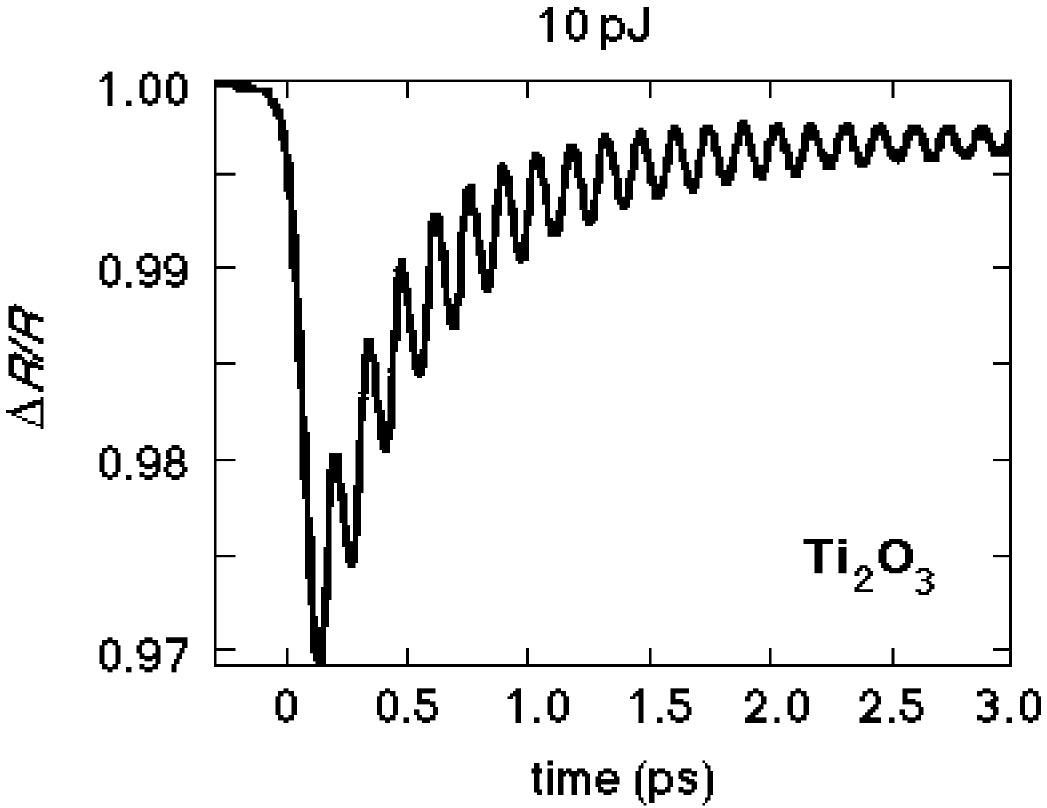


L. Dhar *et al., Chem. Rev.* **94** (1994) 157

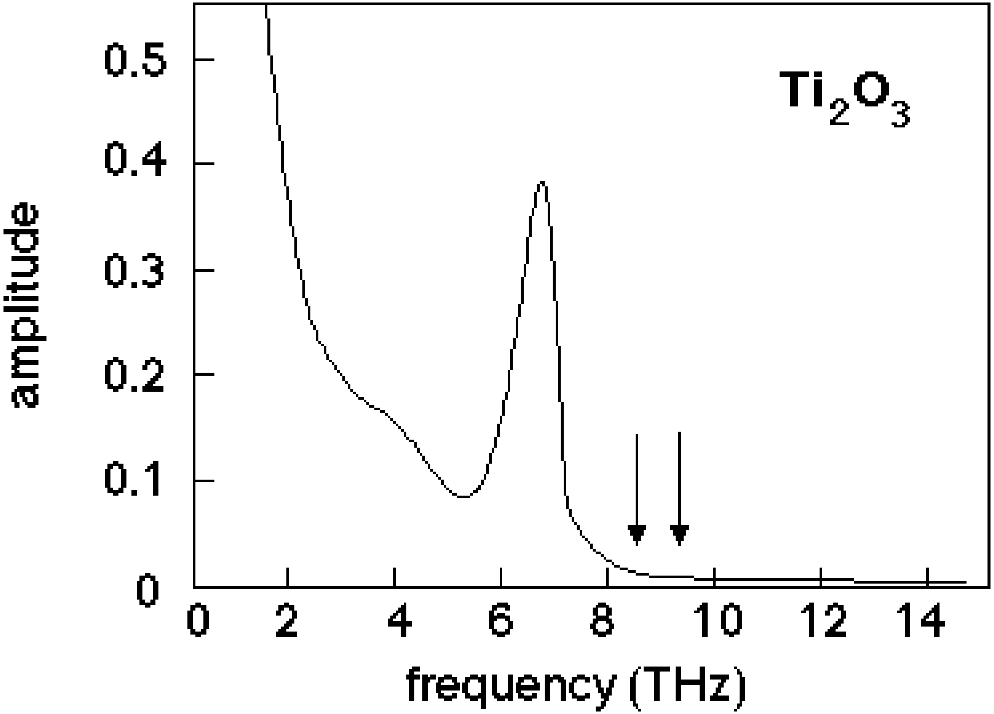
LARGE AMPLITUDE OSCILLATIONS



T.K. Cheng *et al., Appl. Phys. Lett.* **57** (1990) 1004

H.J. Zeiger *et al., Phys. Rev. B* **45** (1992) 768

LARGE AMPLITUDE OSCILLATIONS



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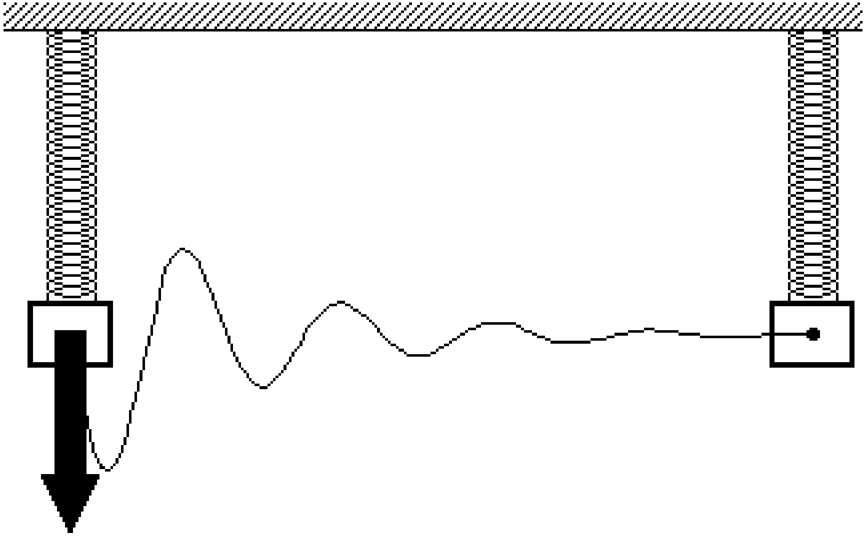
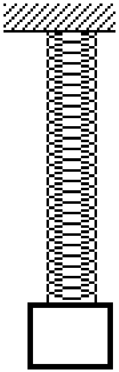
IMPULSIVE RAMAN SCATTERING



- need to match *q*
- sinusoidal oscillation

DISPLACIVE EXCITATION



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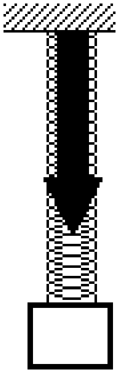

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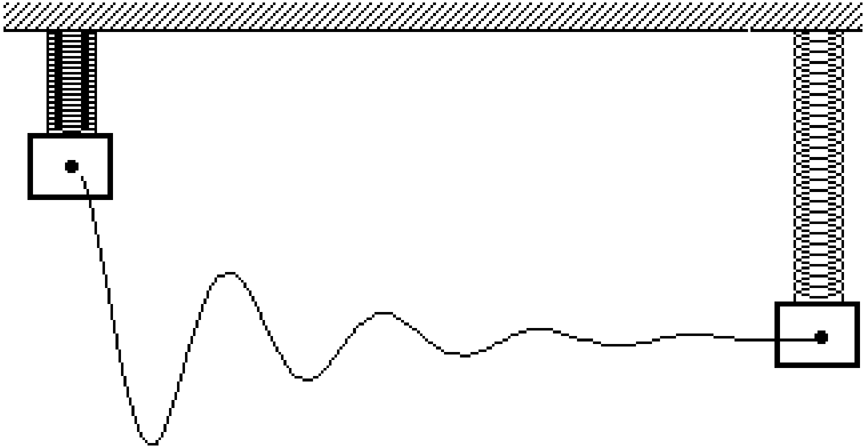

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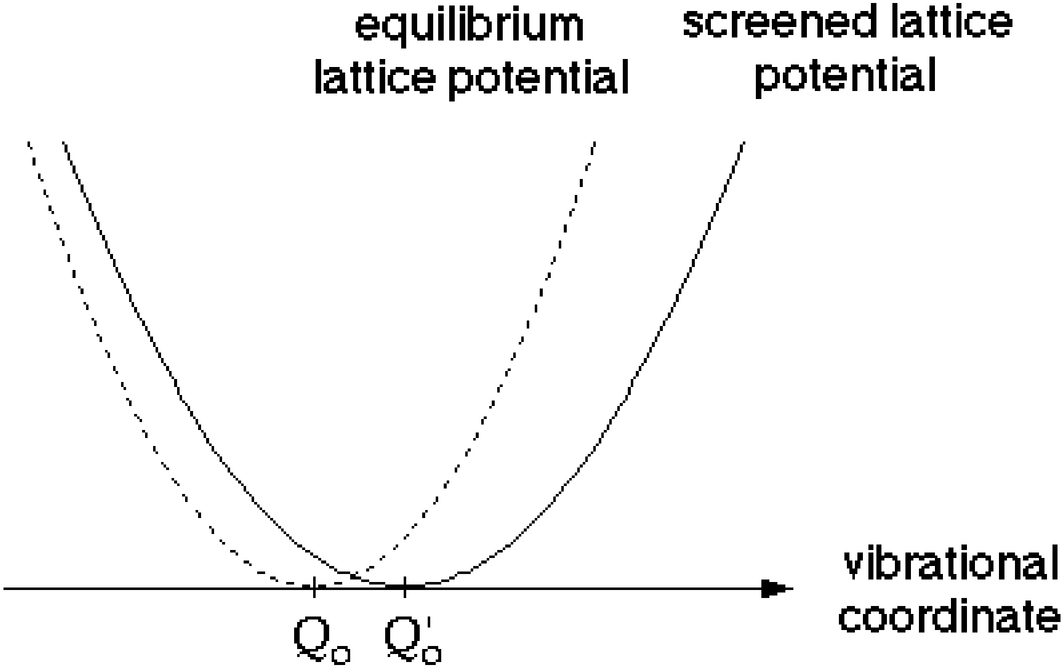

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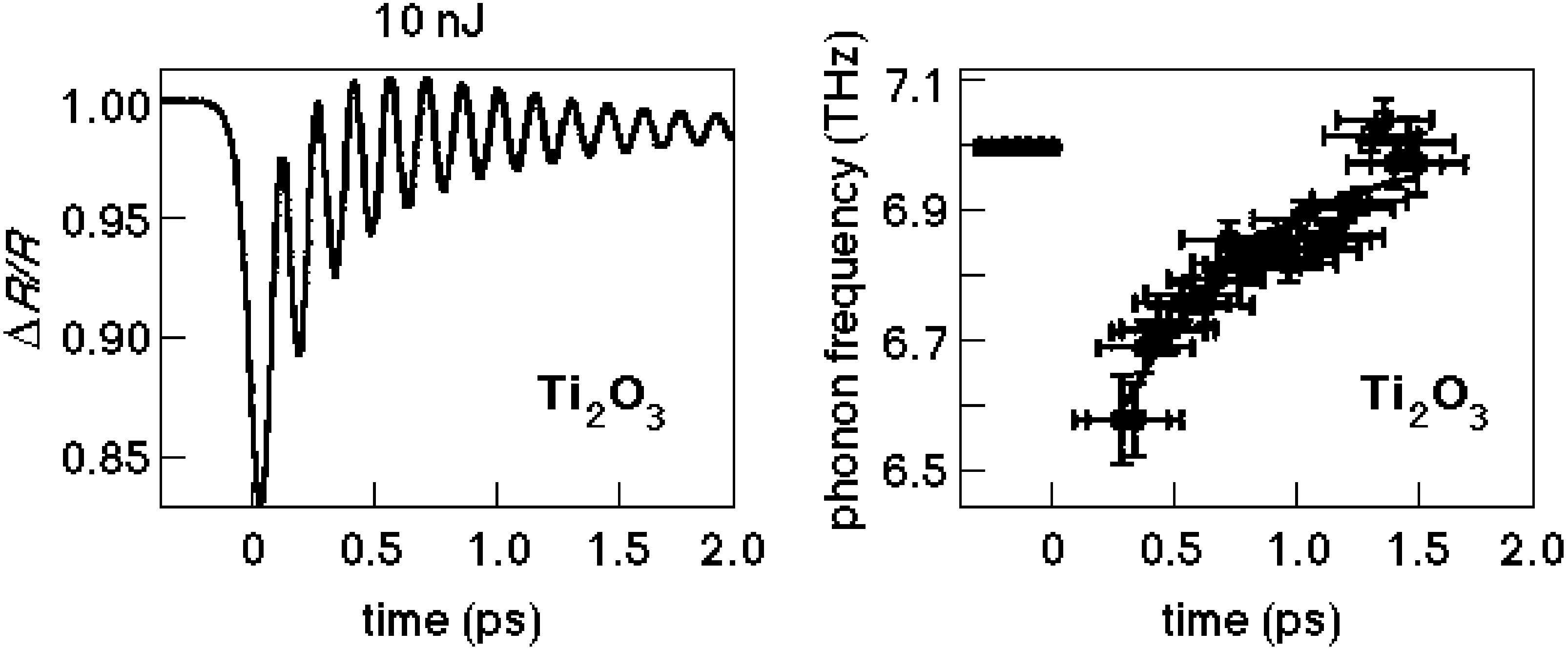
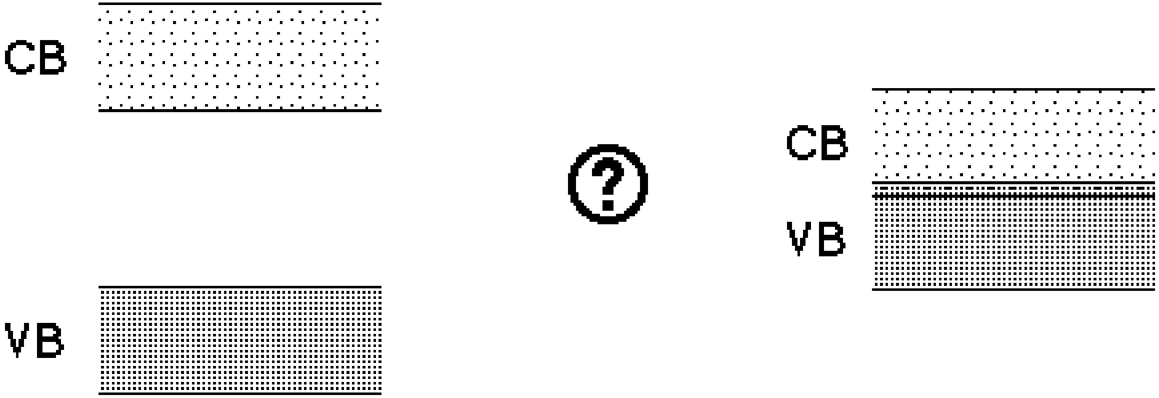


T.K. Cheng *et al.*, *Appl. Phys. Lett.* **62** (1993) 1901

BEYOND PHONONS

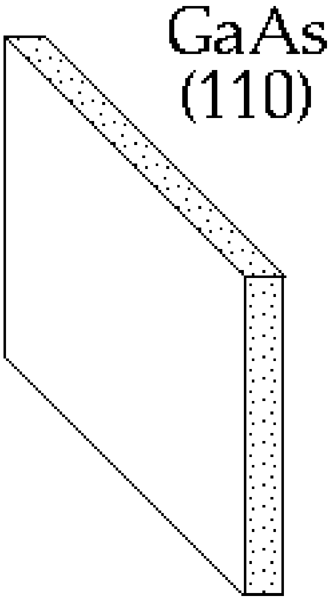
intense pulses can ‘melt’ GaAs, Si, but how?



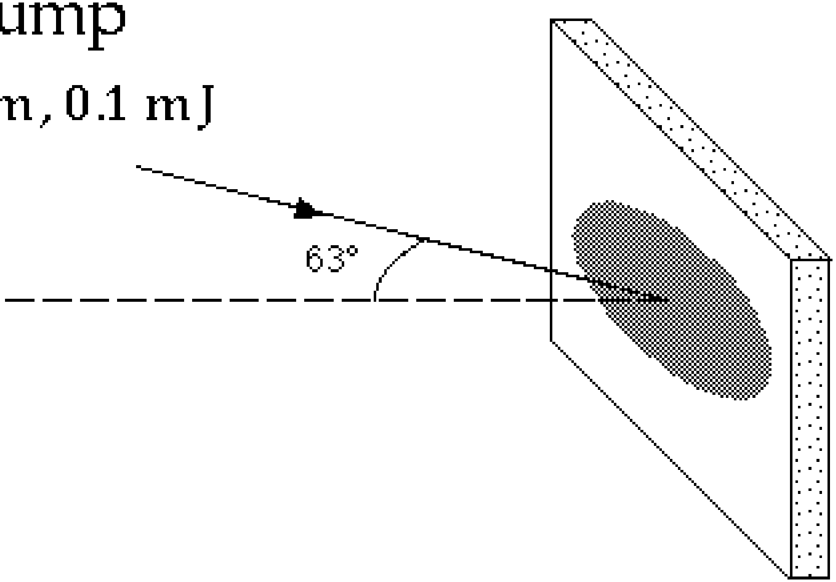
BEYOND PHONONS

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

2.2 eV DIELECTRIC CONSTANT



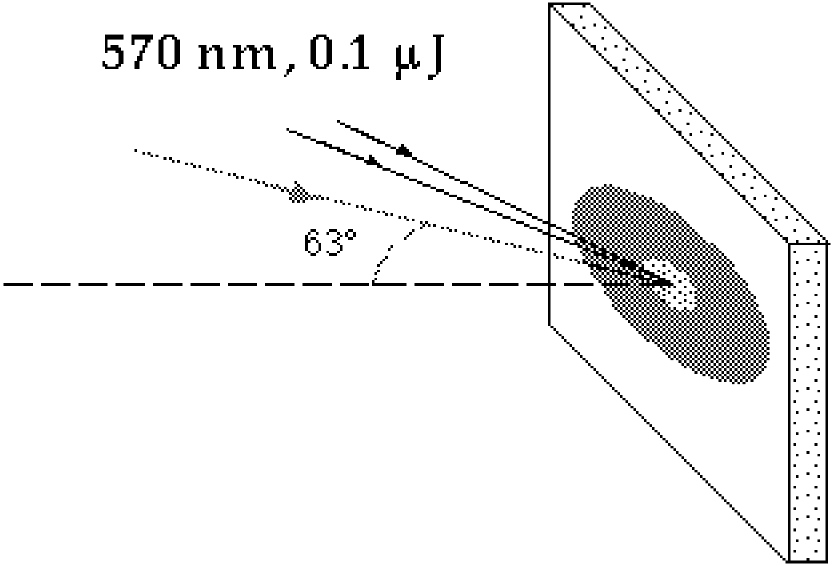
2.2 eV DIELECTRIC CONSTANT



pump

640 nm, 0.1 mJ

2.2 eV DIELECTRIC CONSTANT



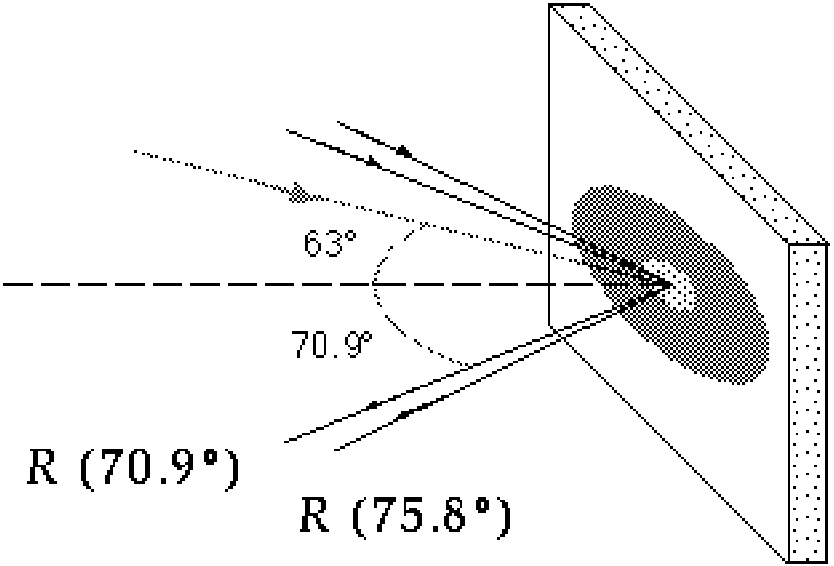
probes

570 nm, 0.1 μJ

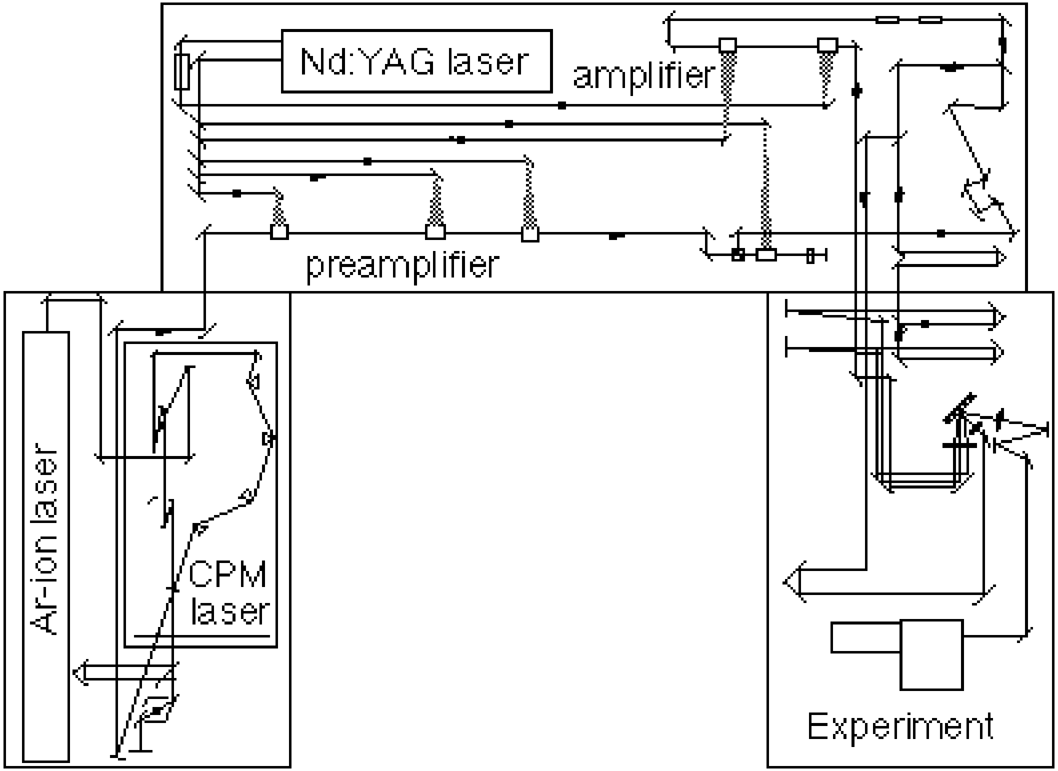
all beams

70 fs, *p*-pol

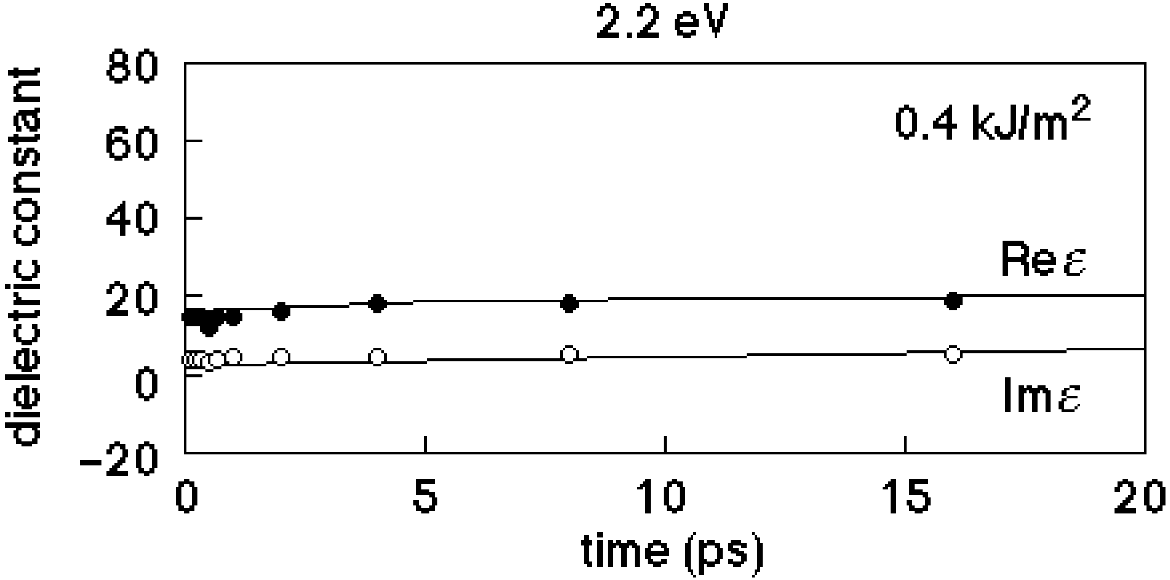
2.2 eV DIELECTRIC CONSTANT



SETUP


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

TIME DEPENDENCE



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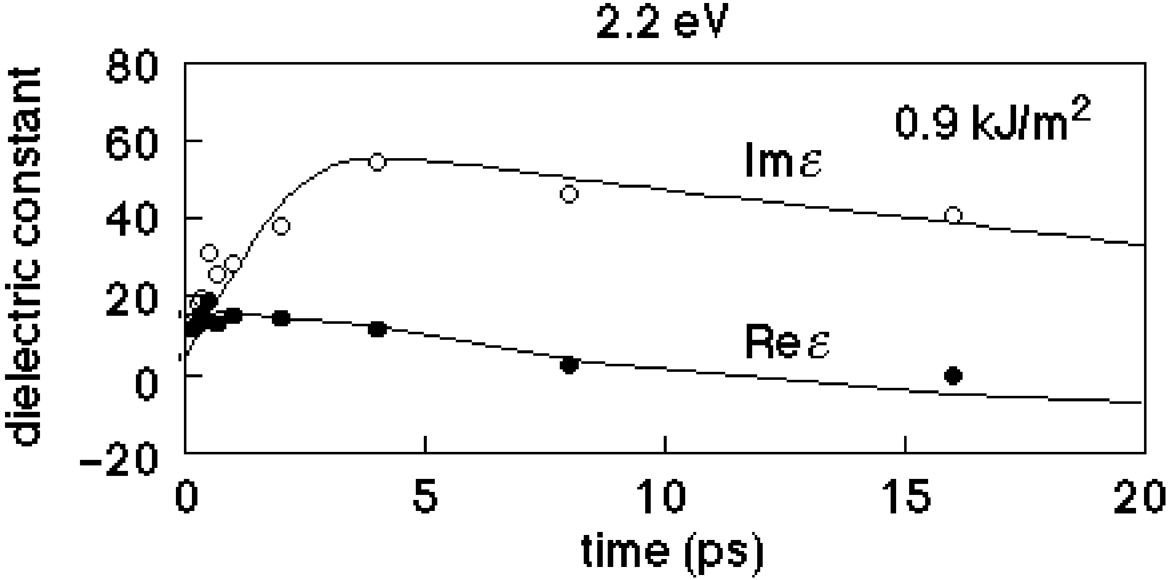
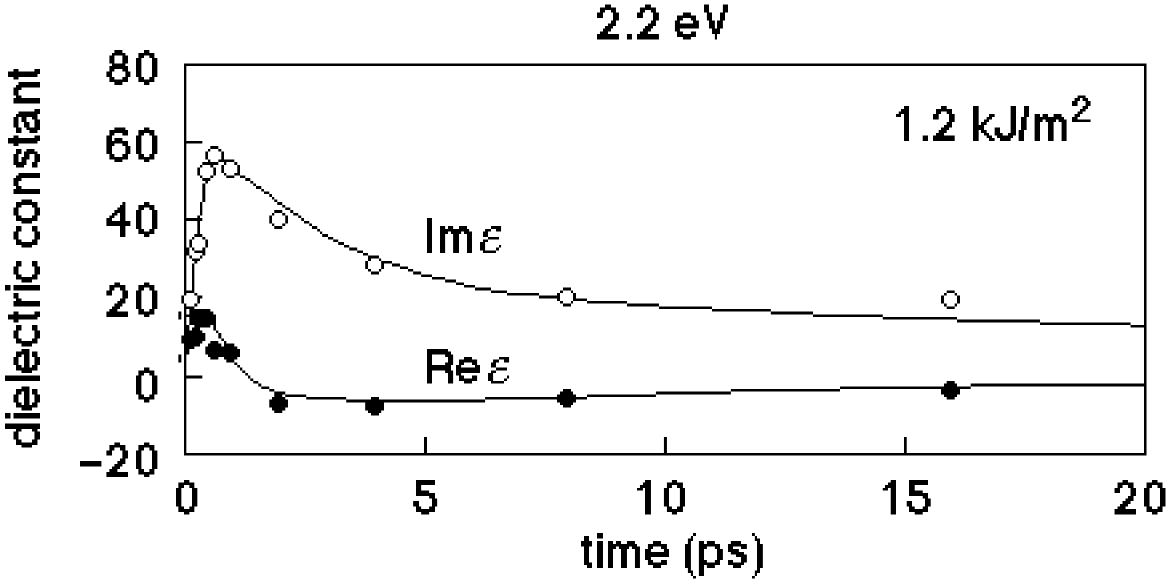
TIME DEPENDENCE



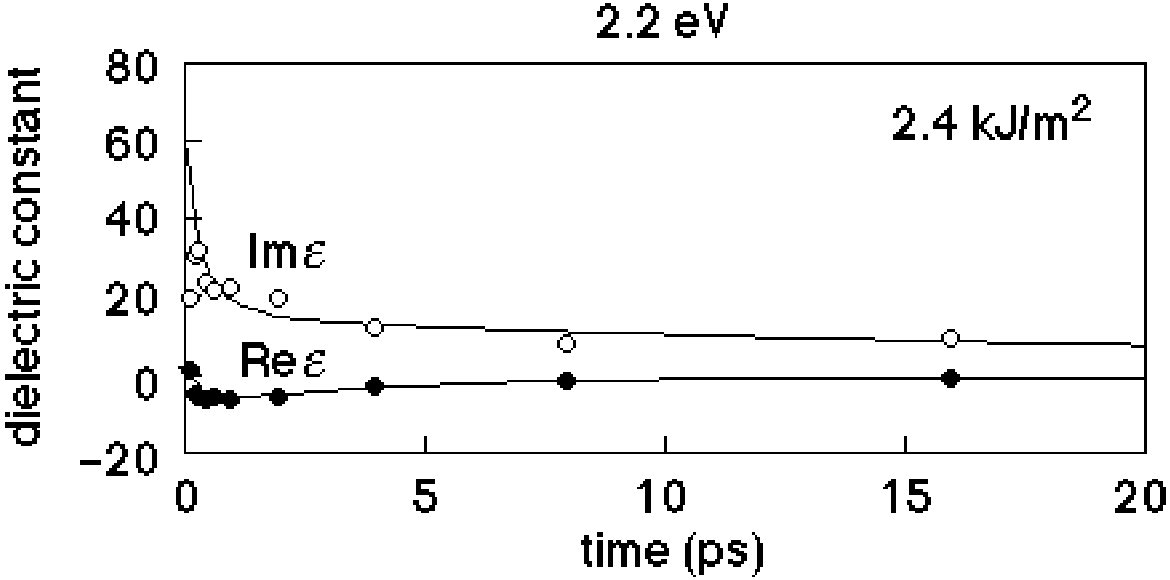
Phys. Rev. B **51** (1995) 6959

TIME DEPENDENCE



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

TIME DEPENDENCE



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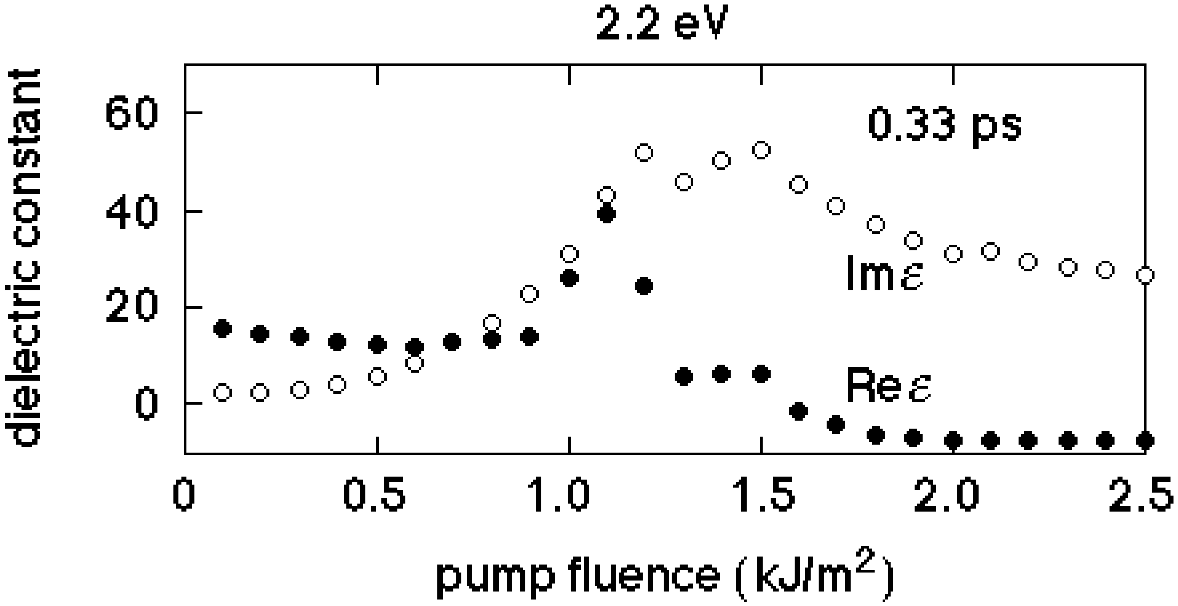
FLUENCE DEPENDENCE



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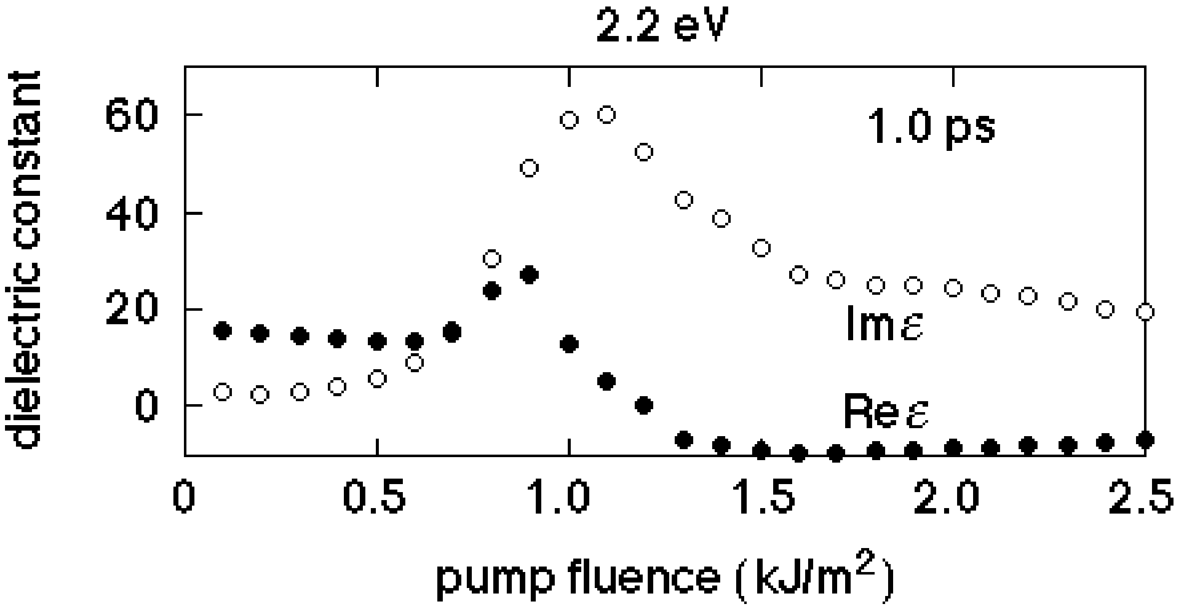
FLUENCE DEPENDENCE



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

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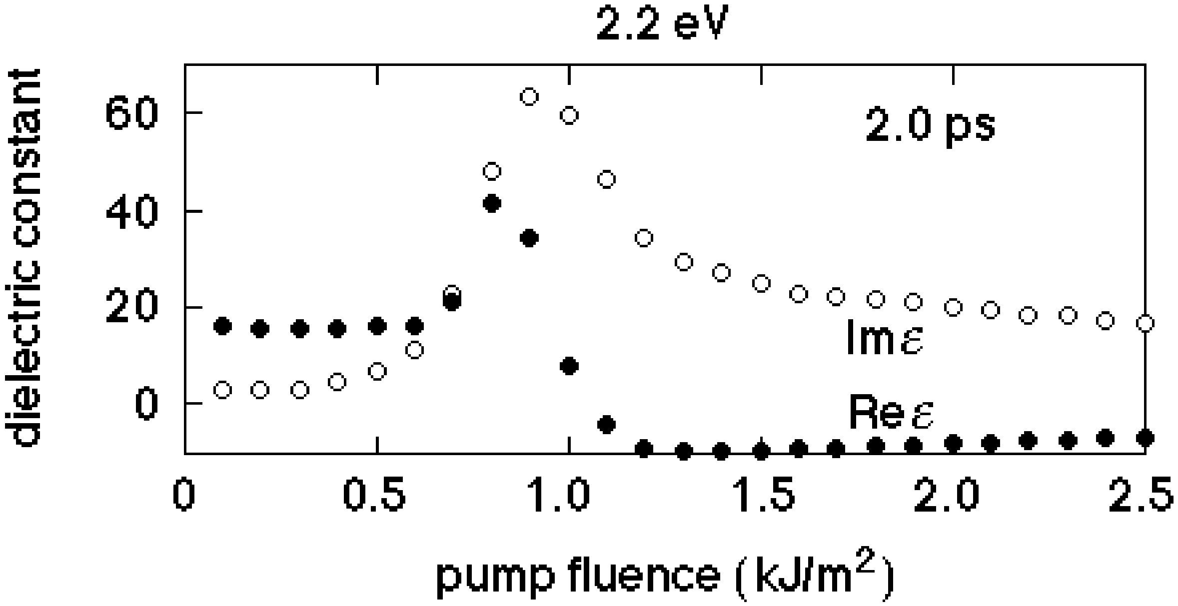

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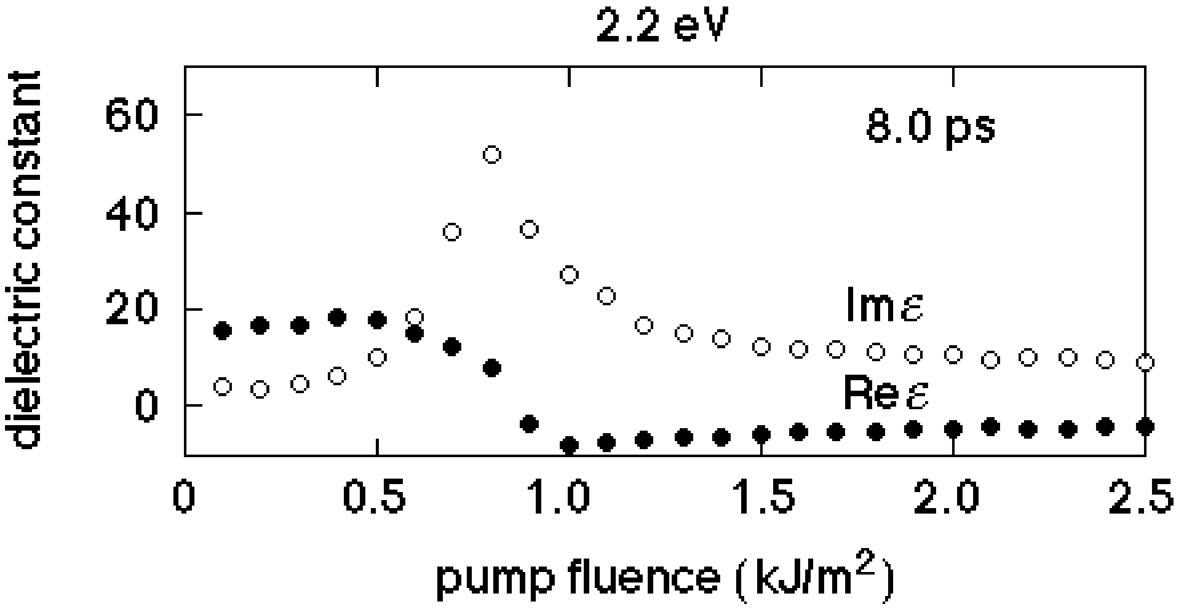
FLUENCE DEPENDENCE



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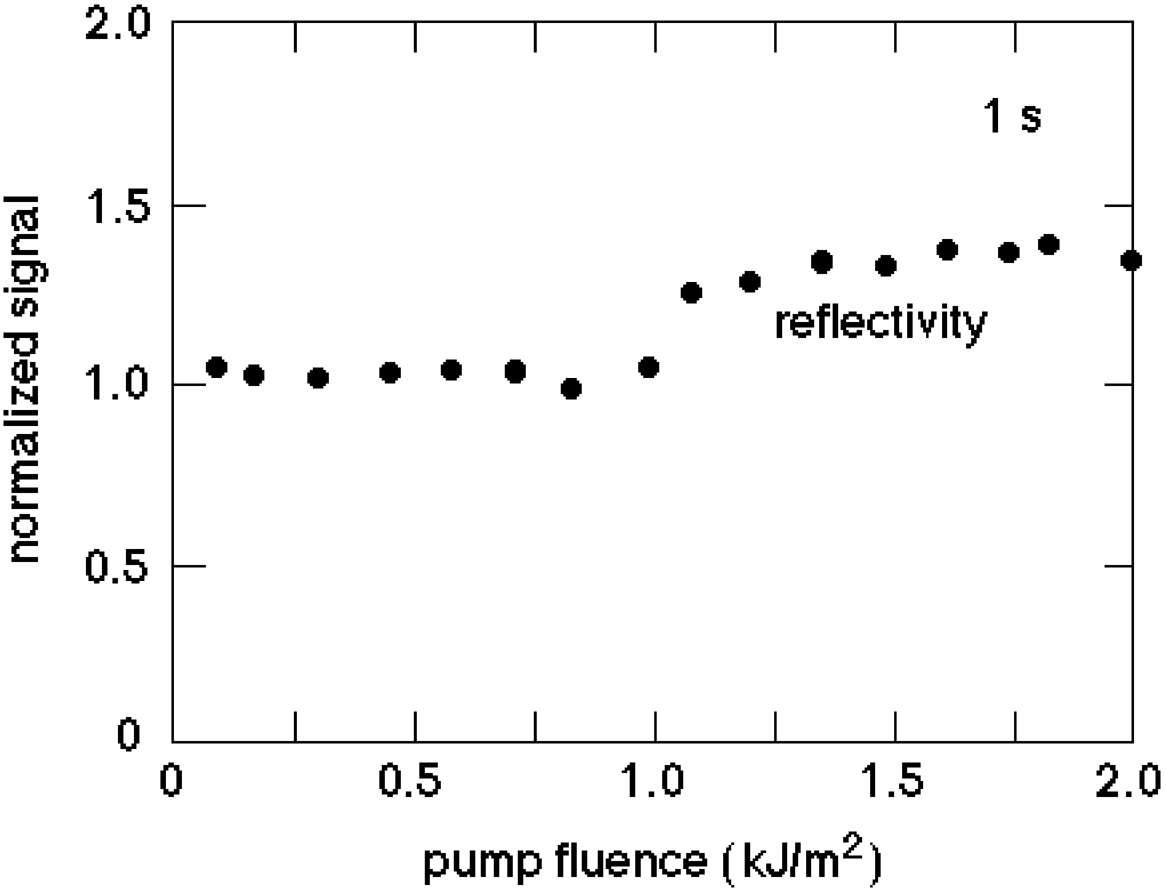
REVERSIBLE vs. IRREVERSIBLE



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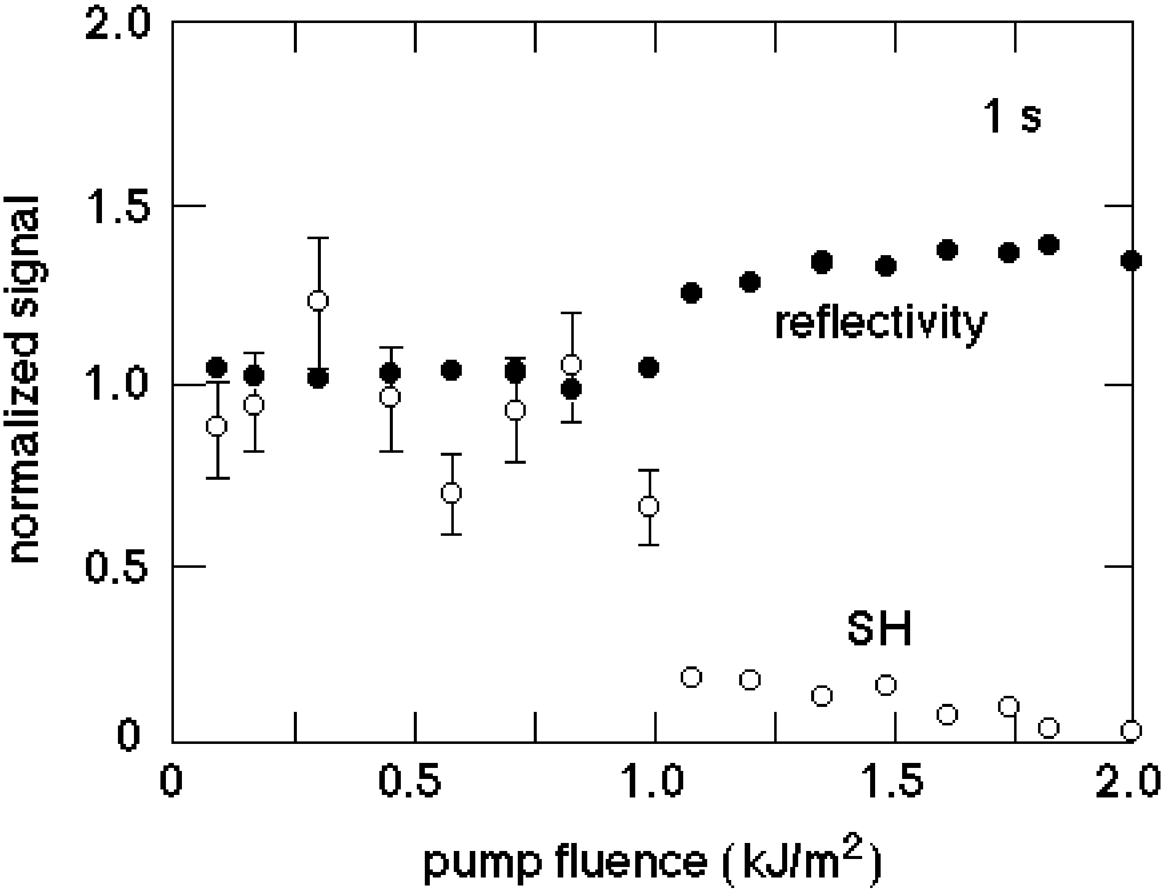
REVERSIBLE vs. IRREVERSIBLE



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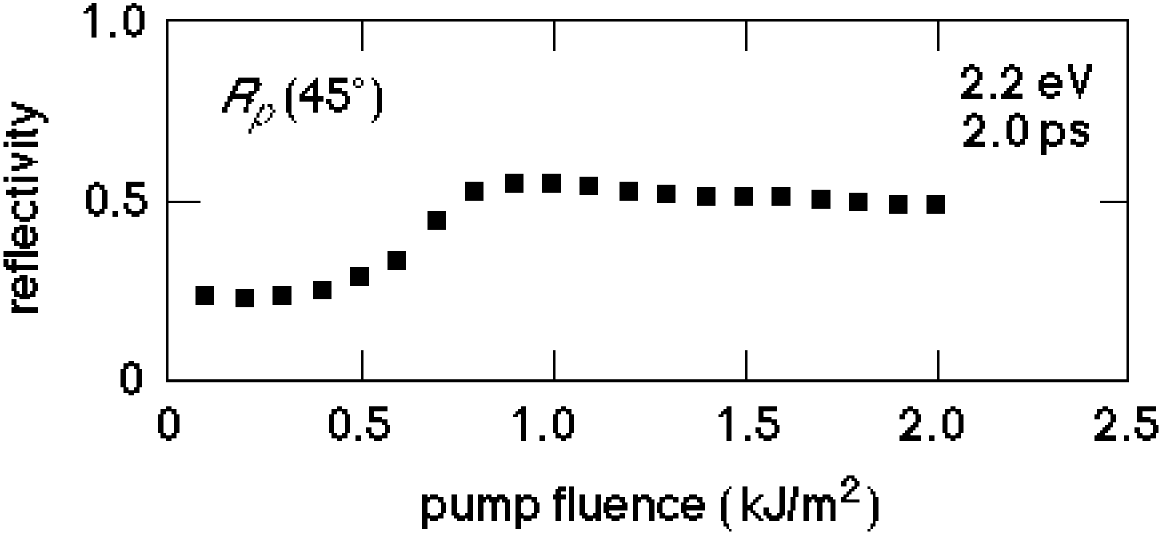
SIMPLE MODEL



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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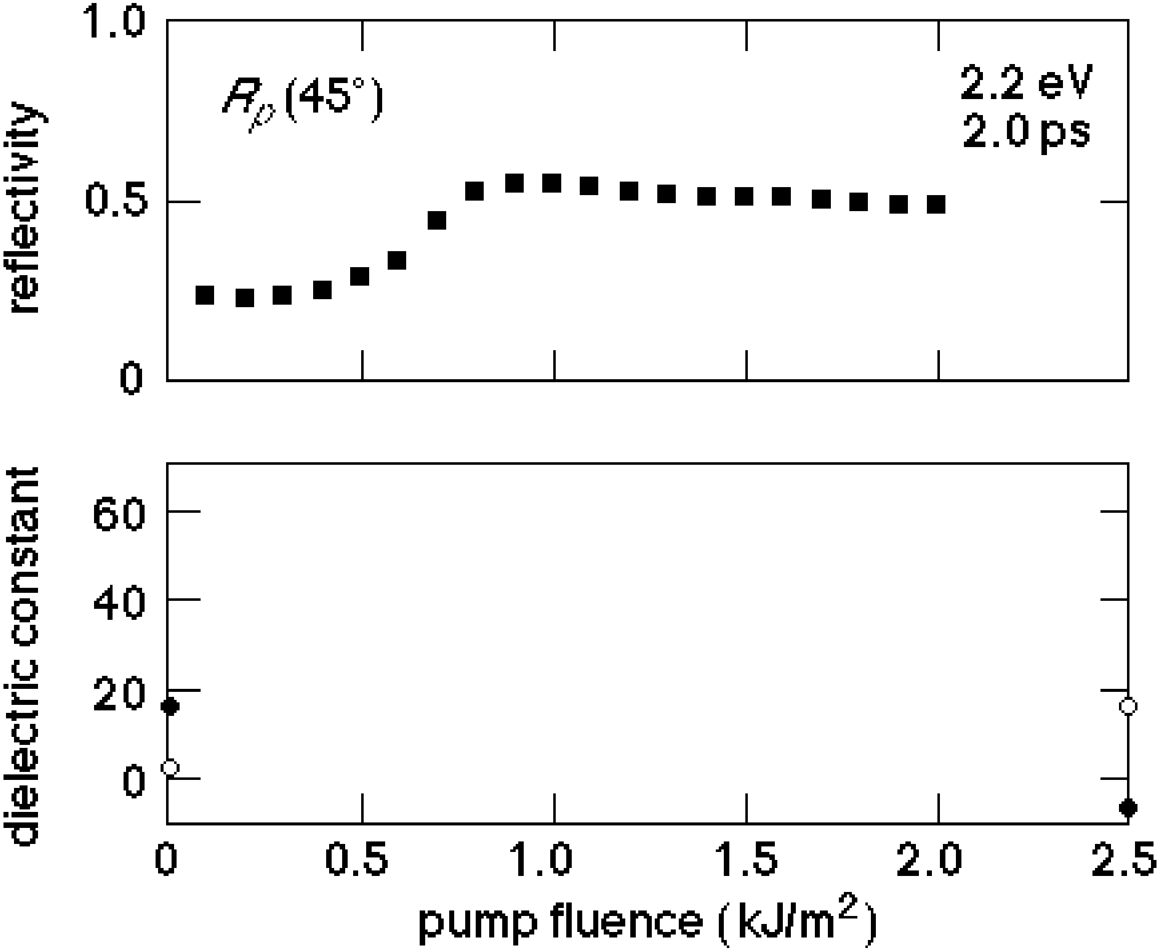
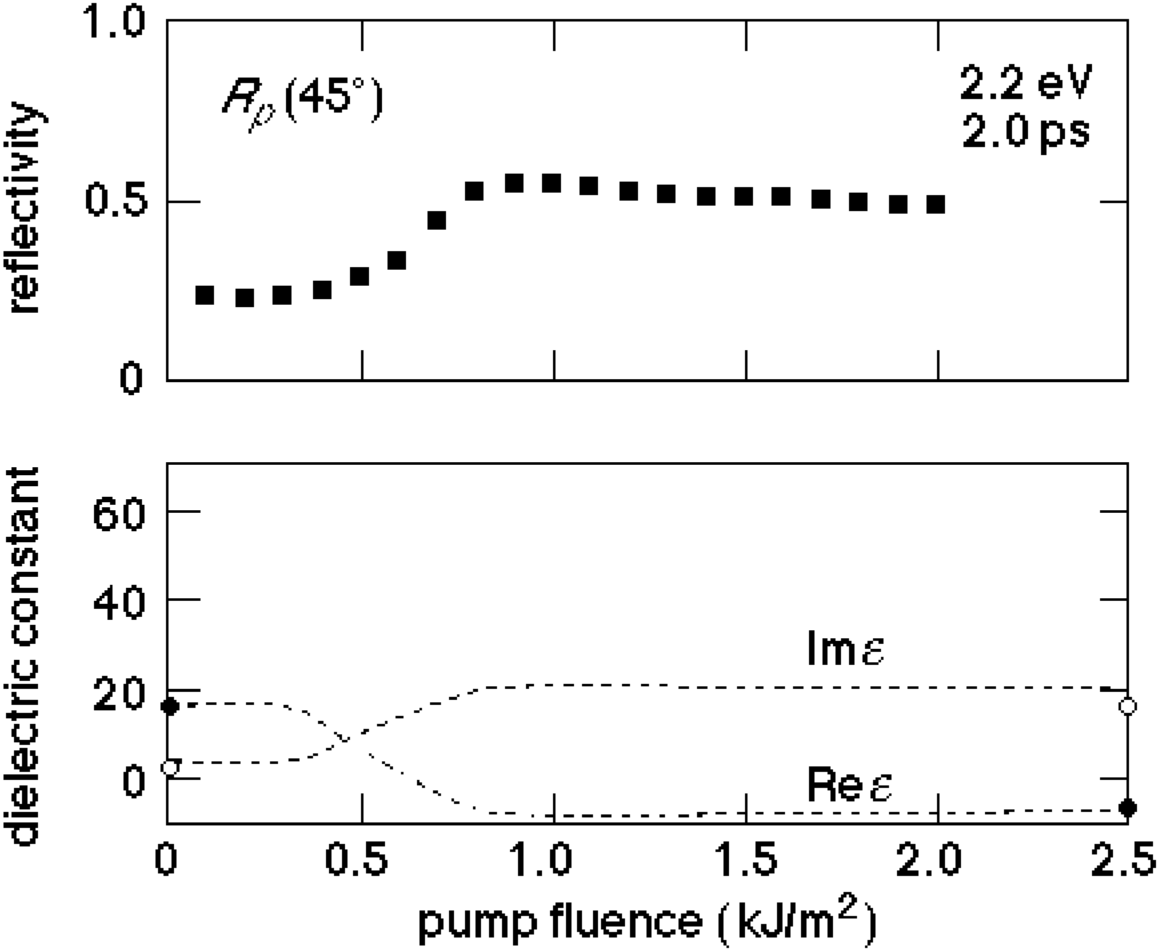
SIMPLE MODEL



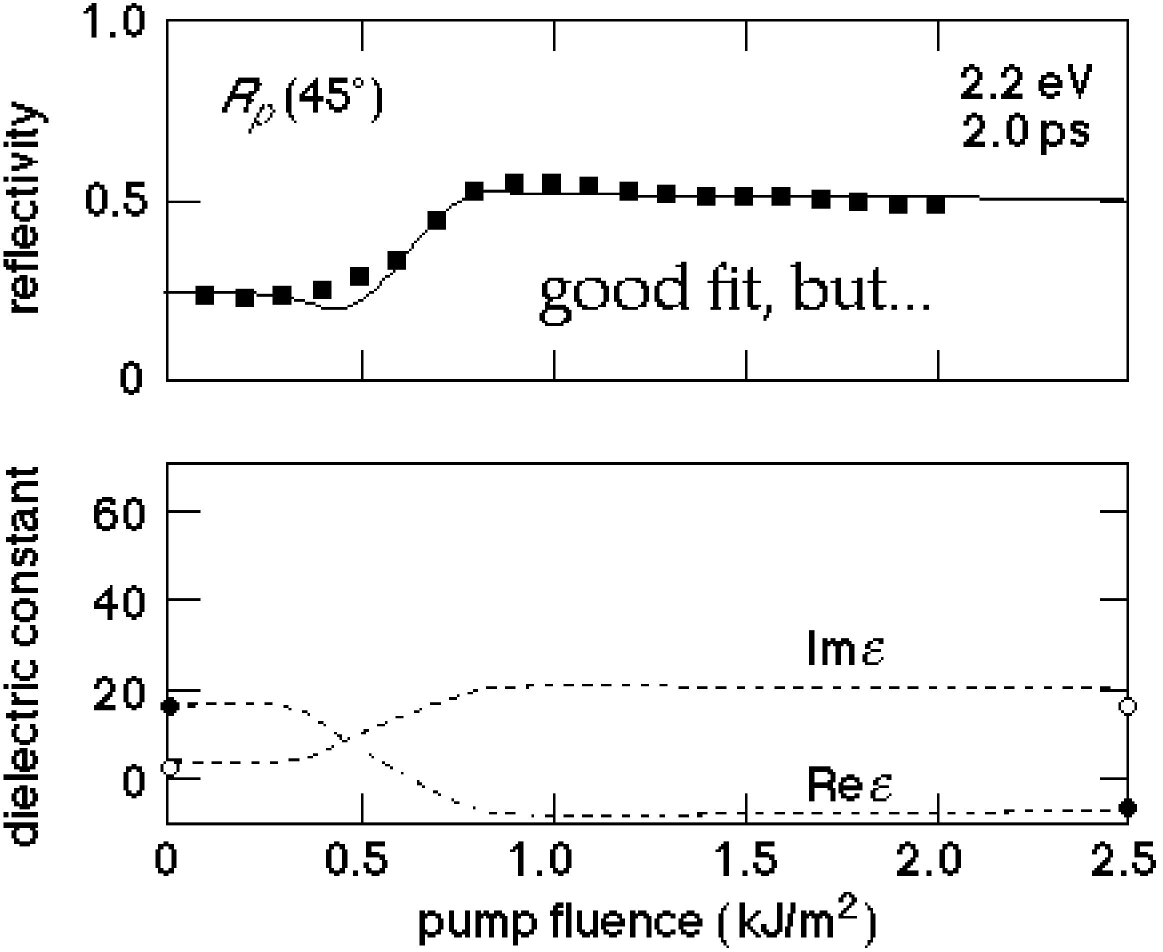
Annu. Rev. Mater. Sci. **25** (1995) 223

SIMPLE MODEL



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

SIMPLE MODEL



good fit, but...

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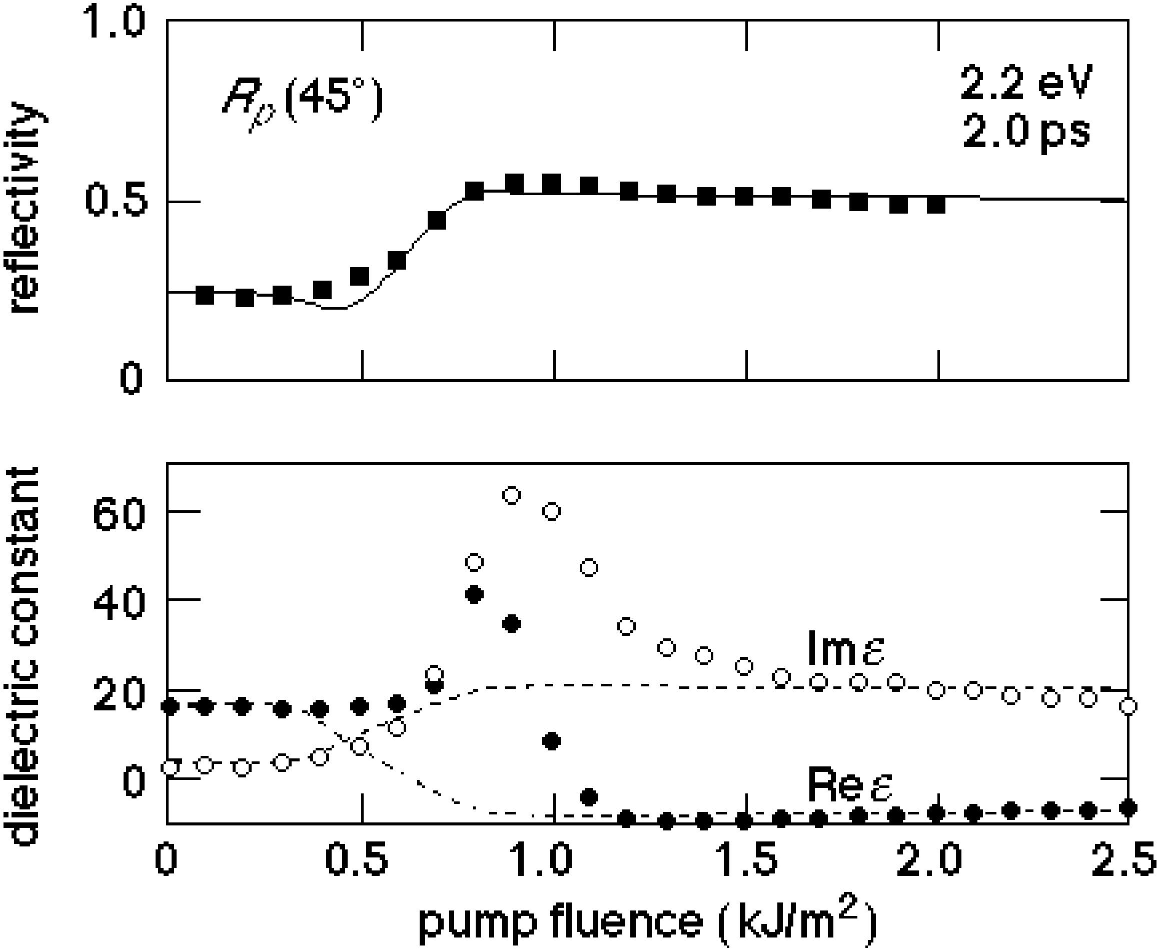
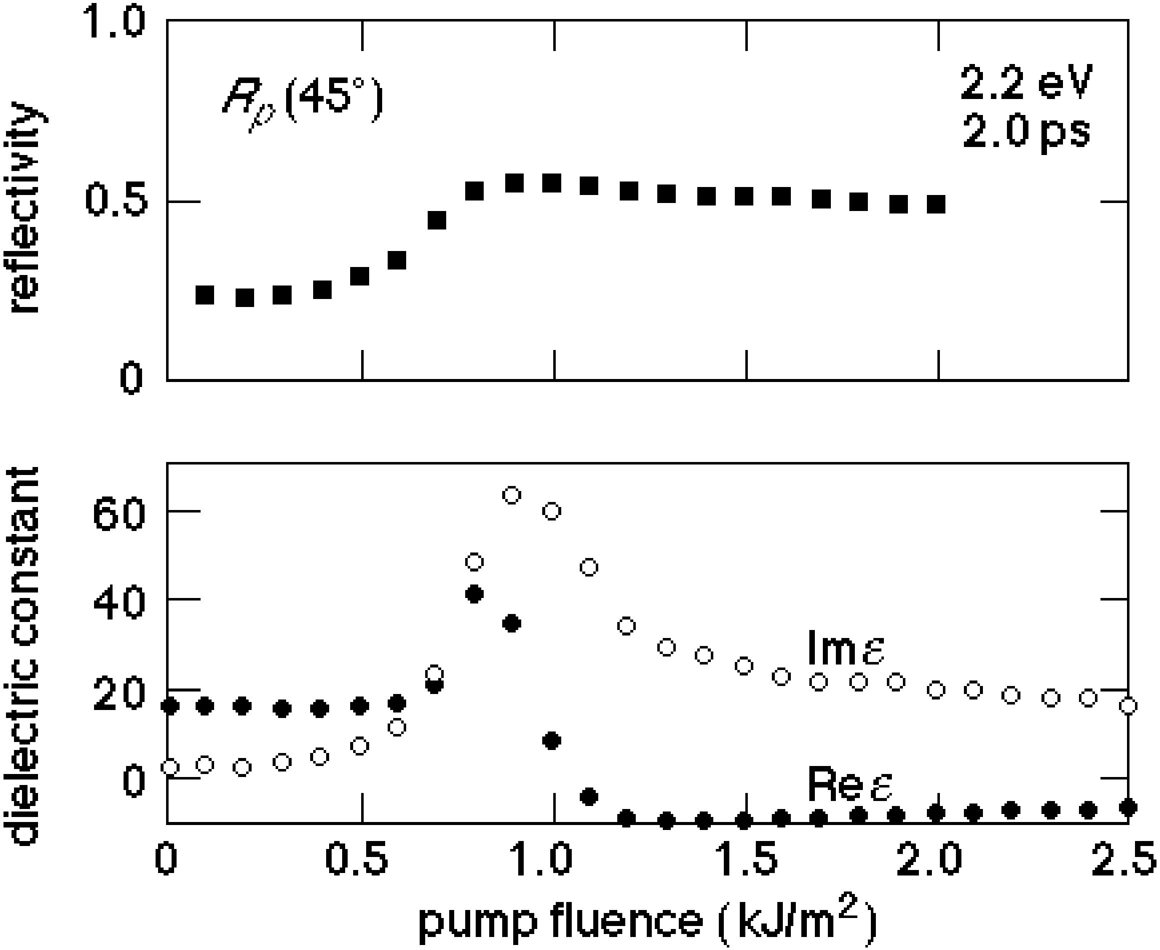
SIMPLE MODEL



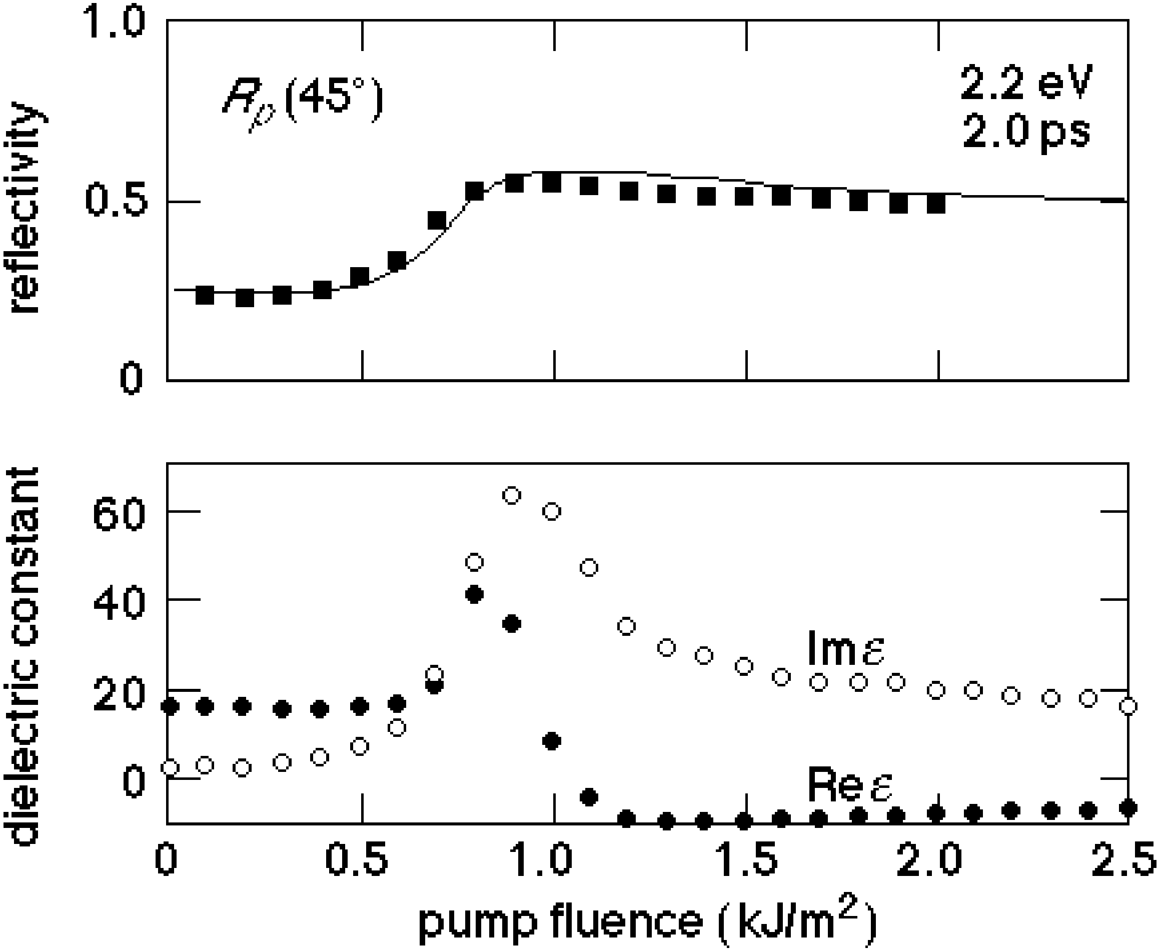
Annu. Rev. Mater. Sci. **25** (1995) 223

SIMPLE MODEL



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

SIMPLE MODEL



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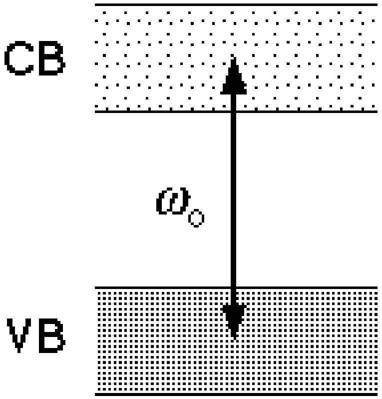
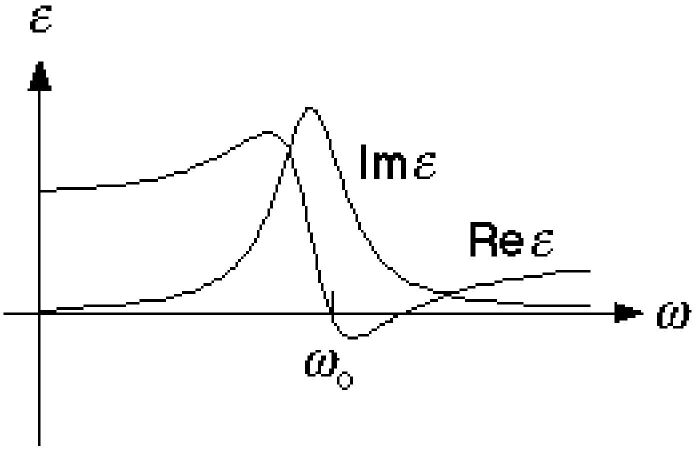
WHAT IS HAPPENING?

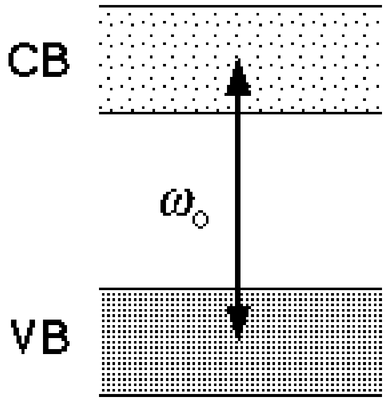


semiconductor

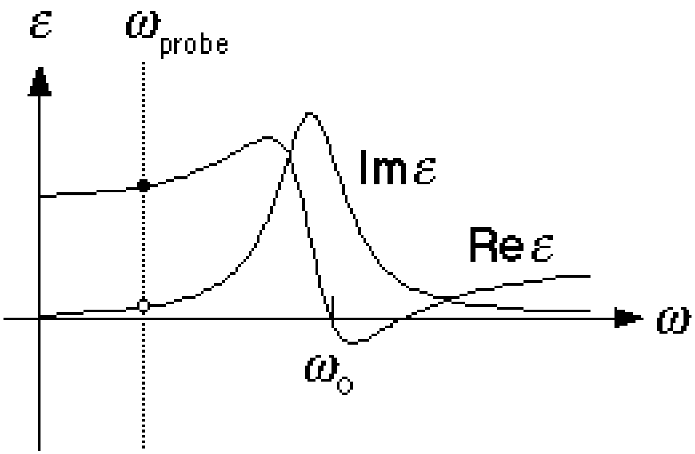


$\omega_0 \approx 4.5$ eV

WHAT IS HAPPENING?

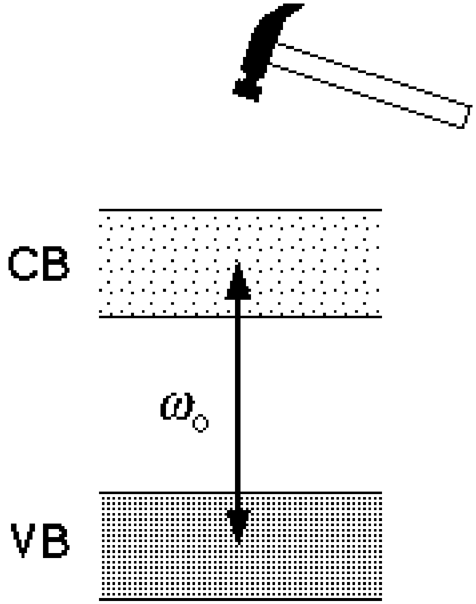


semiconductor

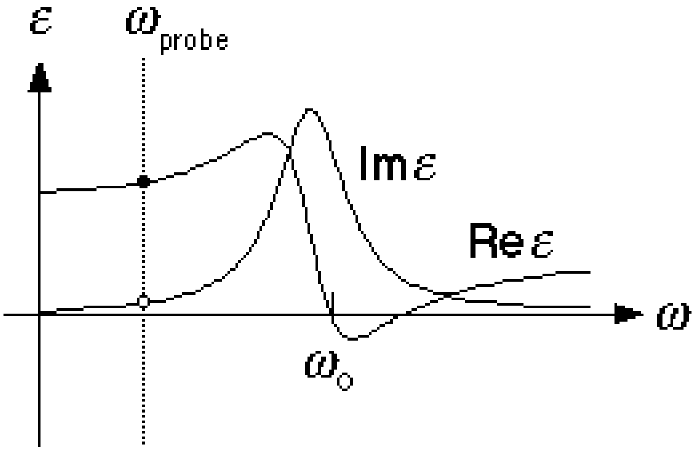


$\omega_{probe} = 2.2$ eV

WHAT IS HAPPENING?

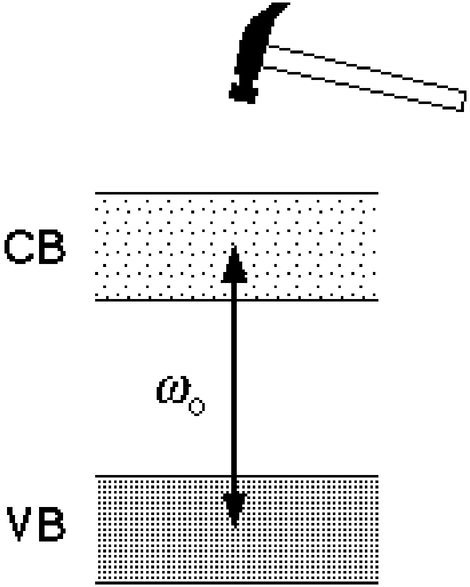


semiconductor

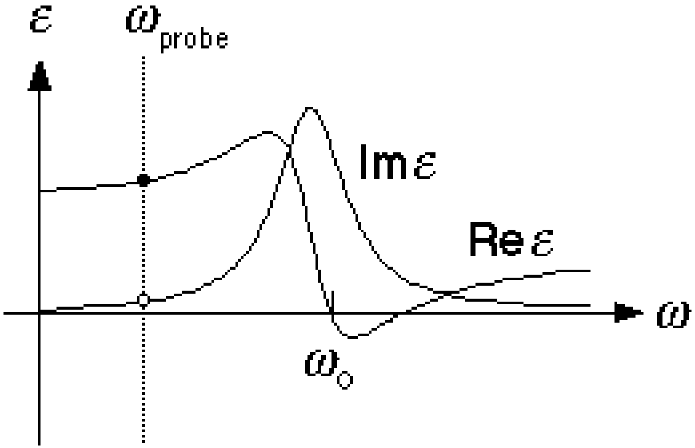


pump pulse excites valence electrons...

WHAT IS HAPPENING?



semiconductor



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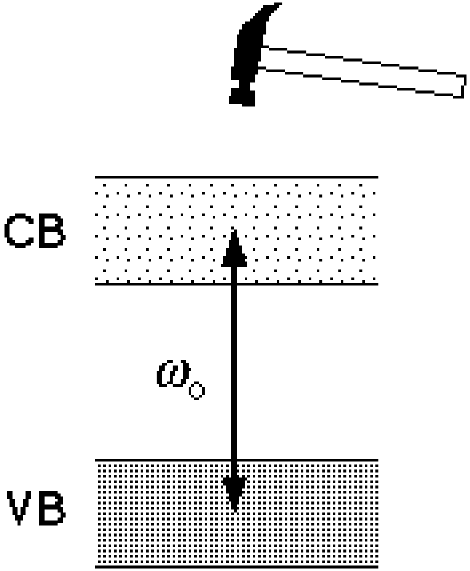
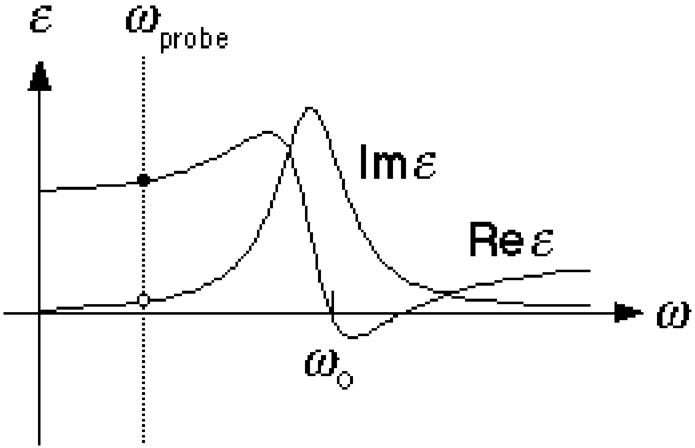


semiconductor



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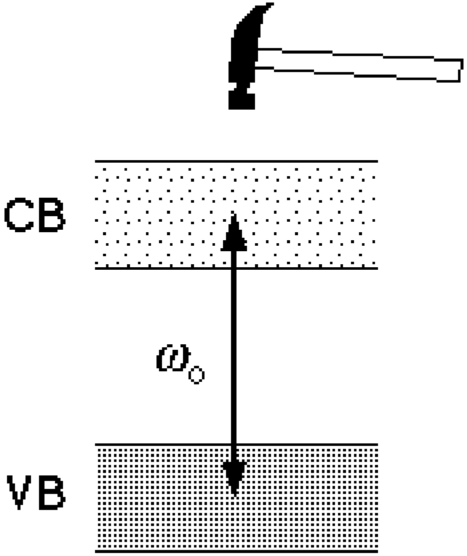

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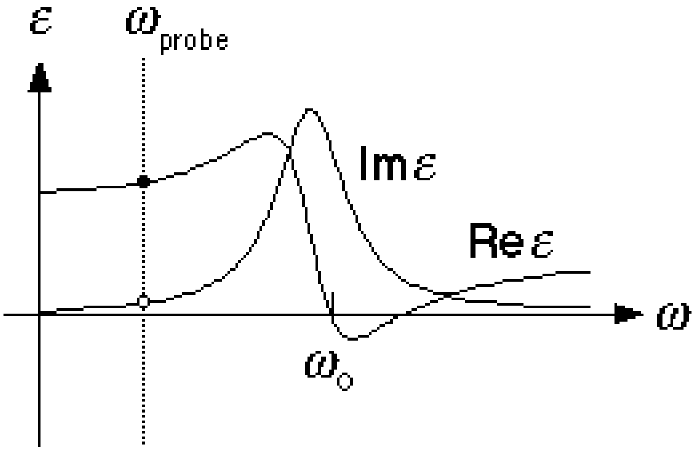

pump pulse excites valence electrons...

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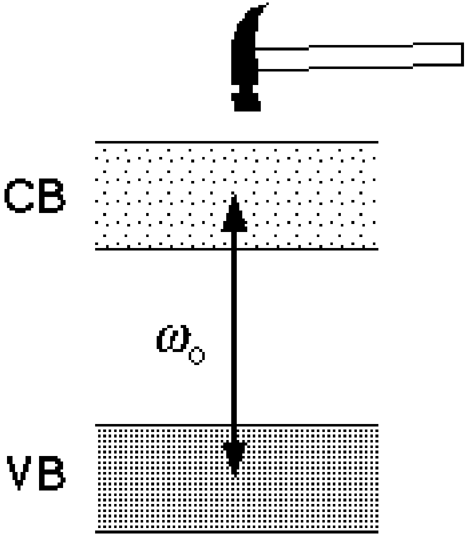
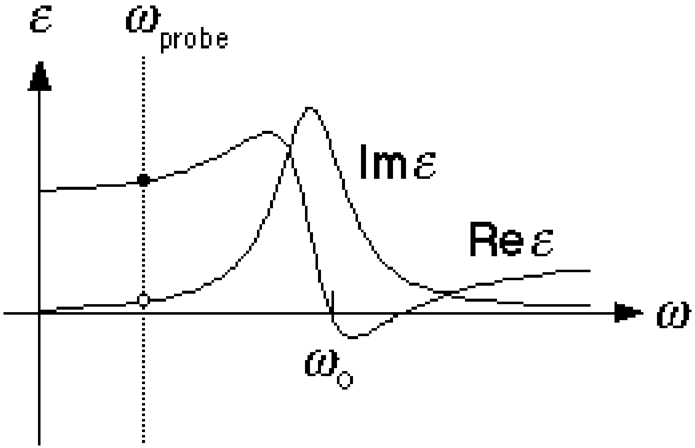


semiconductor



pump pulse excites valence electrons...

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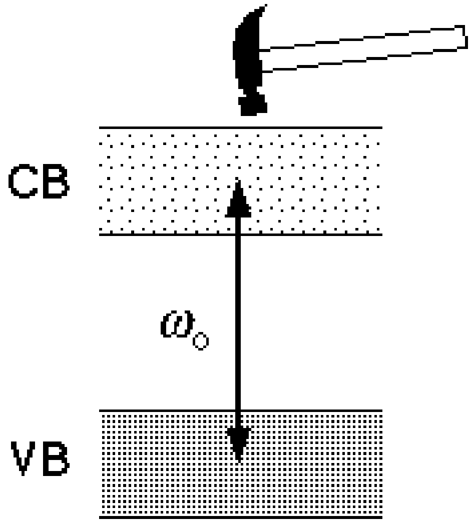
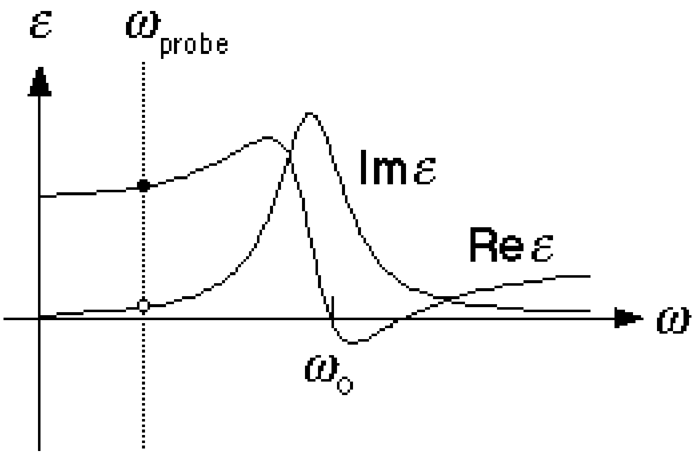


semiconductor



pump pulse excites valence electrons...

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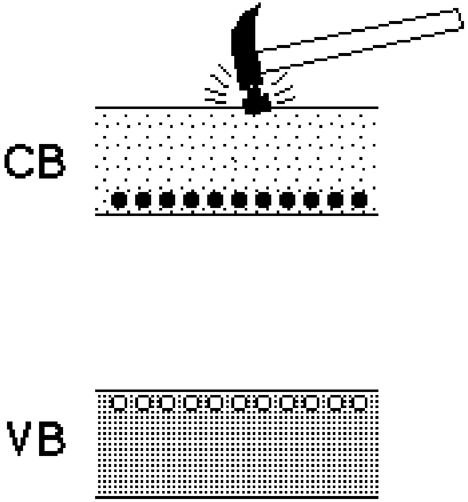

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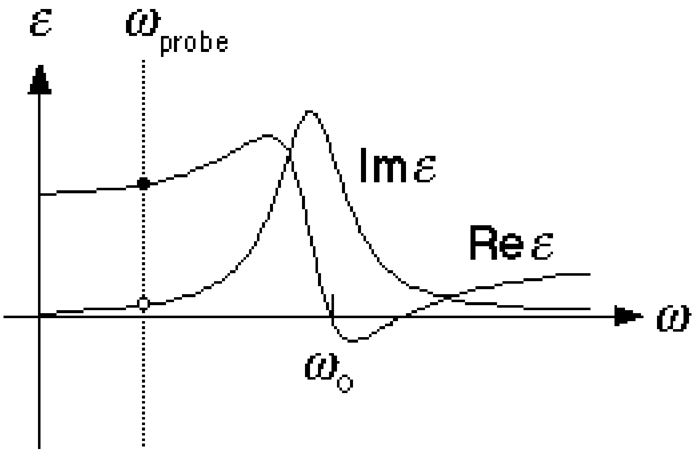
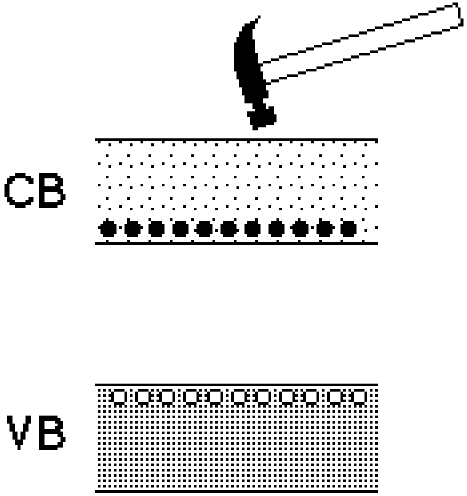


pump pulse excites valence electrons...

WHAT IS HAPPENING?



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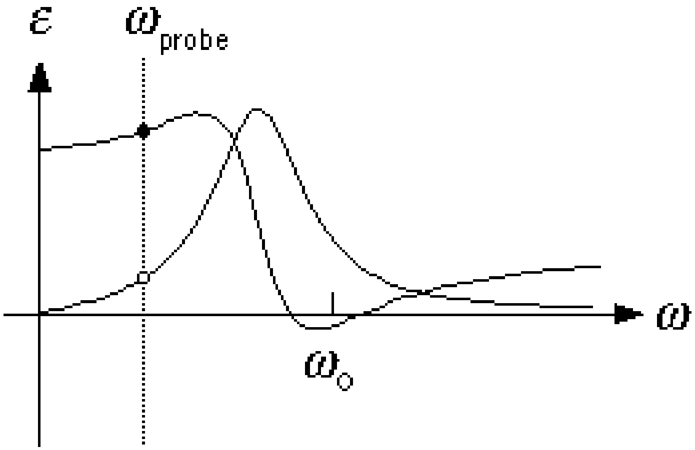
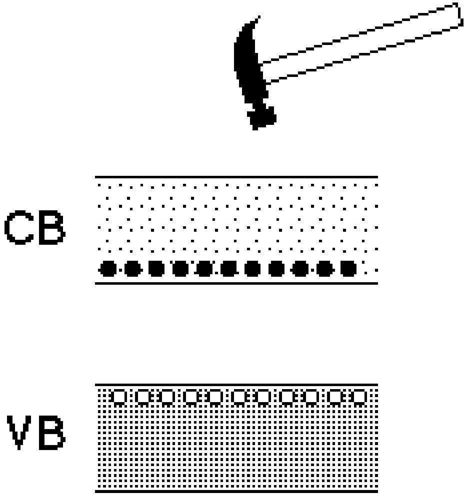


...and induces change in bandstructure

WHAT IS HAPPENING?



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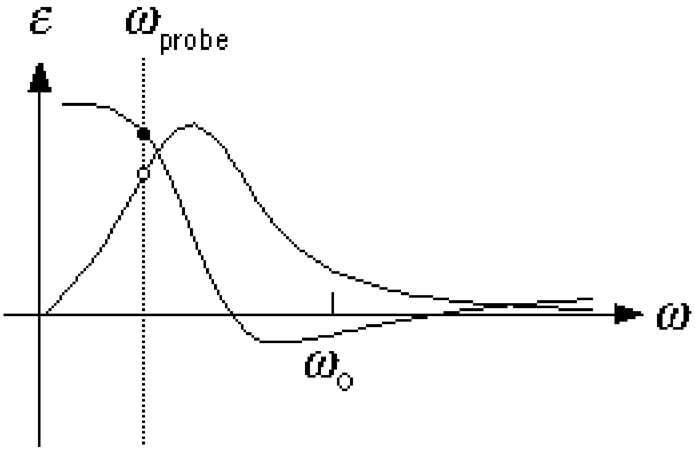

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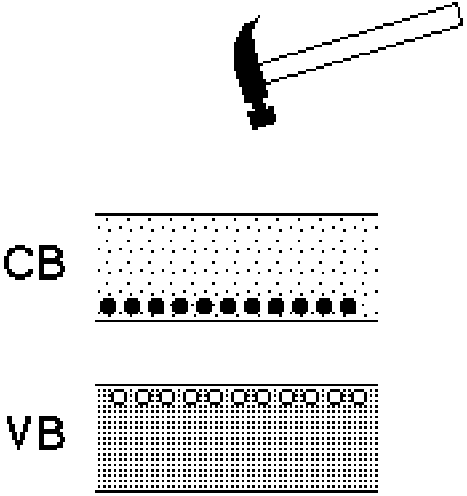

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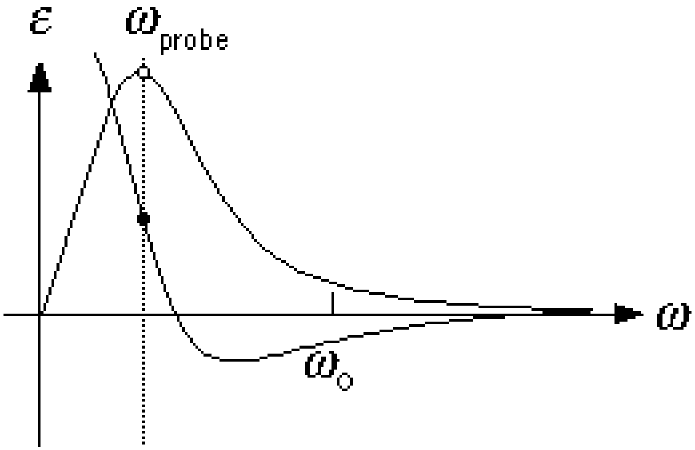

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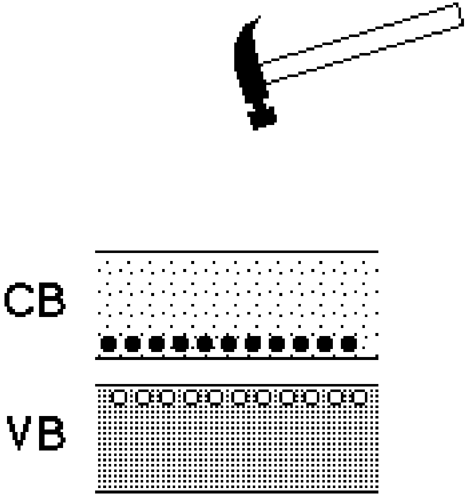

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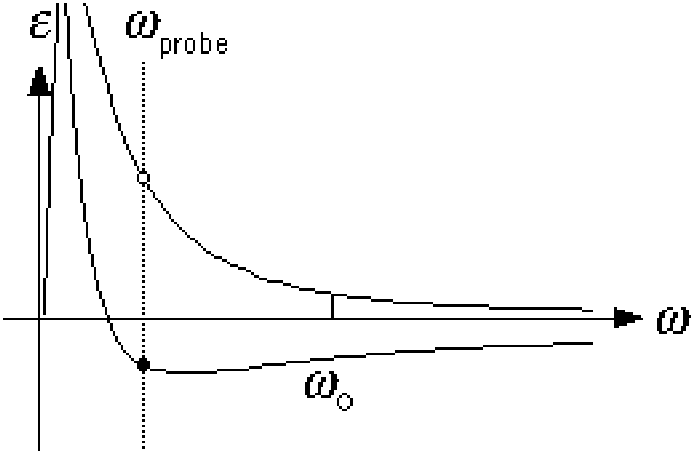
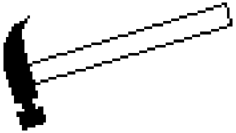
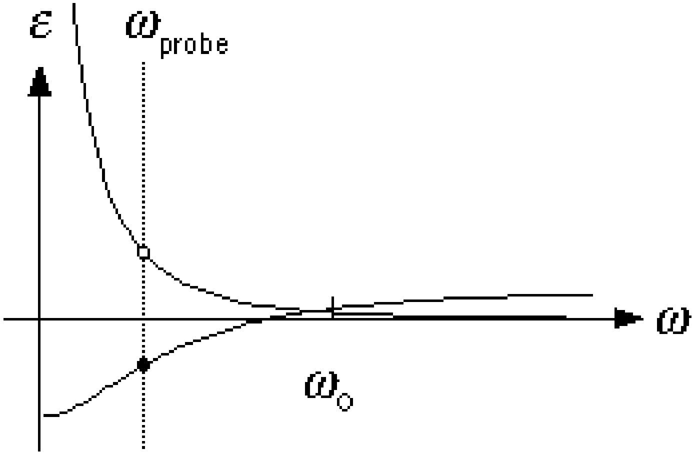
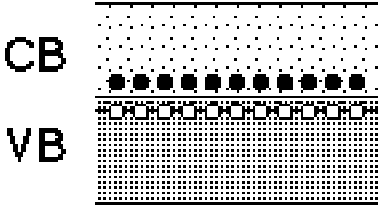


...and induces change in bandstructure

WHAT IS HAPPENING?



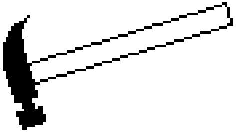


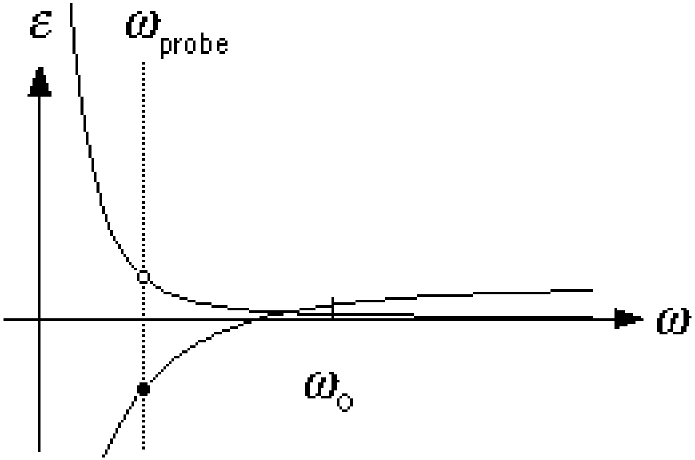


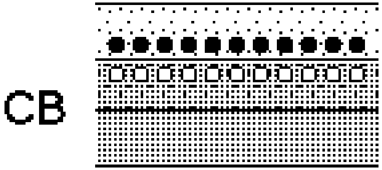
semimetal

...and induces change in bandstructure

WHAT IS HAPPENING?



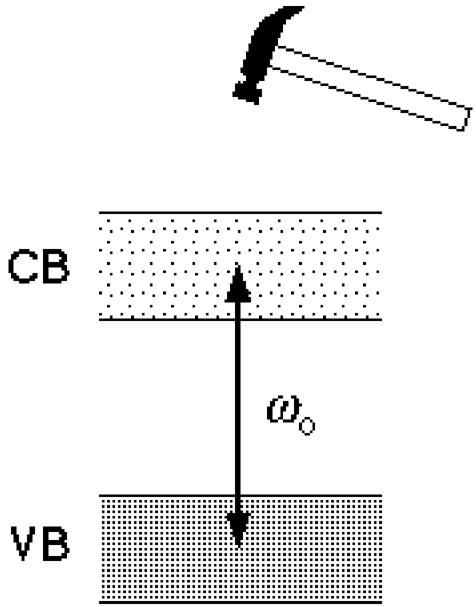




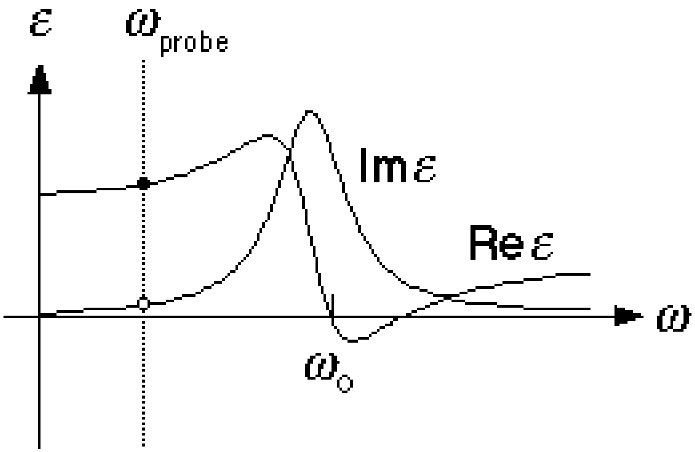
metal

...and induces change in bandstructure

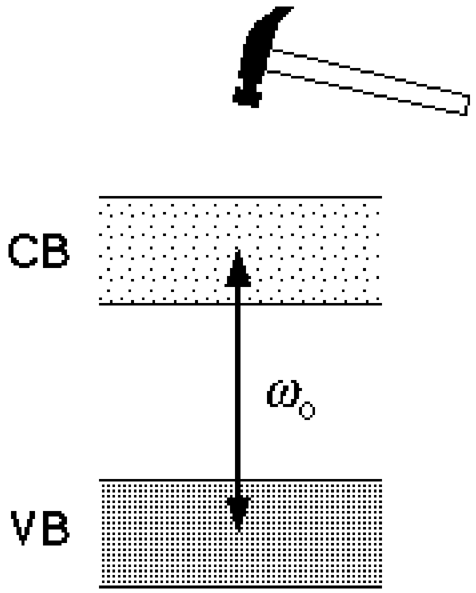
WHAT IS HAPPENING?



semiconductor



WHAT IS HAPPENING?



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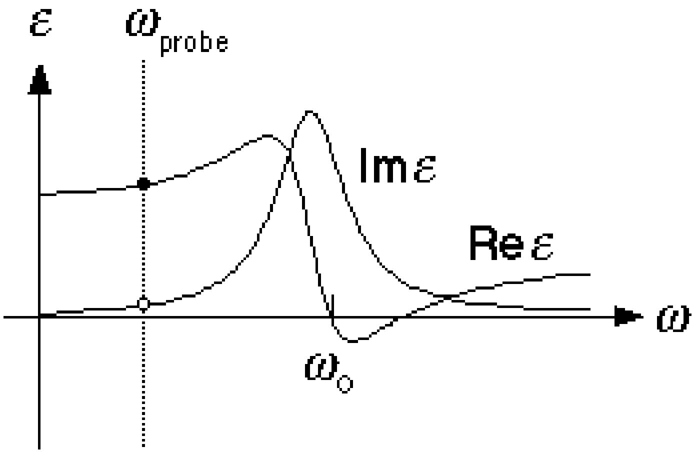

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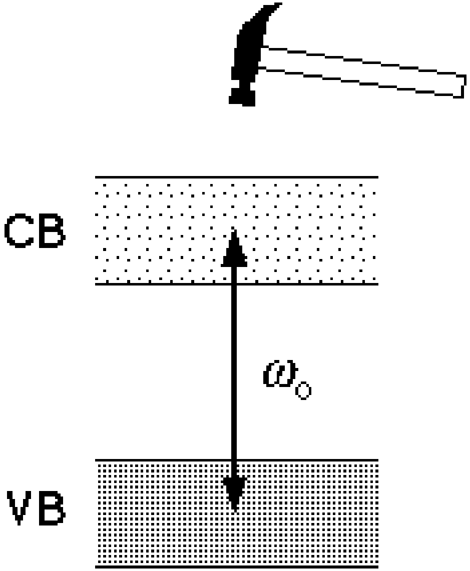

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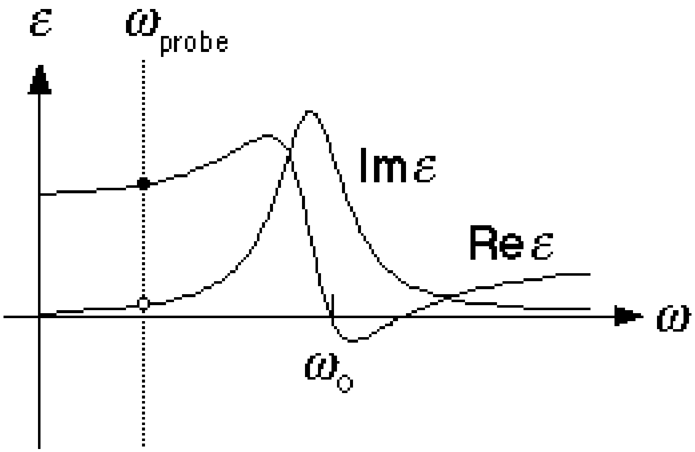
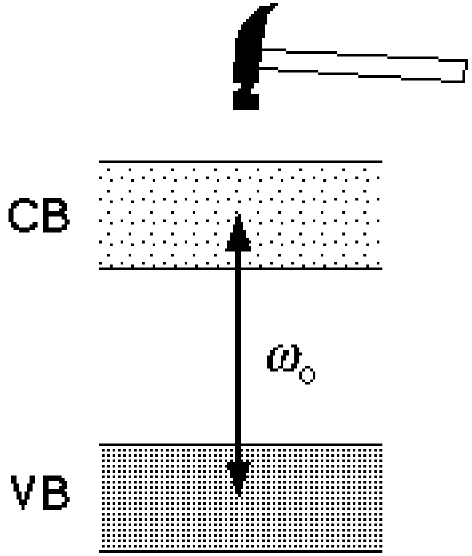


WHAT IS HAPPENING?



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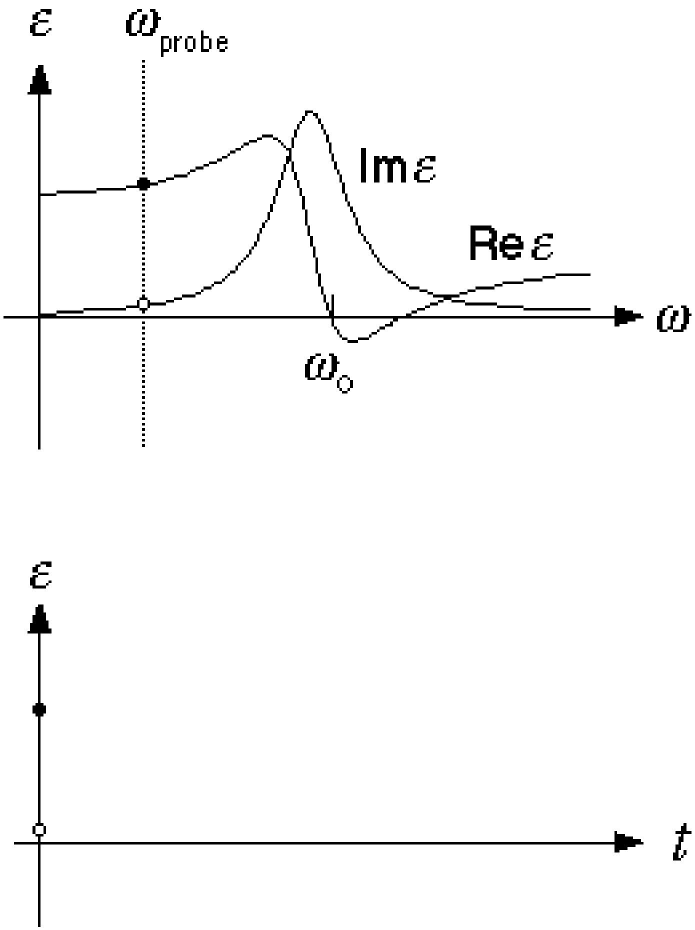

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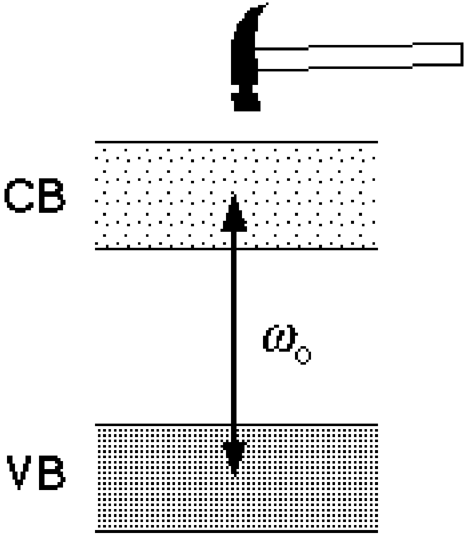

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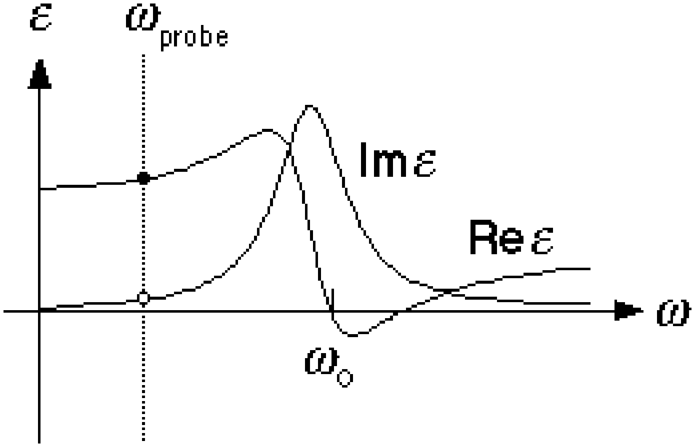
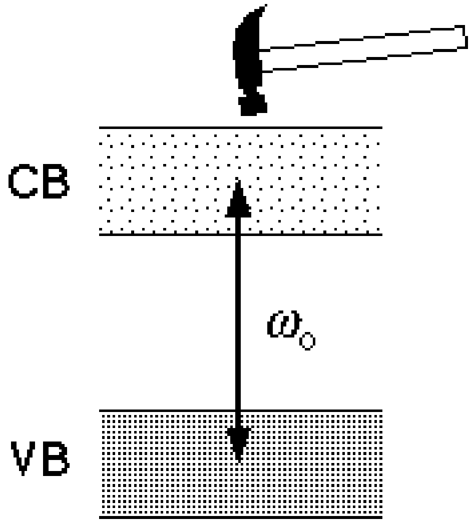


WHAT IS HAPPENING?



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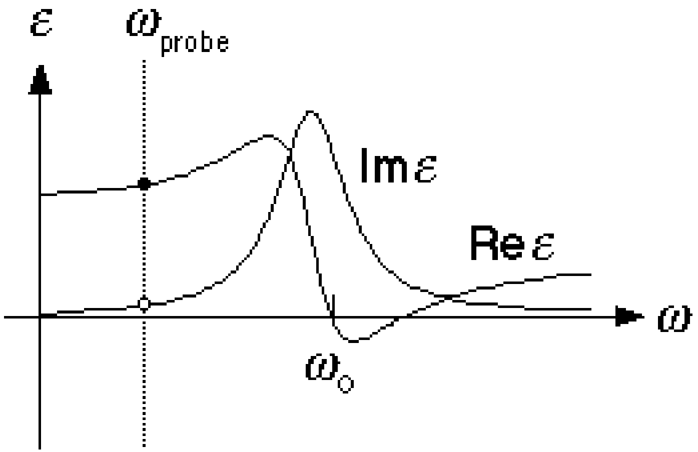
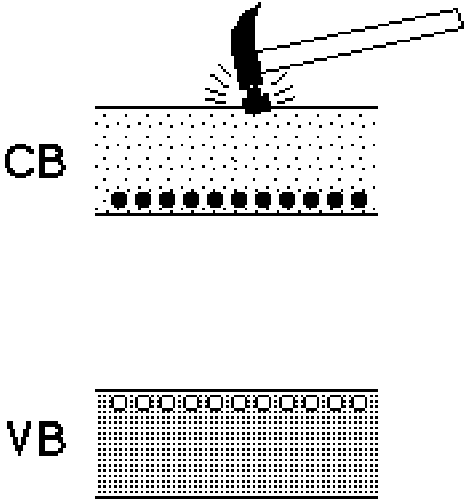
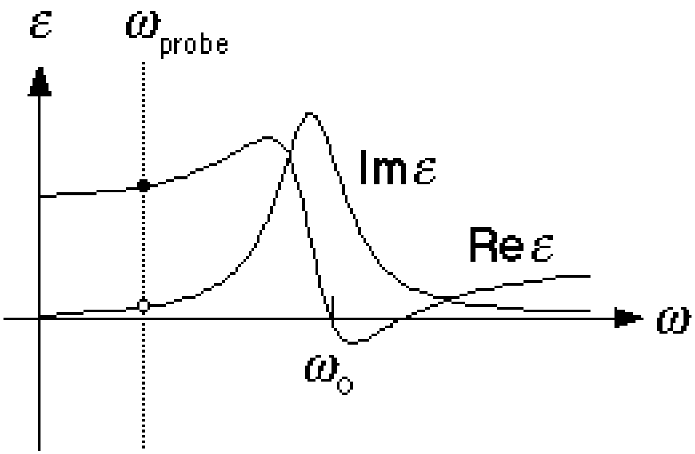


WHAT IS HAPPENING?



semiconductor



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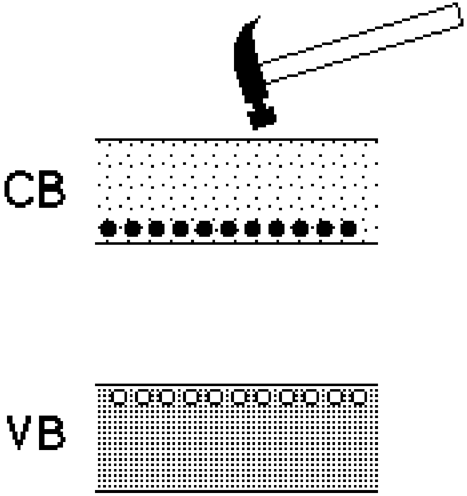

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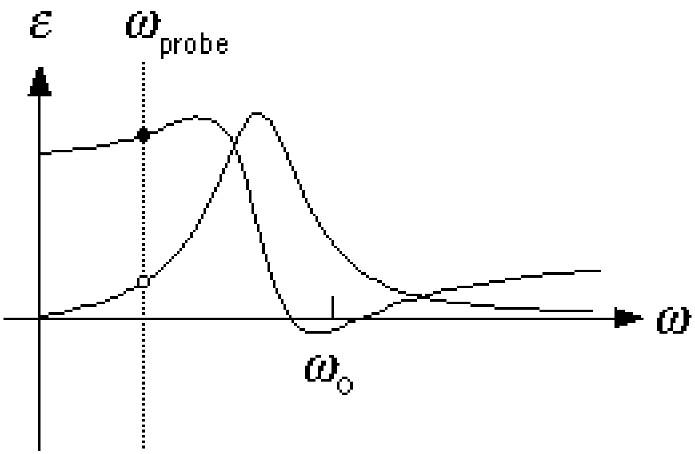

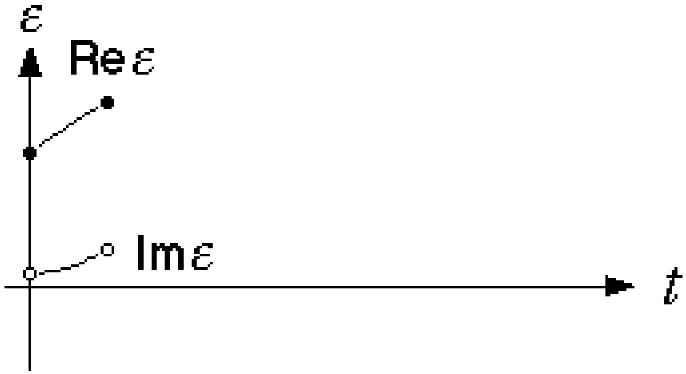

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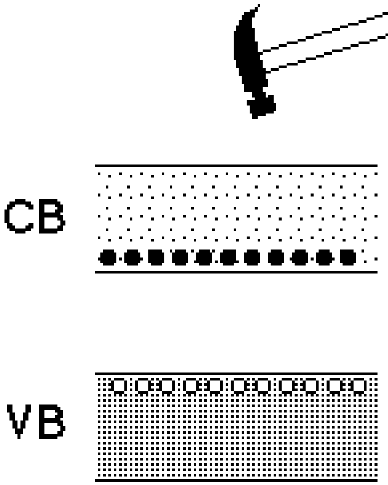

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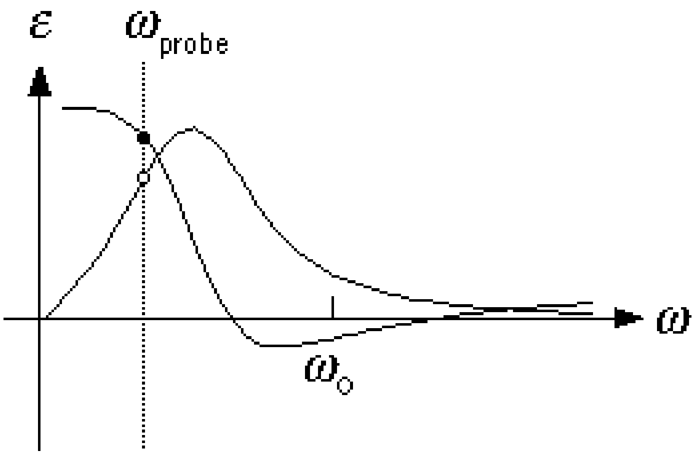

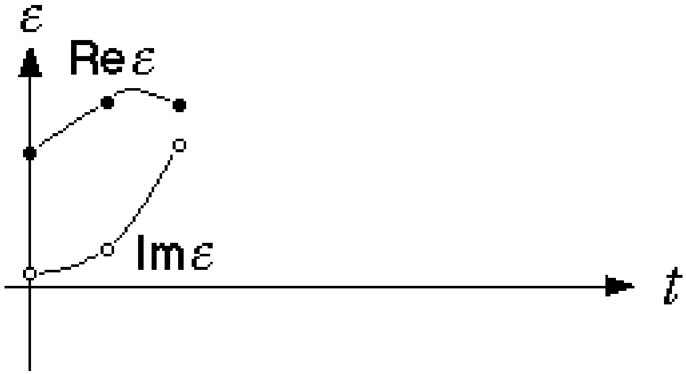

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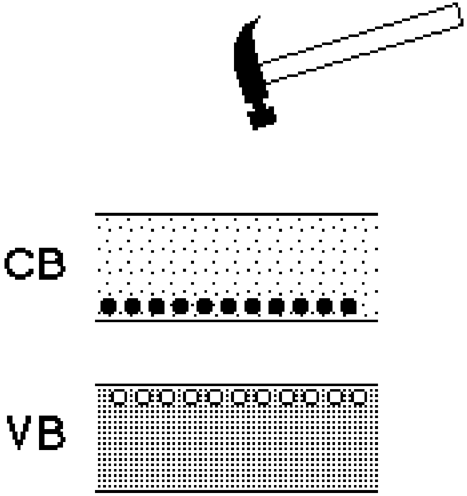

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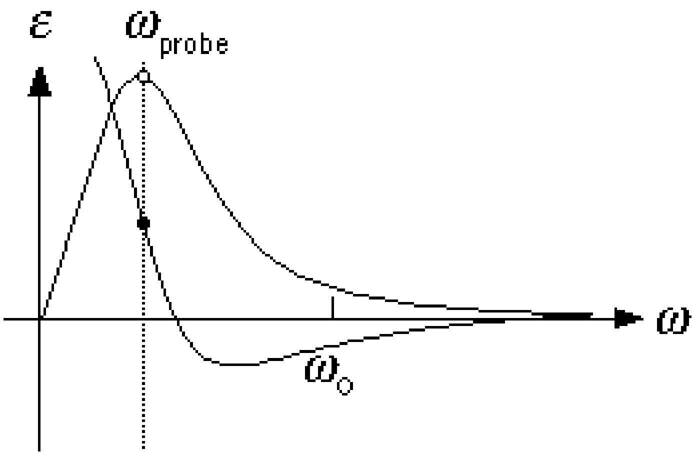

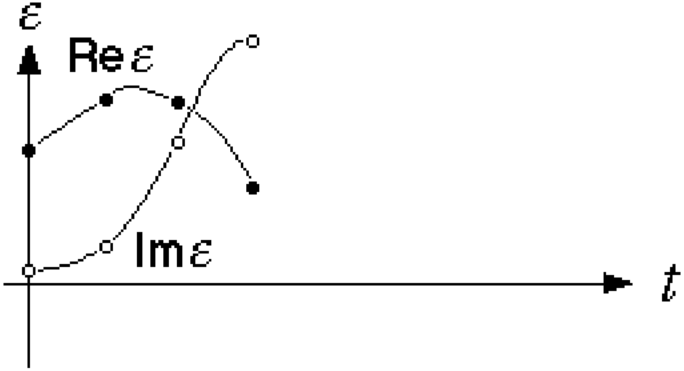

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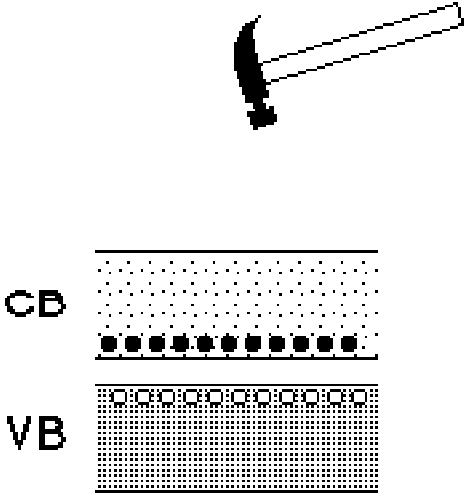

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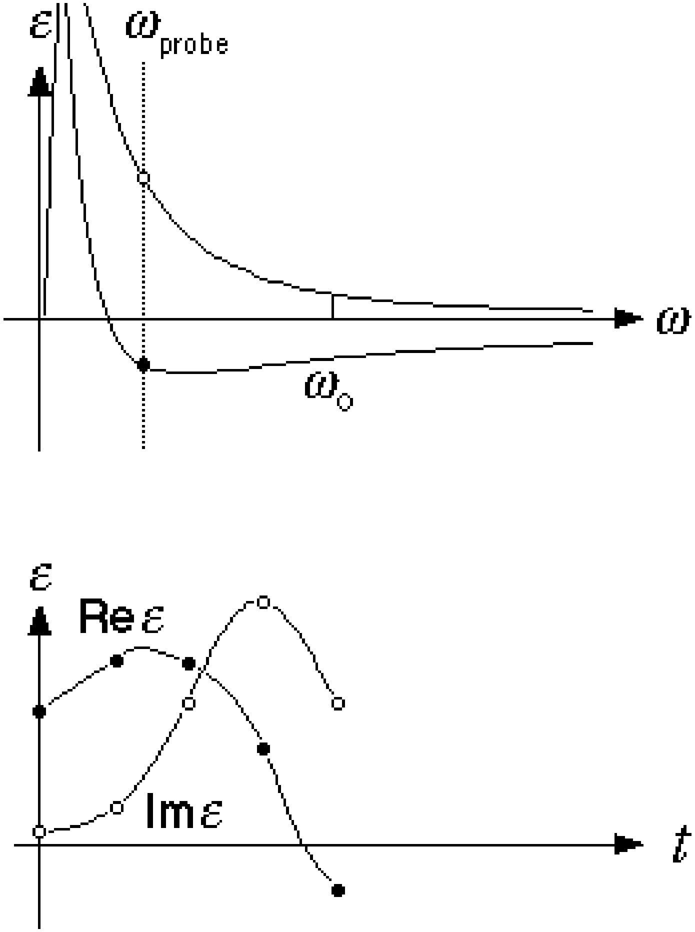

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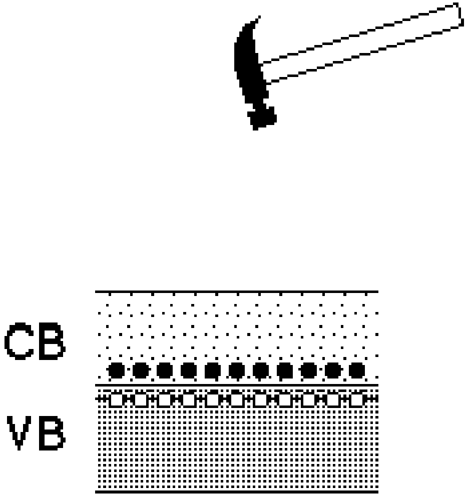

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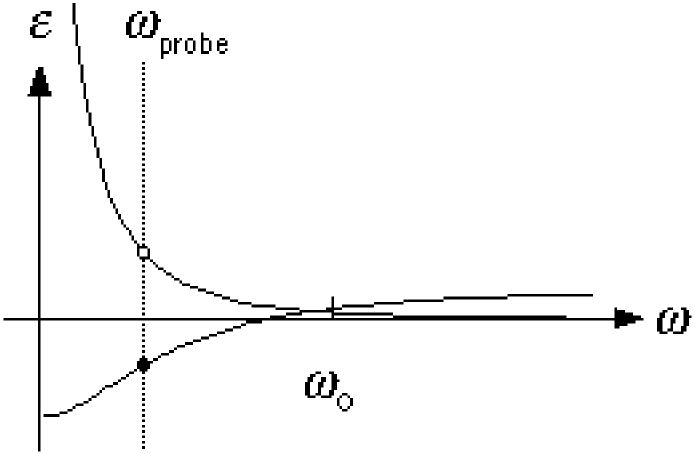

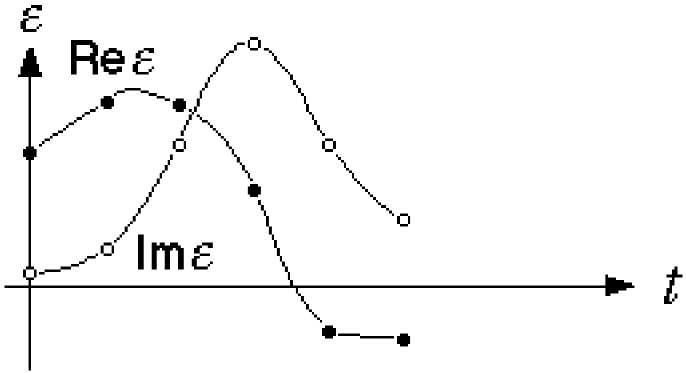

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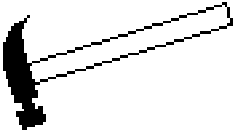

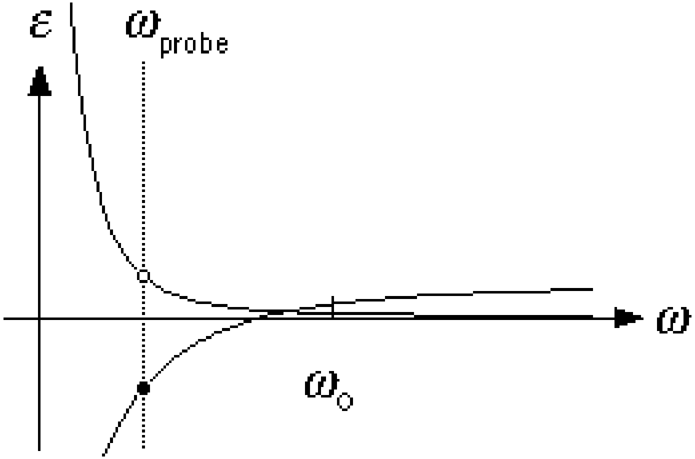

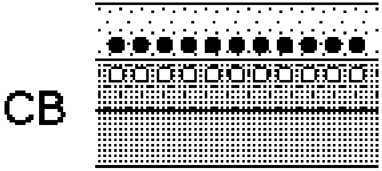

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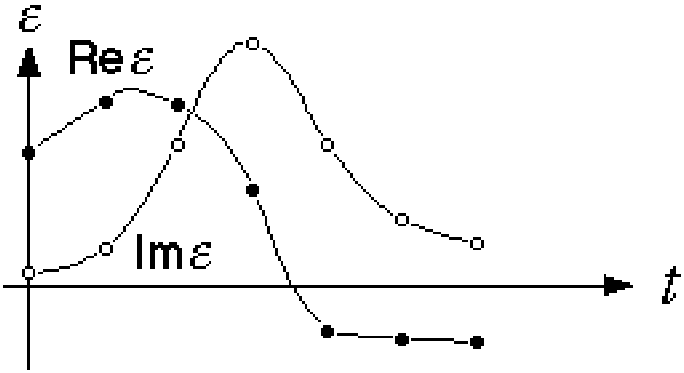
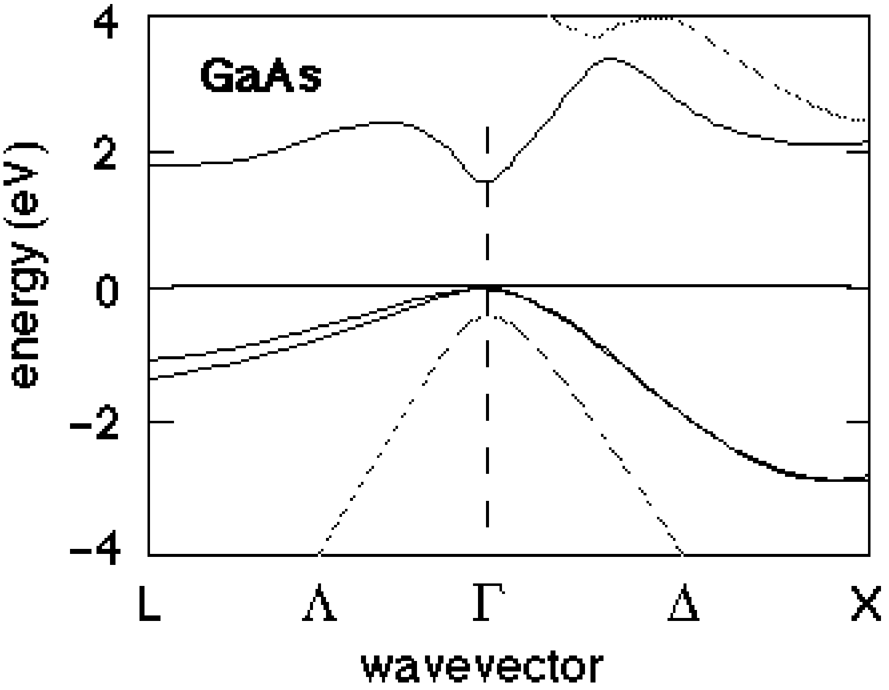
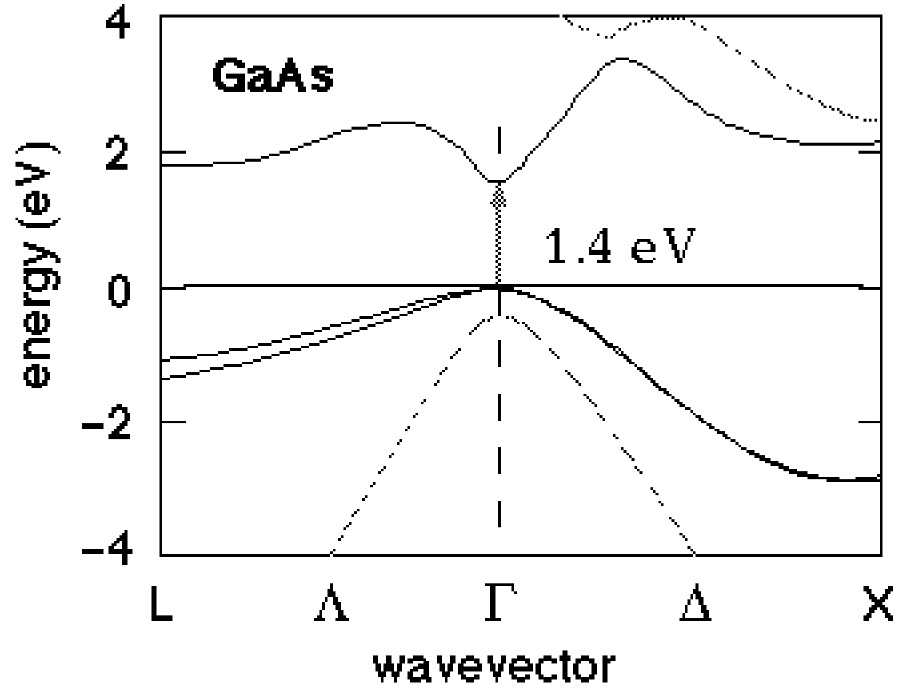


GaAs BANDSTRUCTURE



GaAs BANDSTRUCTURE



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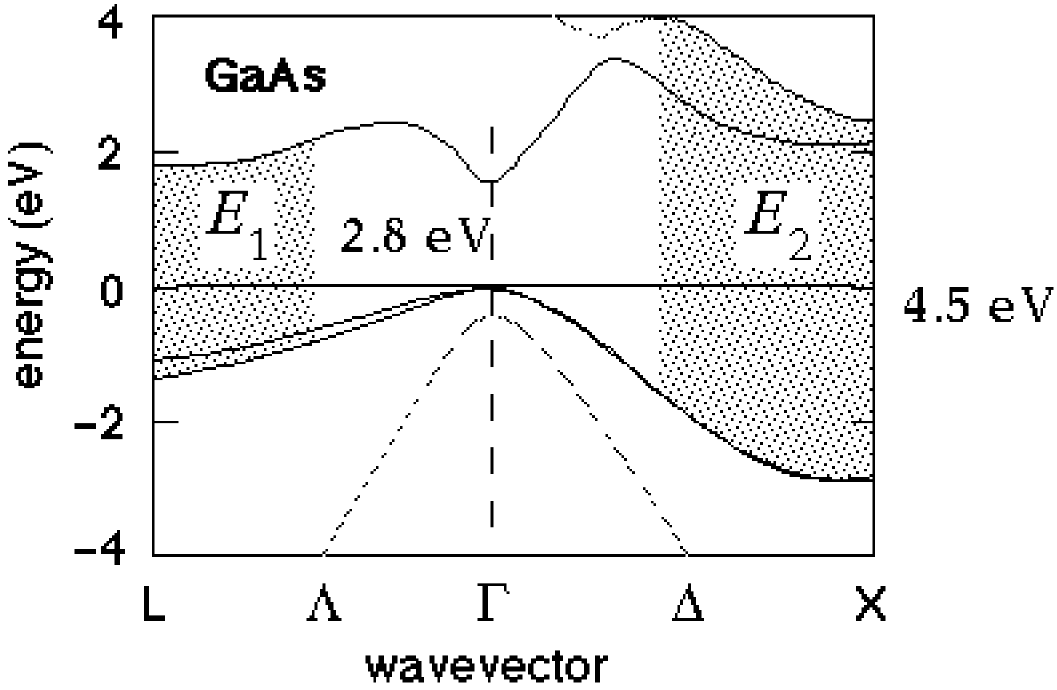

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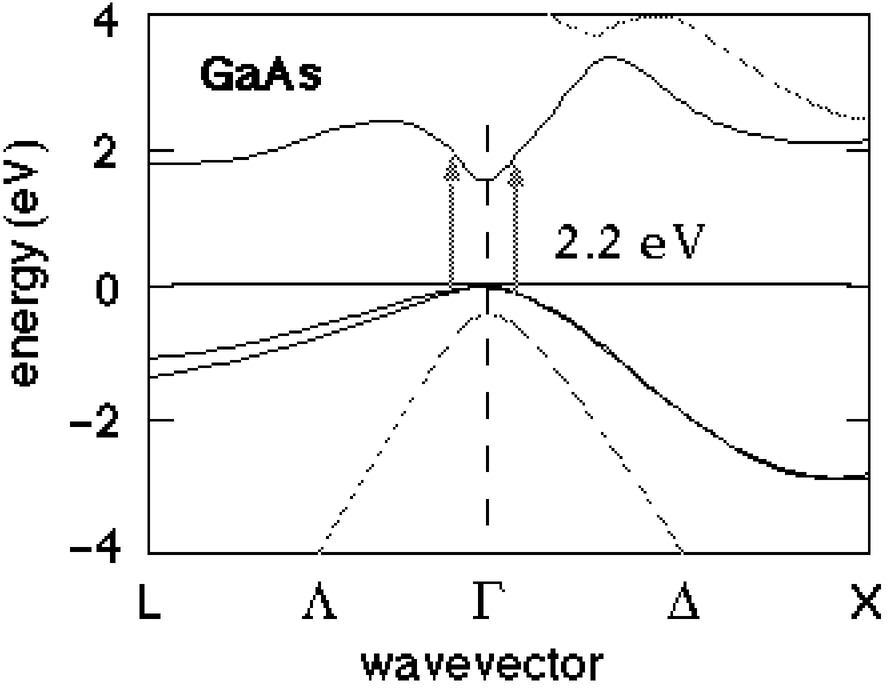

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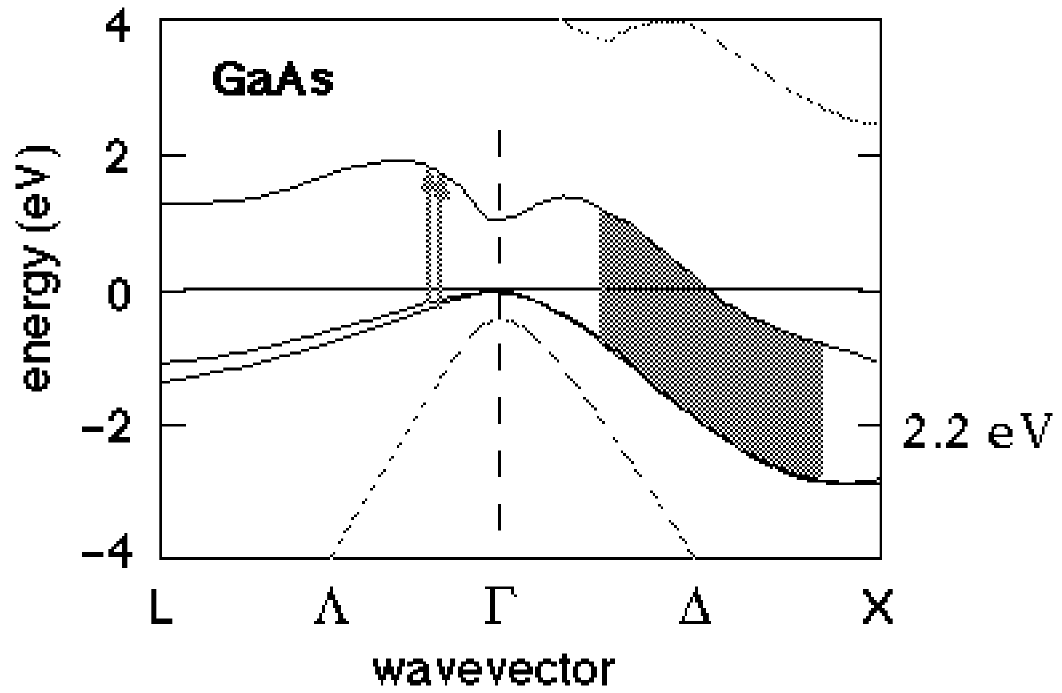
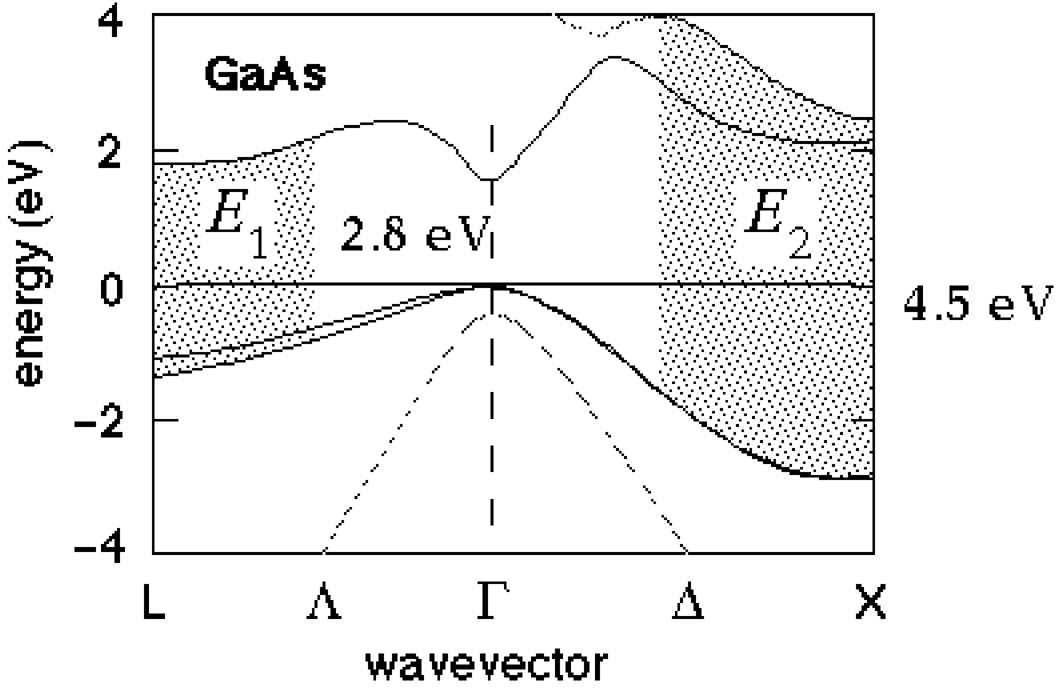


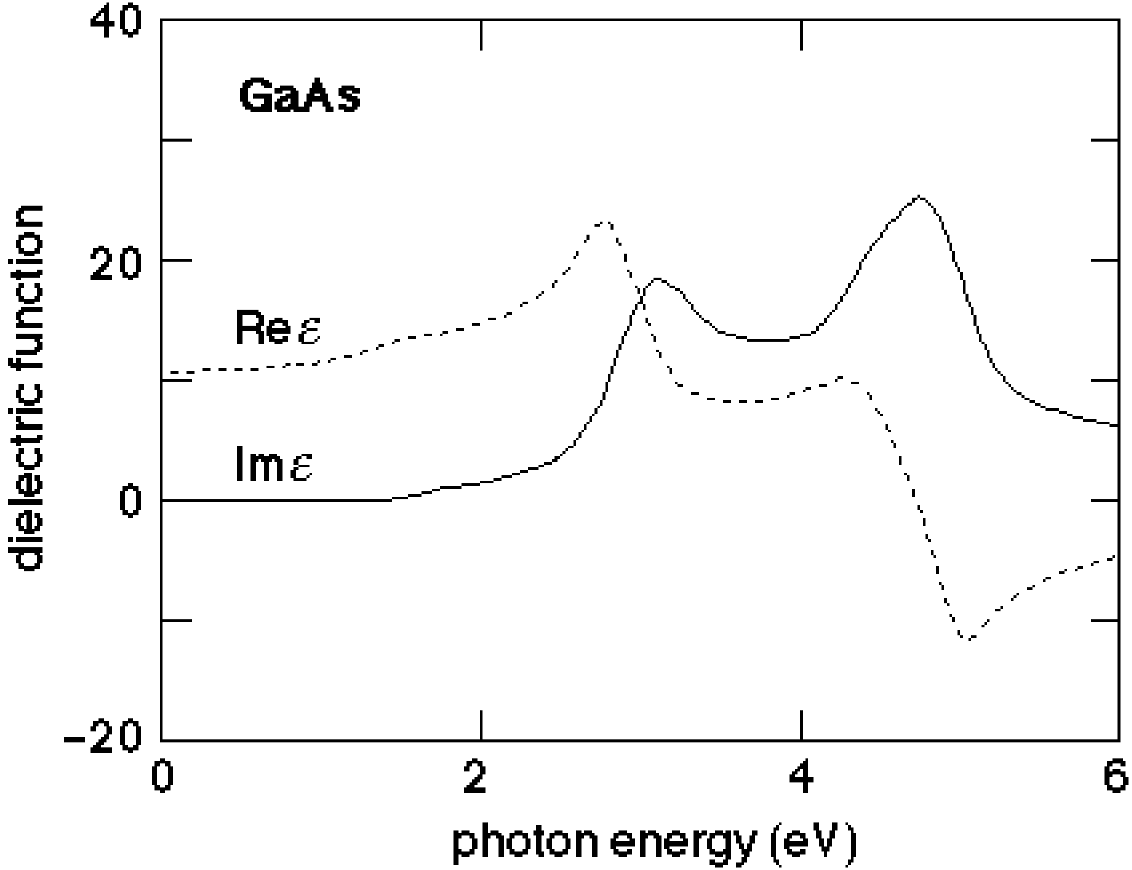
S. Froyen *et al.*, *Phys Rev. B* **28** (1983) 3258

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

GaAs BANDSTRUCTURE



GaAs DIELECTRIC FUNCTION



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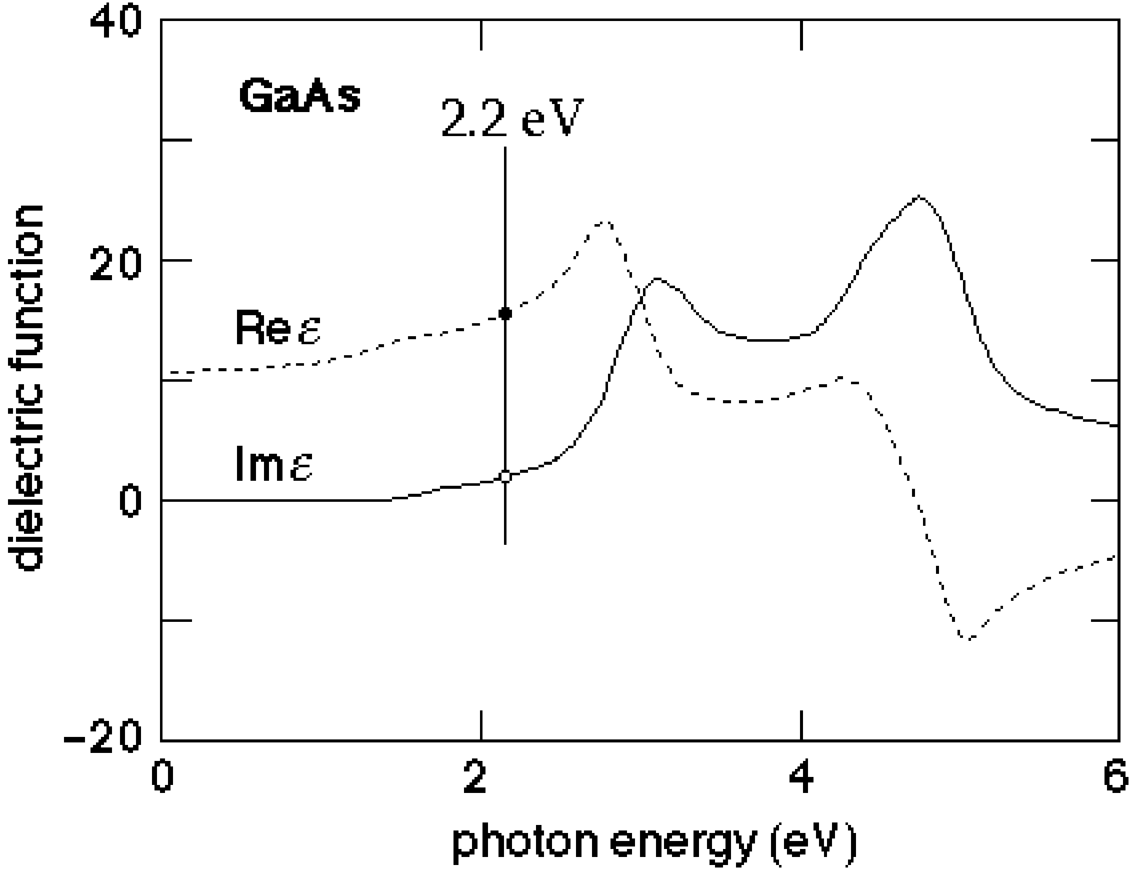
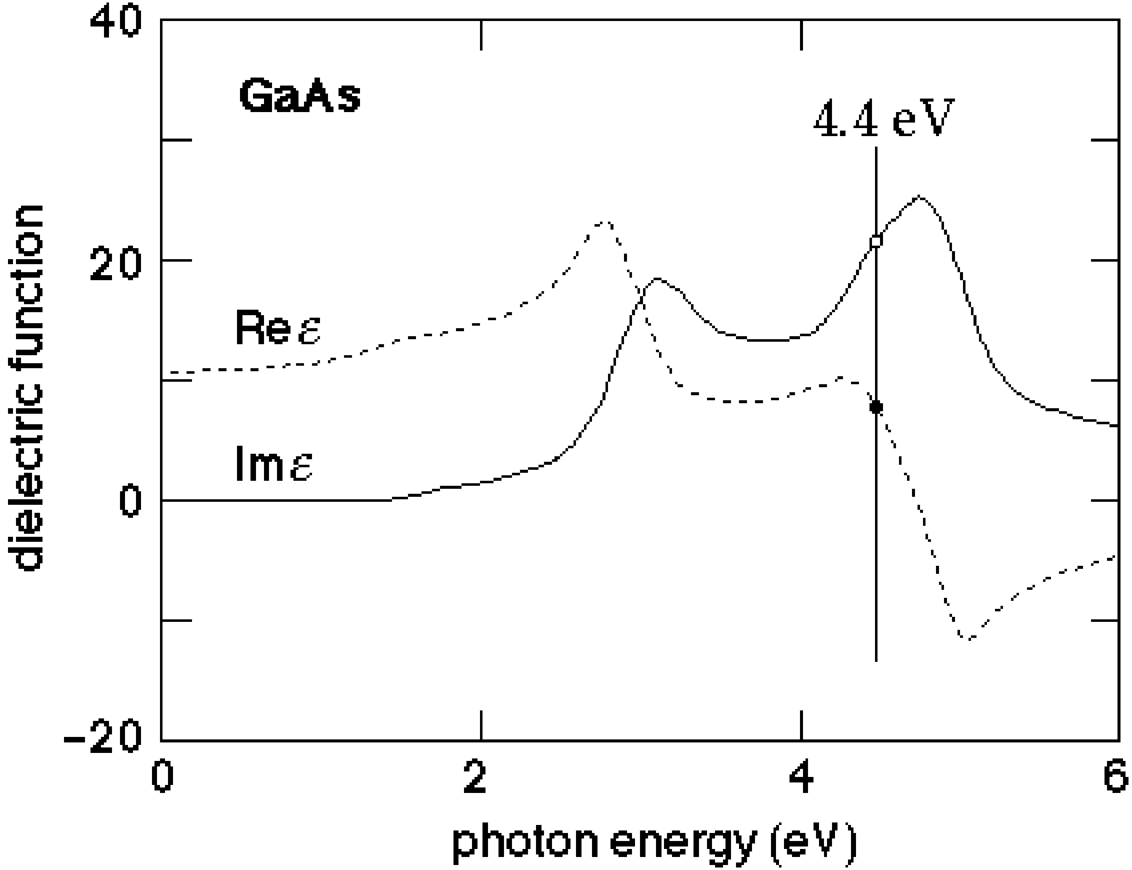
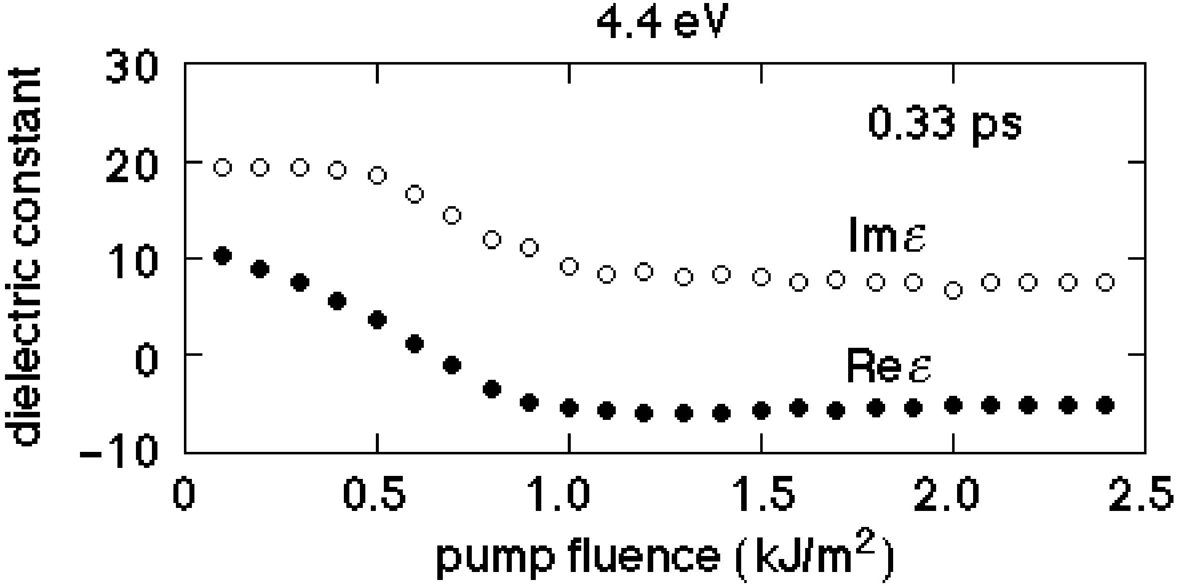


GaAs DIELECTRIC FUNCTION



FLUENCE DEPENDENCE



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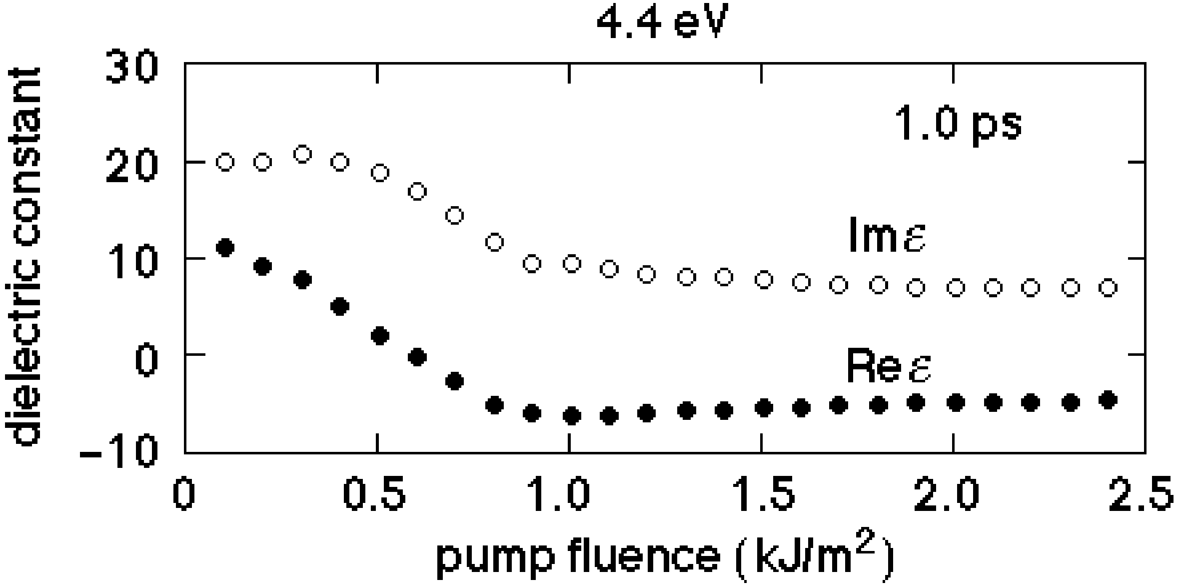

Phys. Rev. B **51** (1995) 6959

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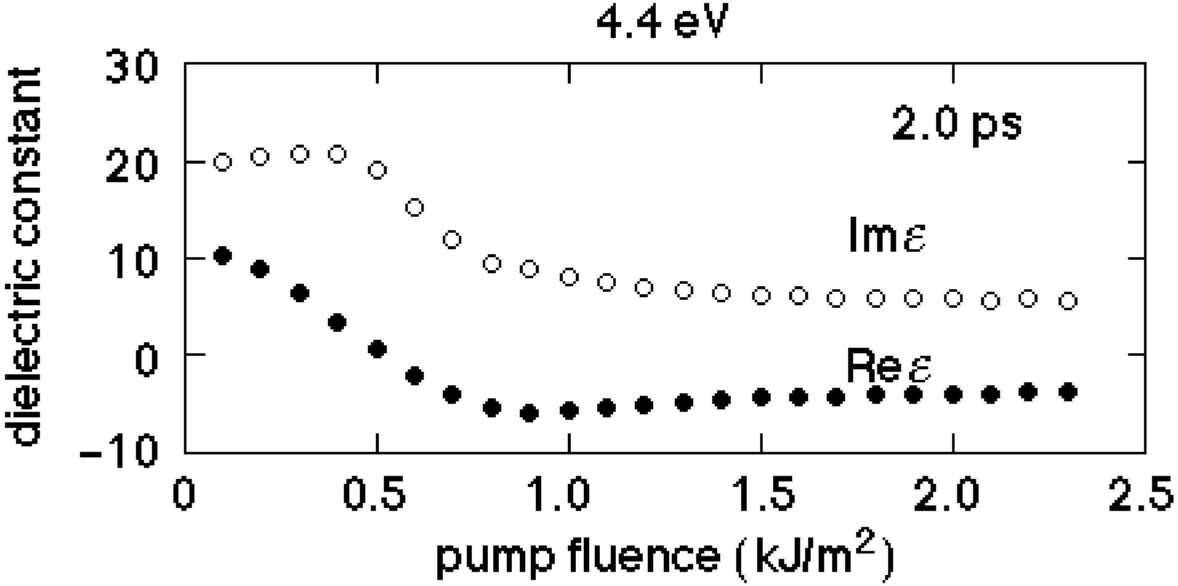

Phys. Rev. B **51** (1995) 6959

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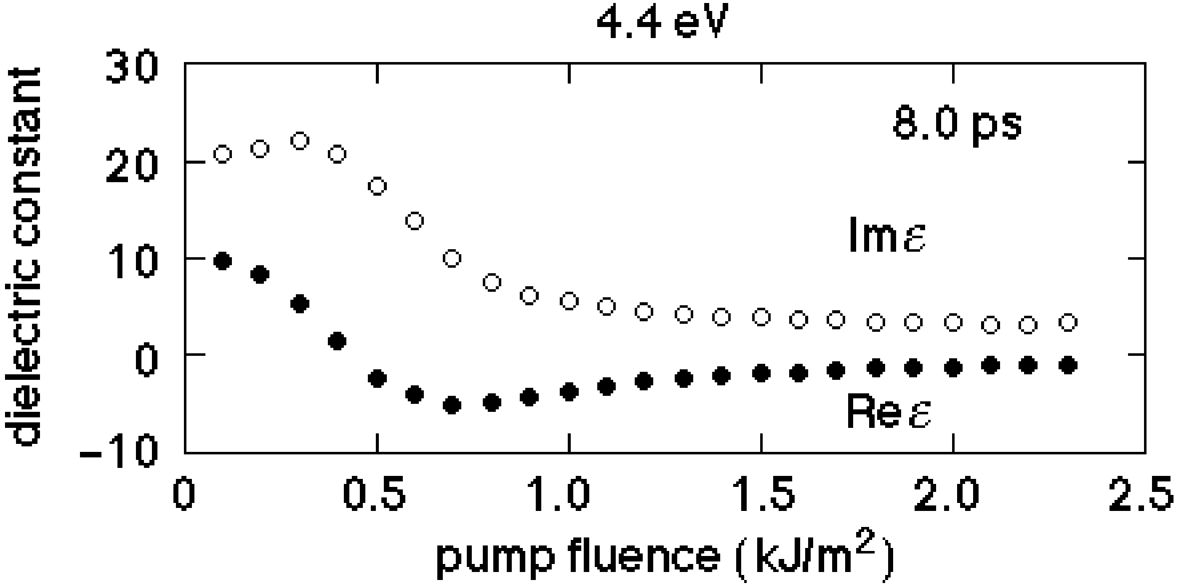

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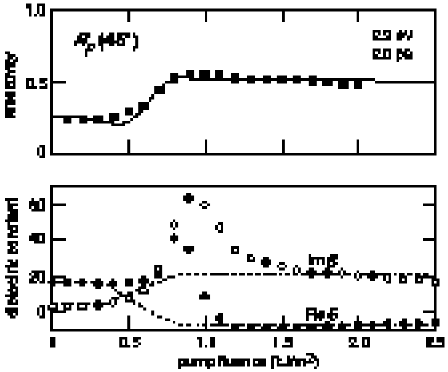
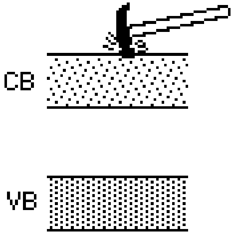
SUMMARY

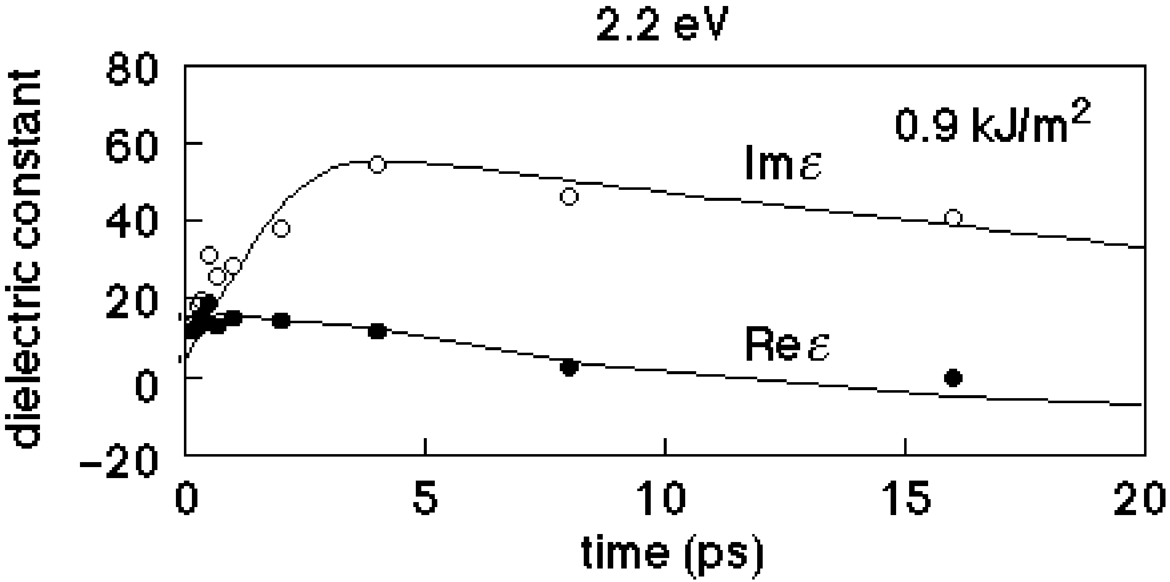


single-angle reflectivity not a good indicator of what happens



laser pulse causes bandgap collapse

CAUSE OF COLLAPSE



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

structural change?

FEMTOSECOND WORK

Field strength at second harmonic:

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

FEMTOSECOND WORK

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:

FEMTOSECOND WORK

Field strength at second harmonic:

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

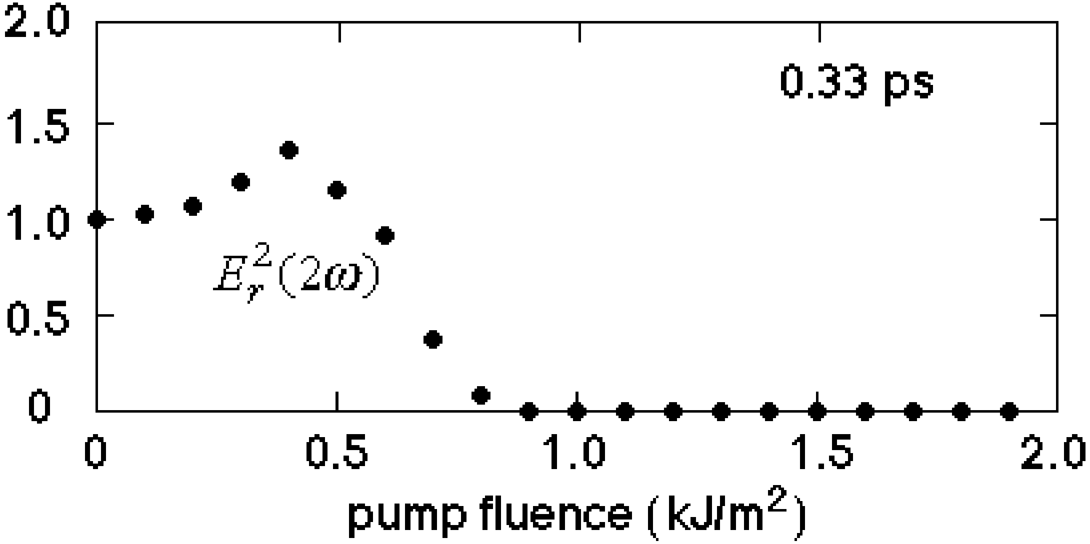
where:

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

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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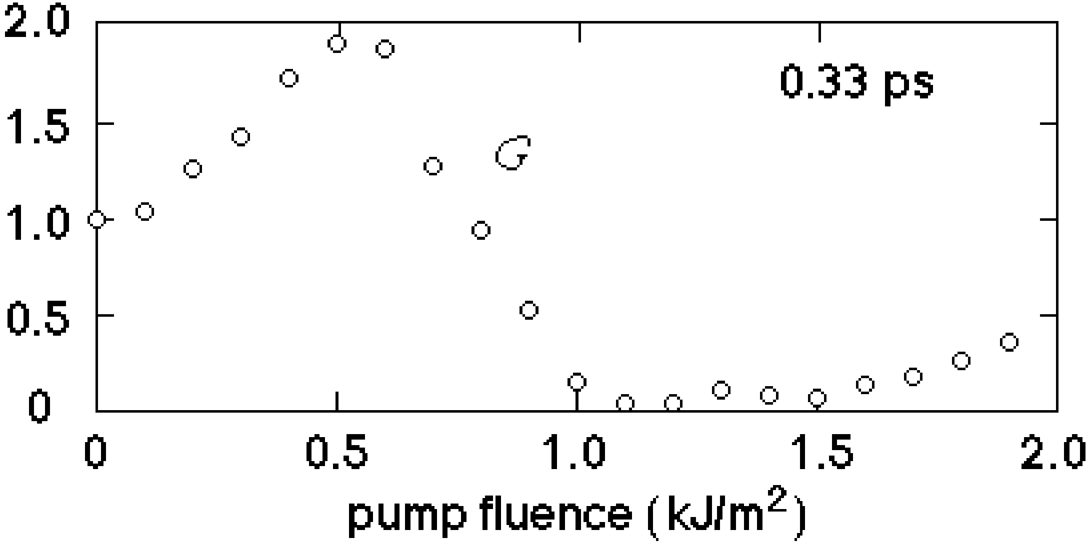

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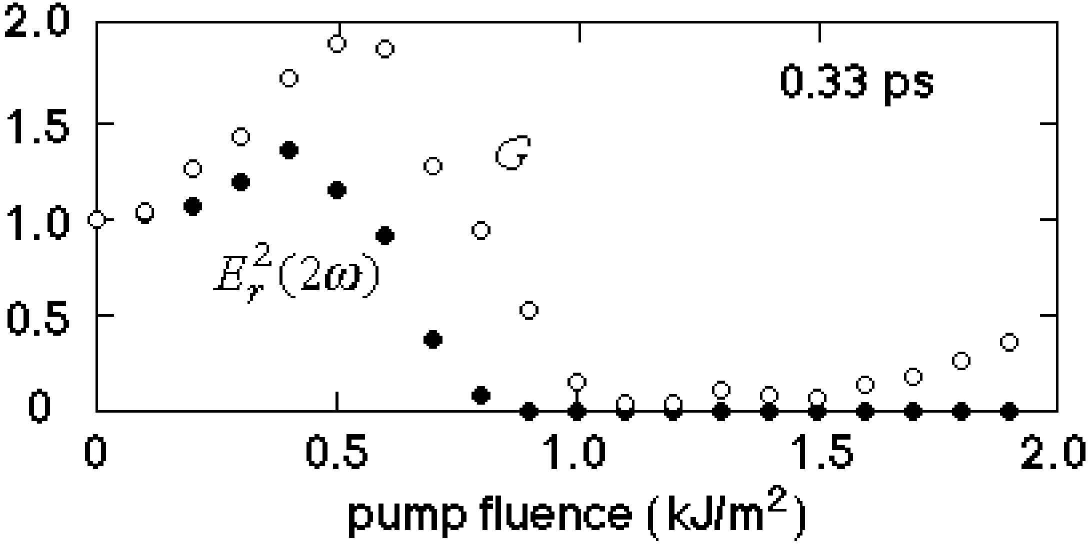

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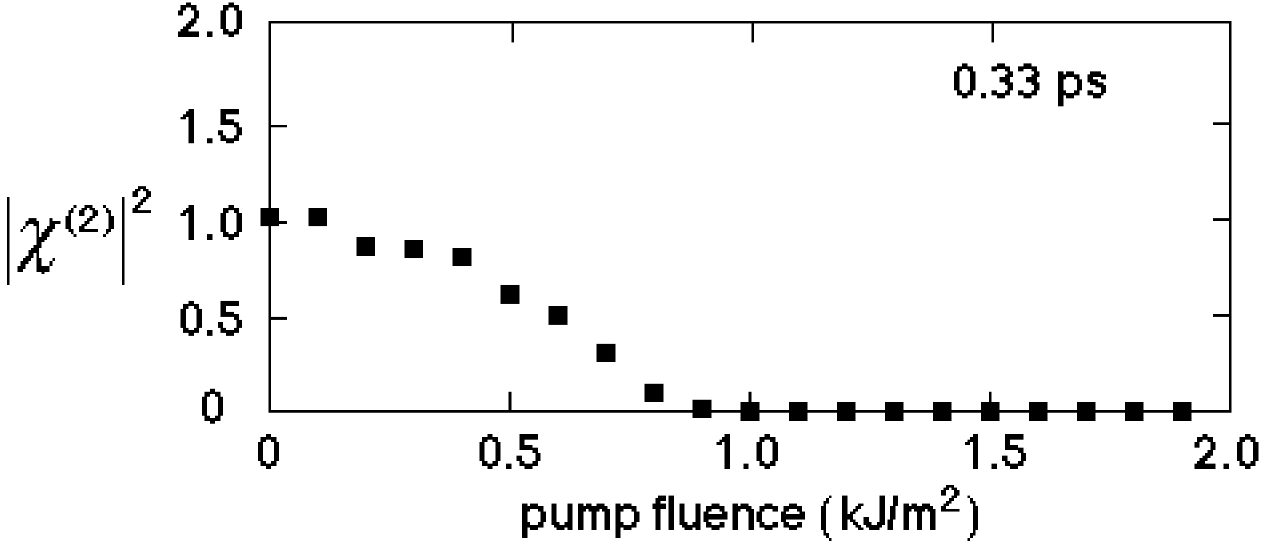

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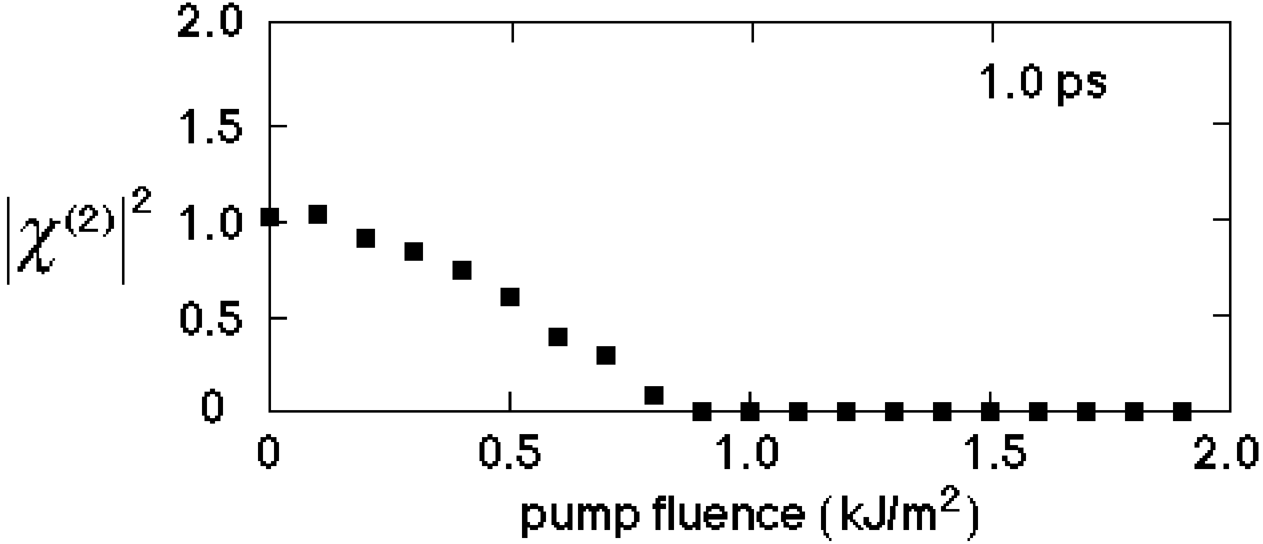

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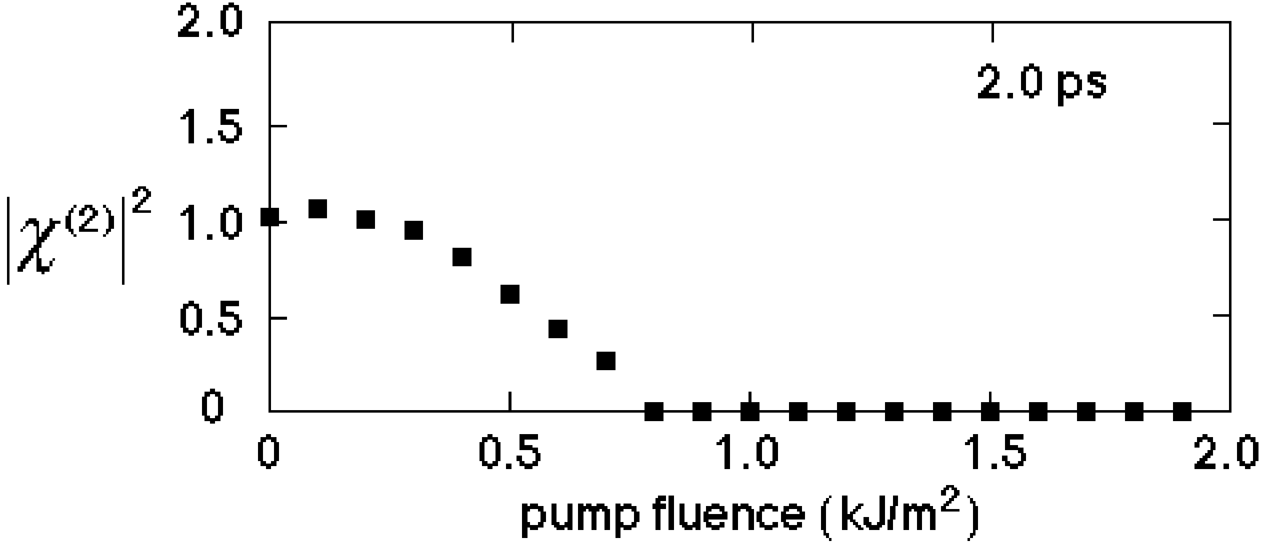

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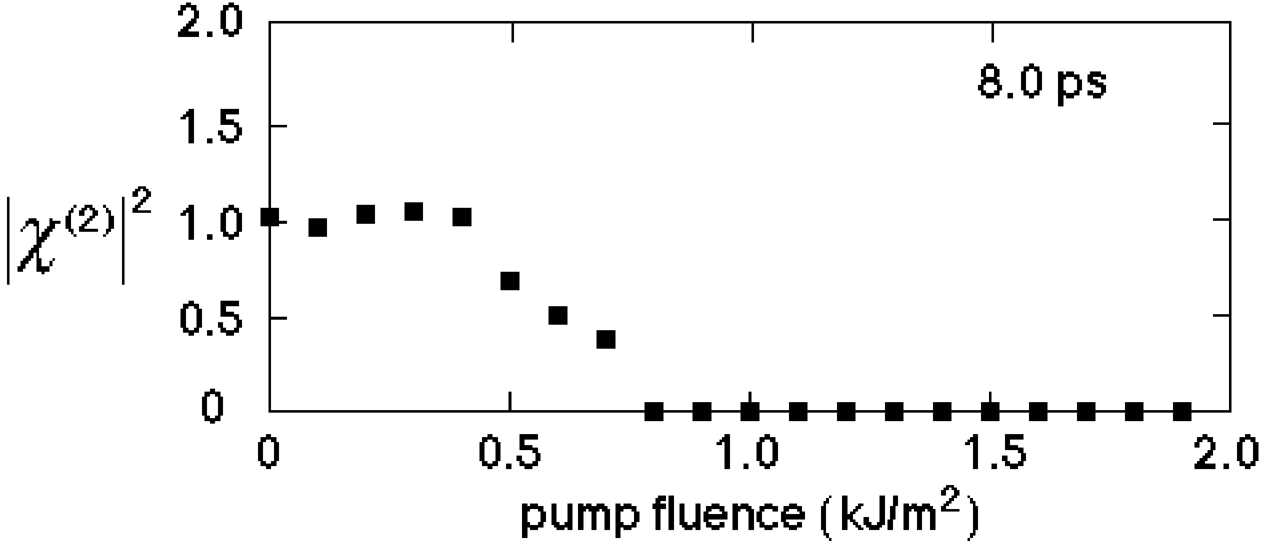

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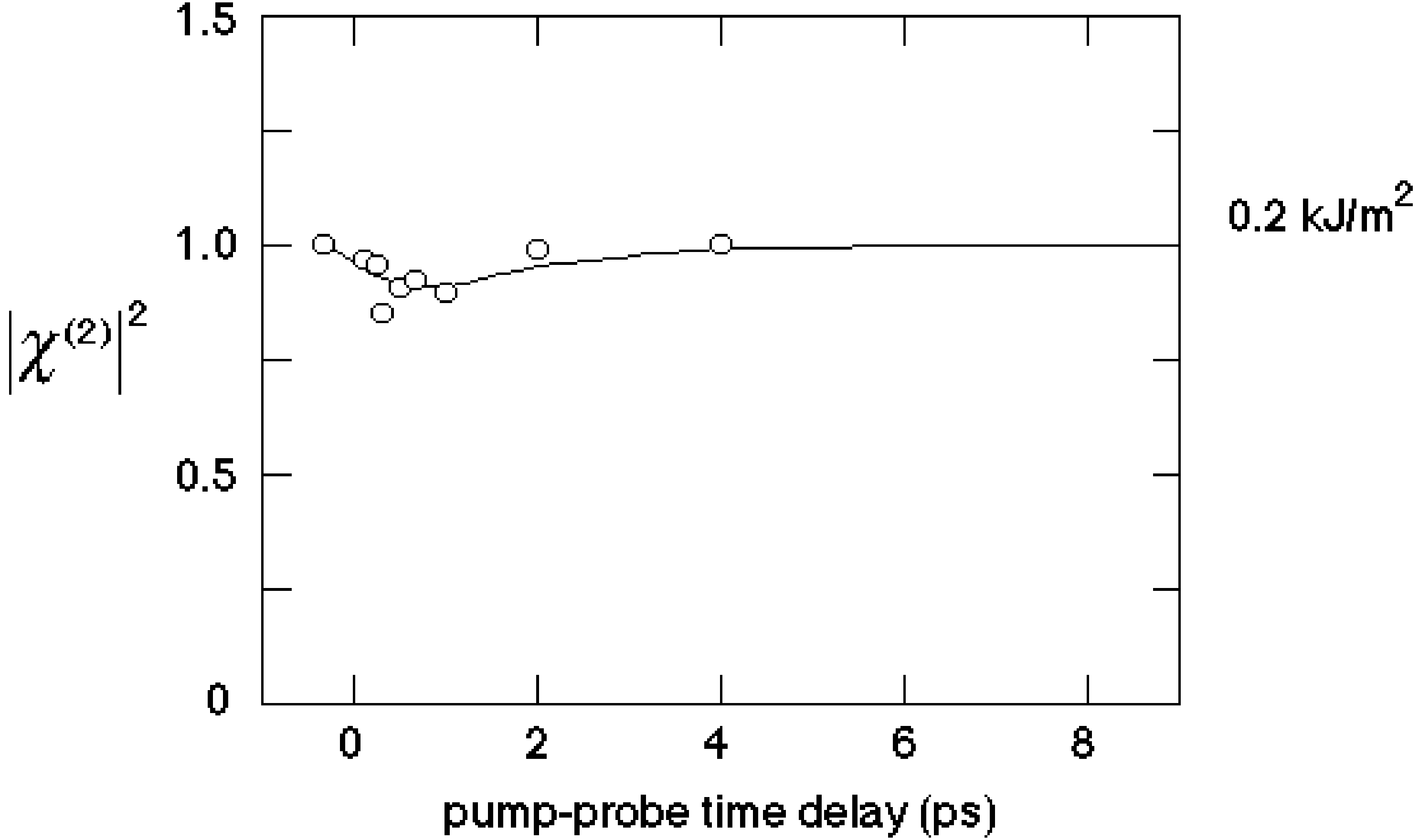

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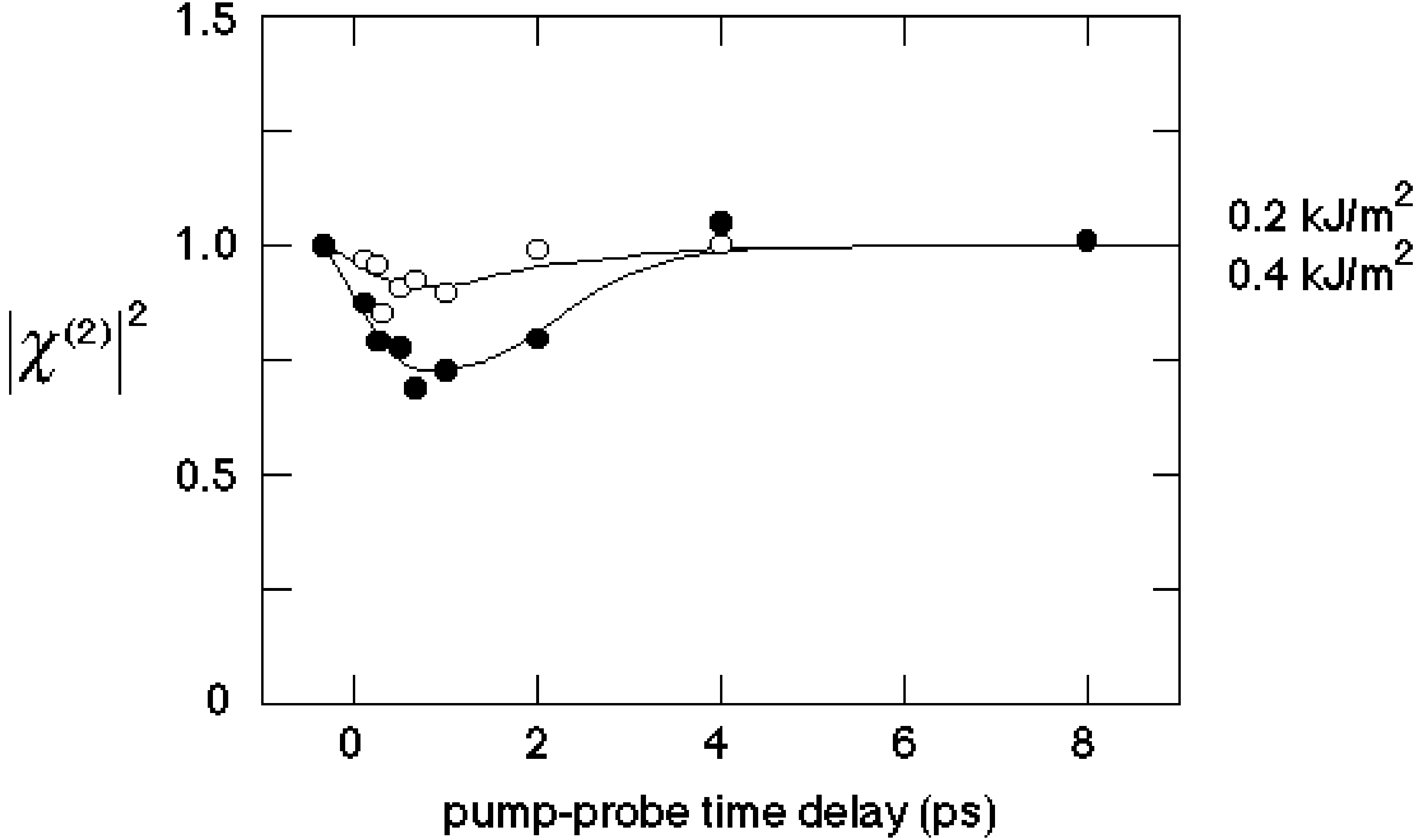

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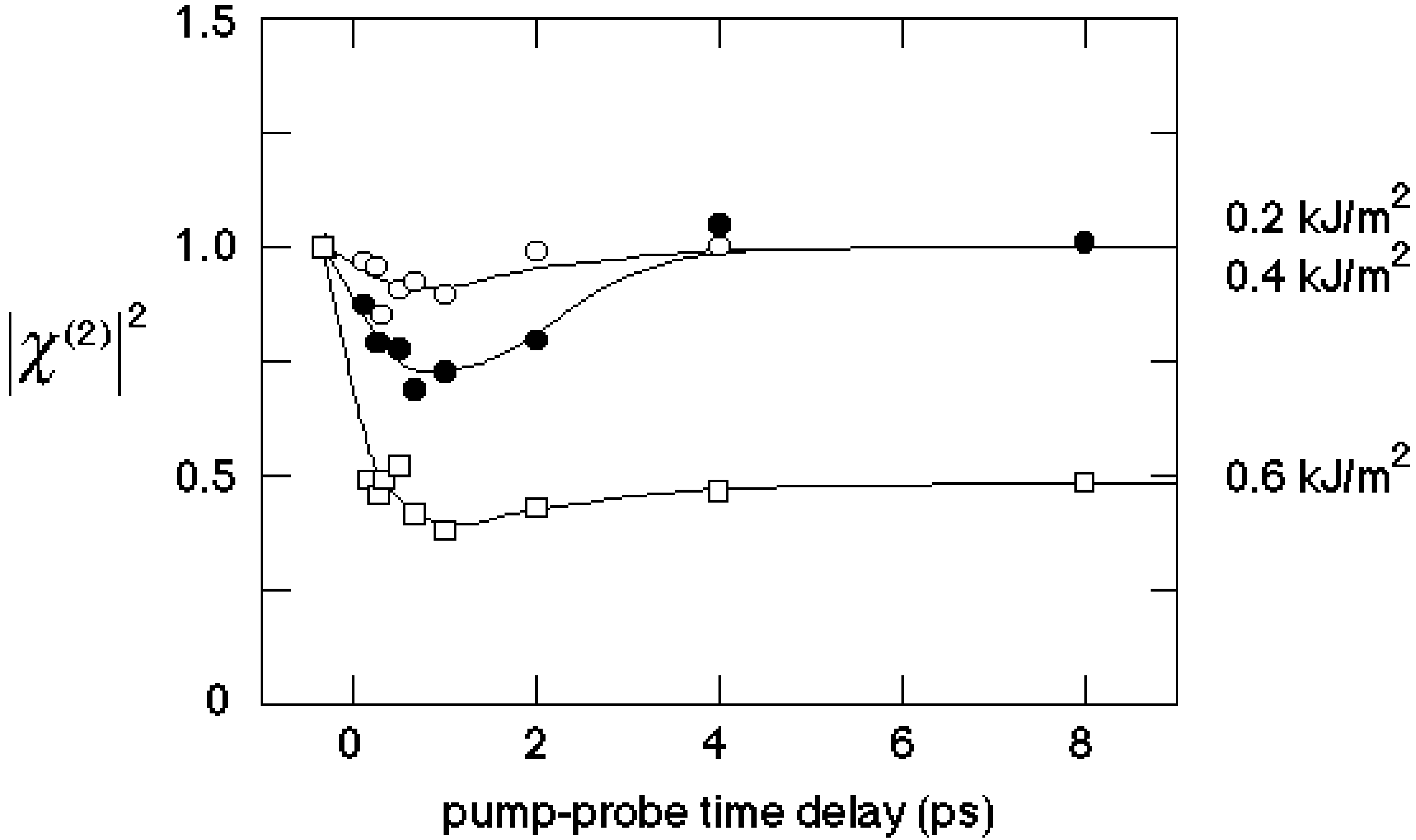

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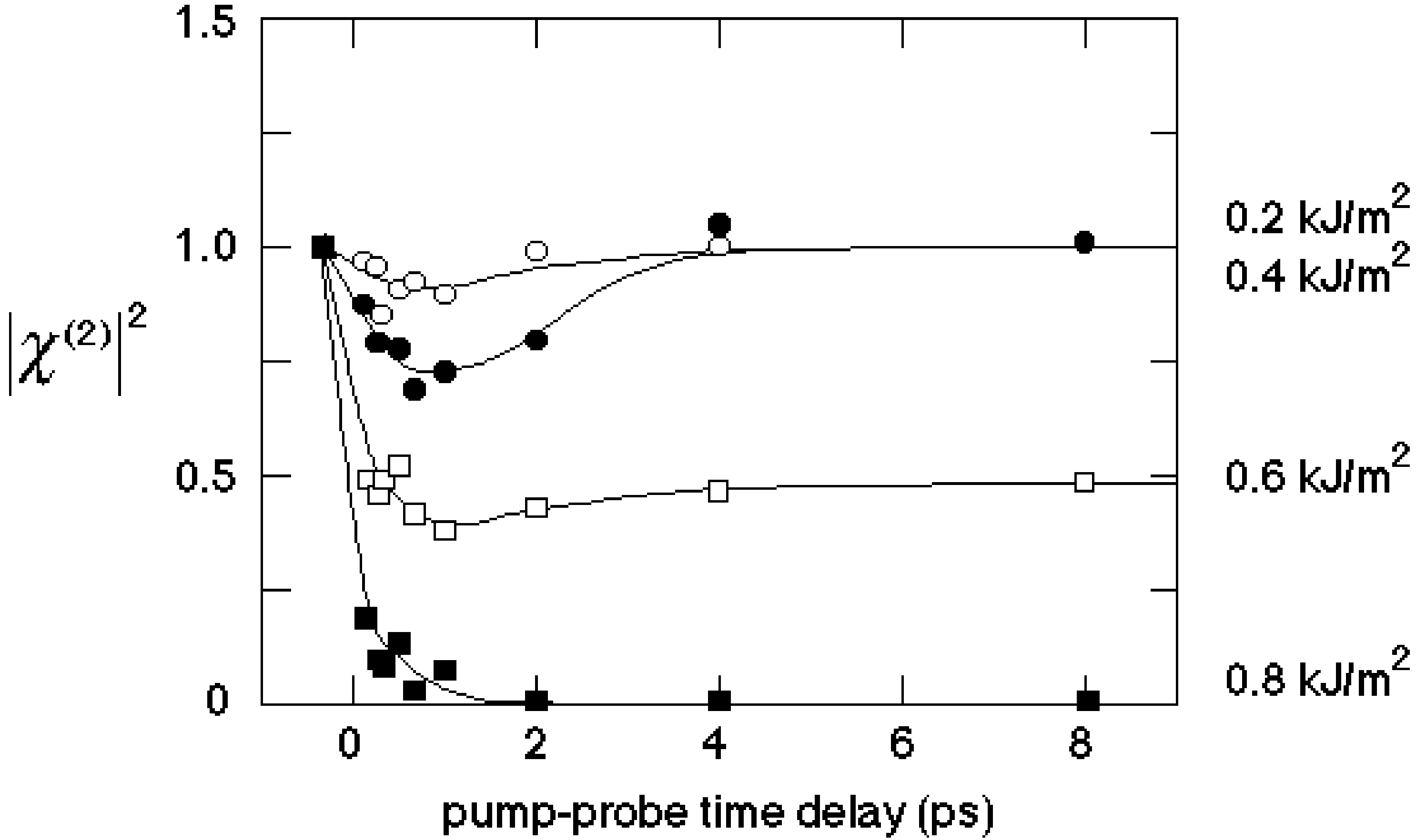

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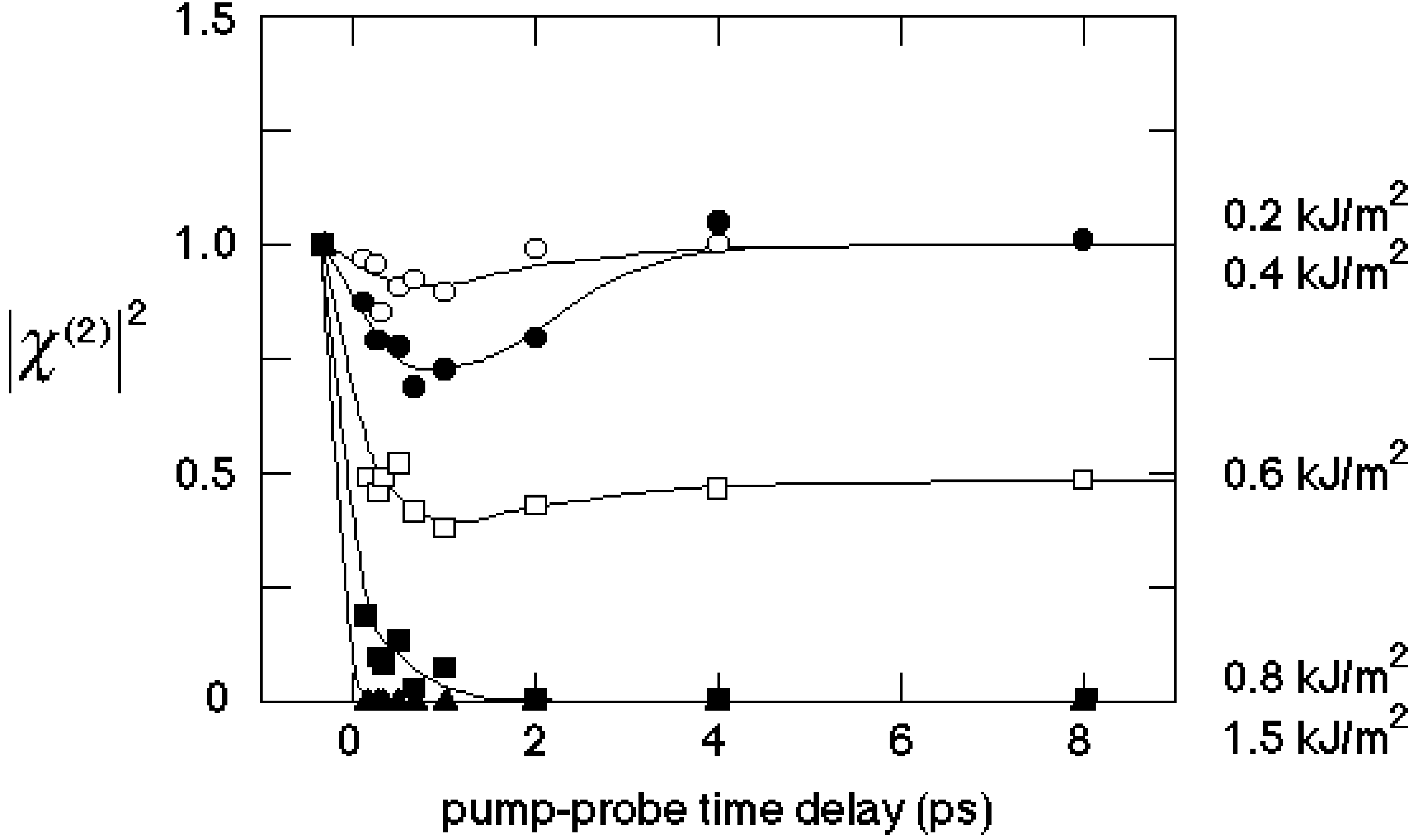

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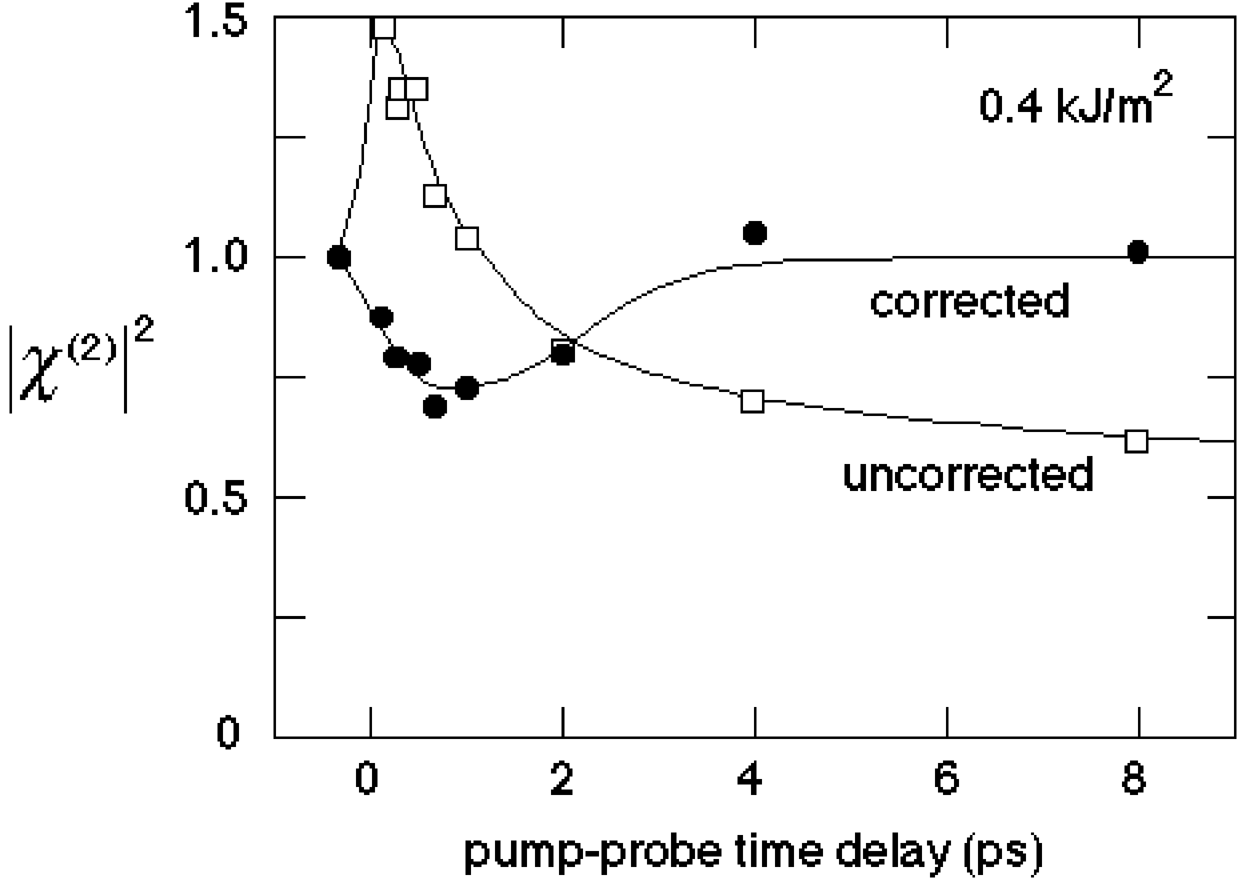
EFFECT OF CHANGES IN ε



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)

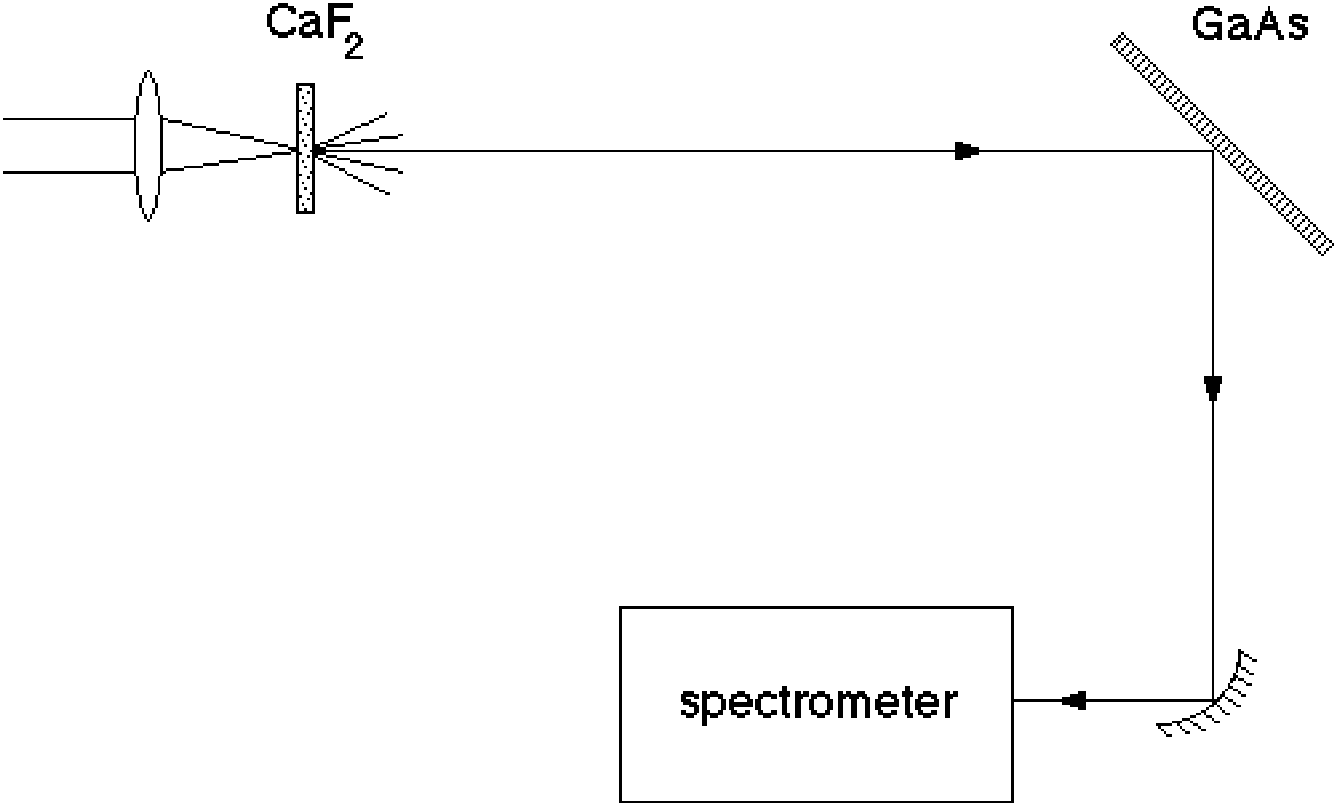
OUTLINE

❶ Background

❷ Femtosecond work

❸ Future

BROADBAND PROBE



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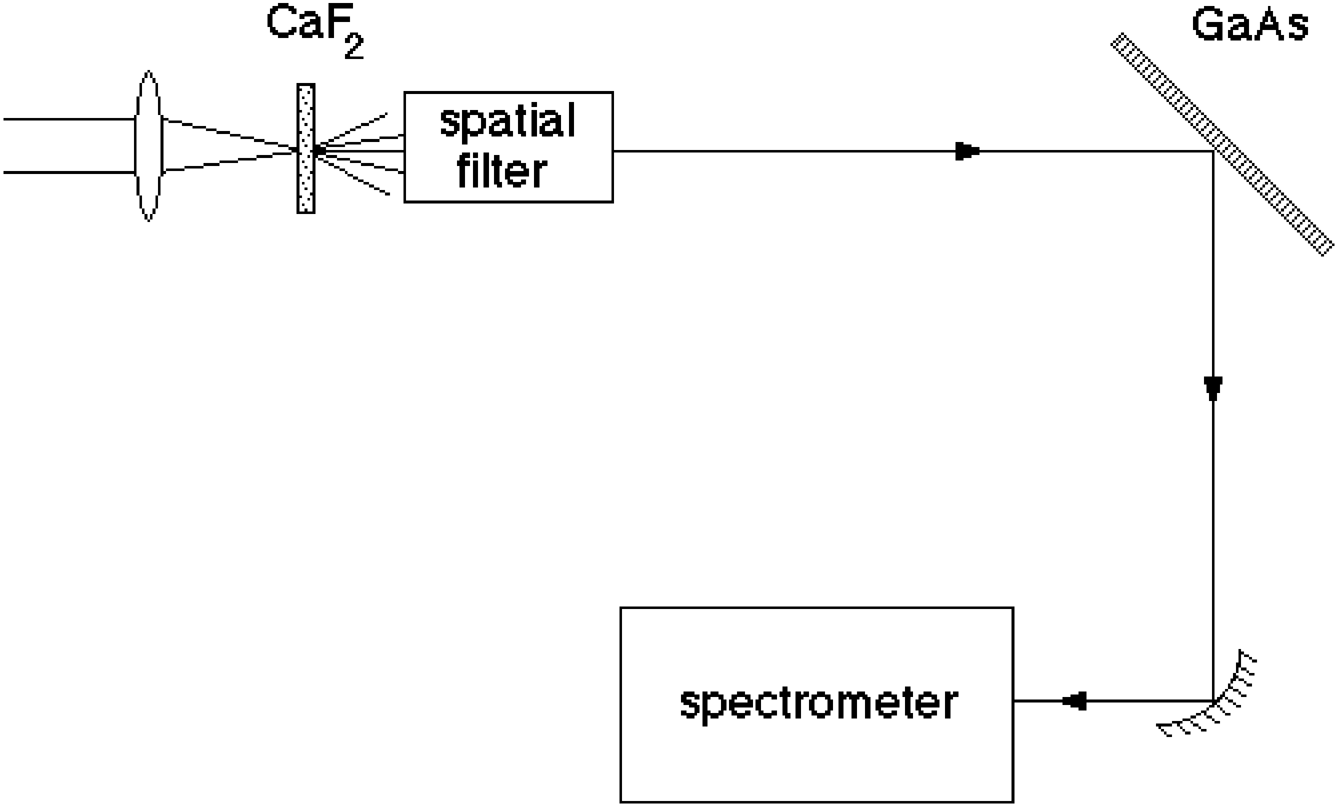

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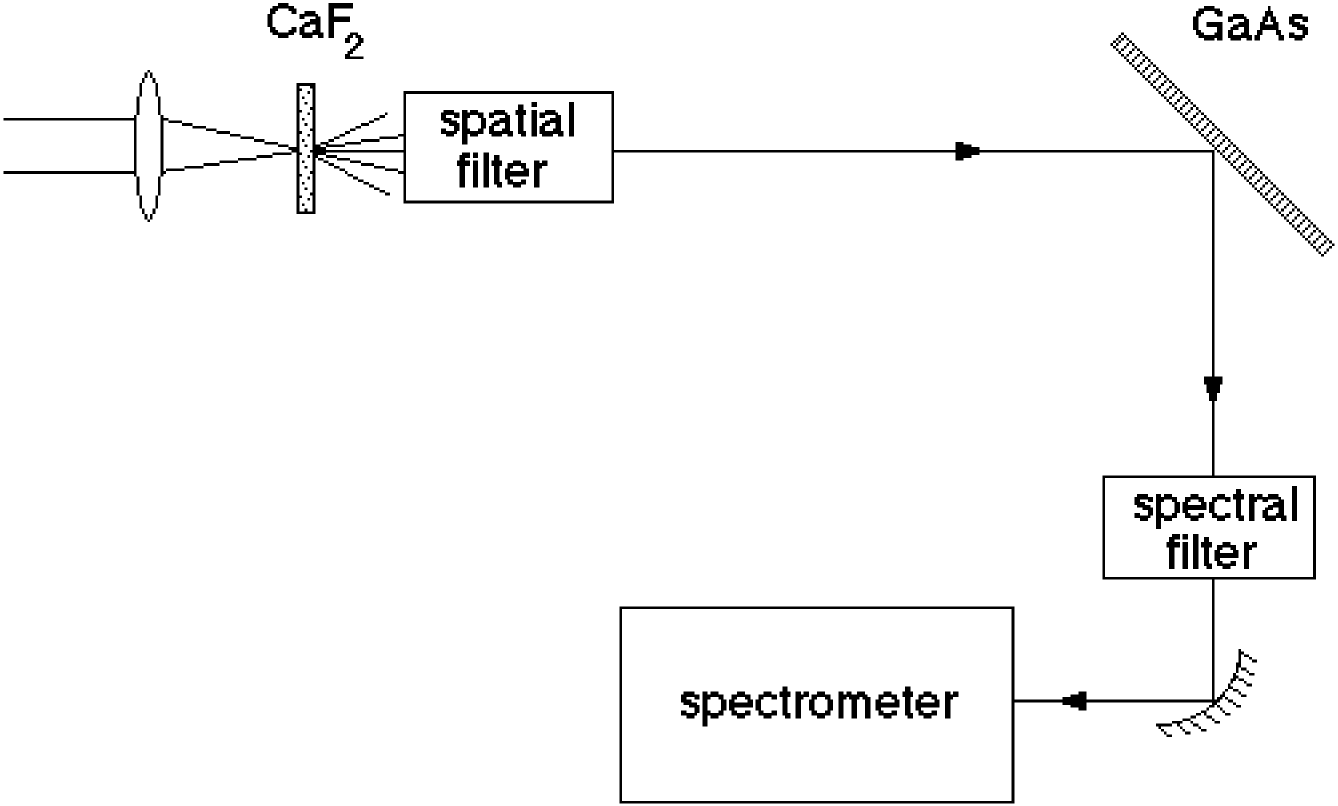

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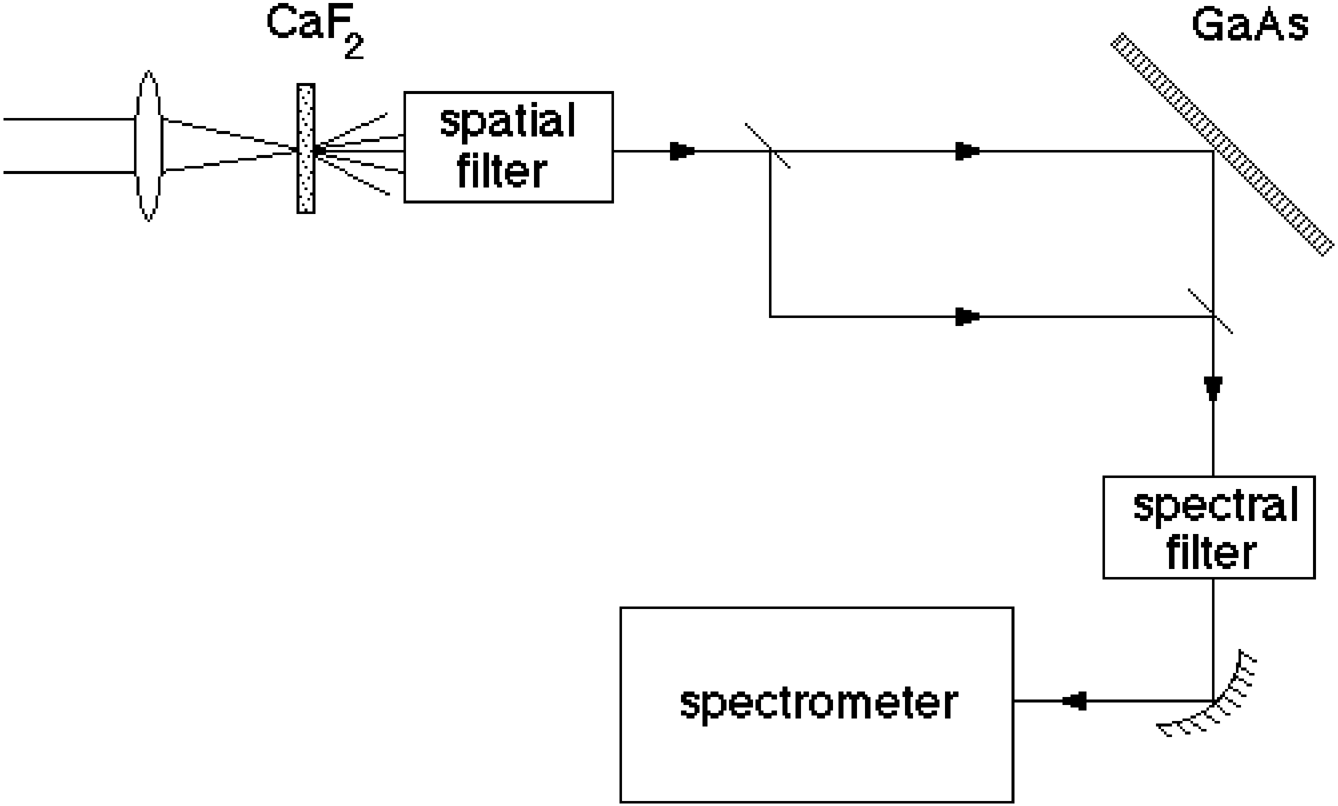
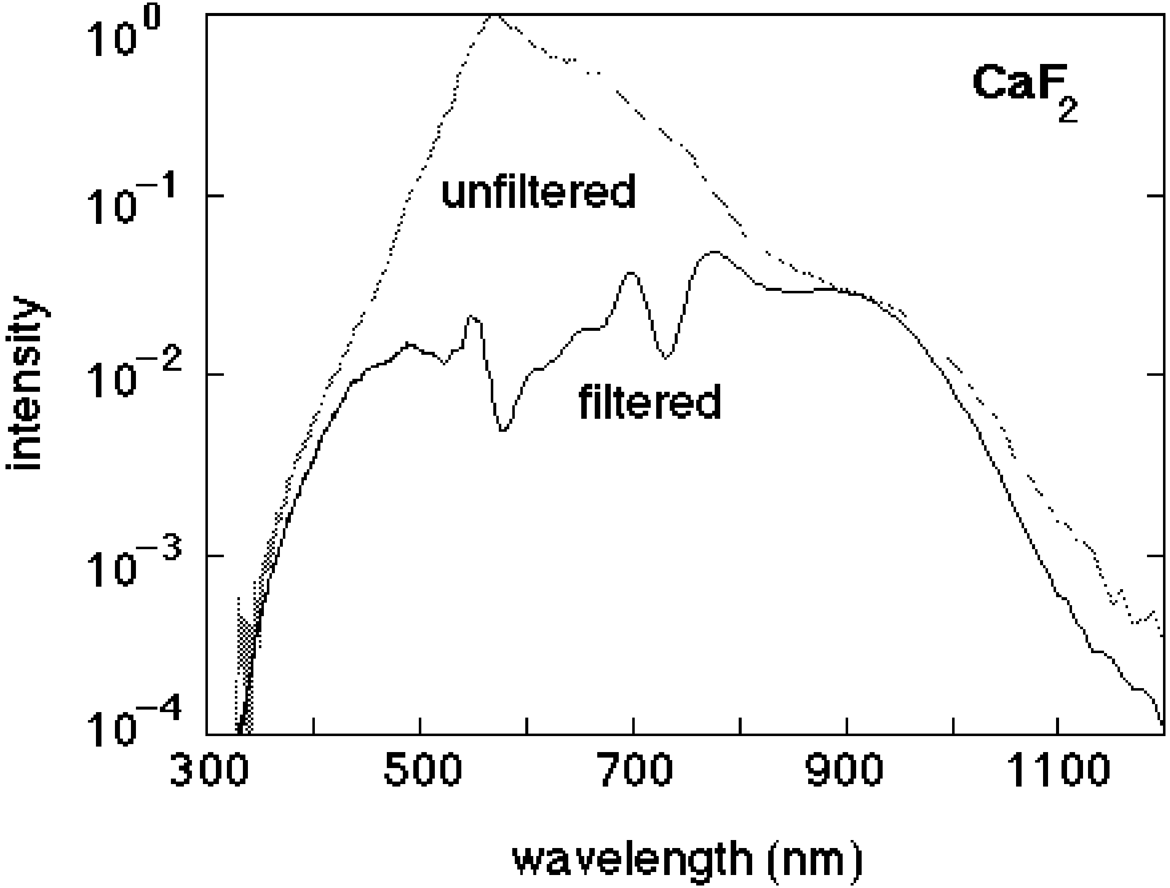


WHITE LIGHT SPECTRUM



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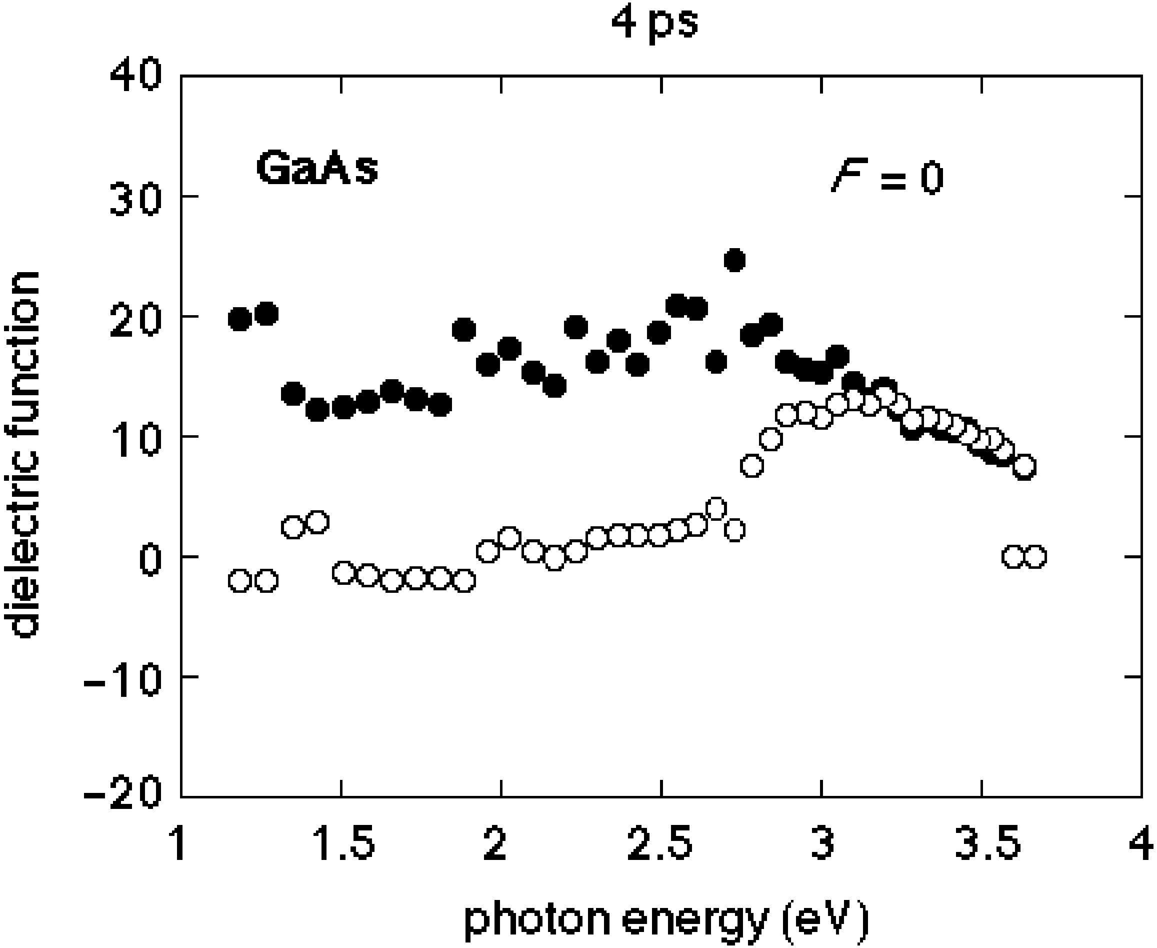

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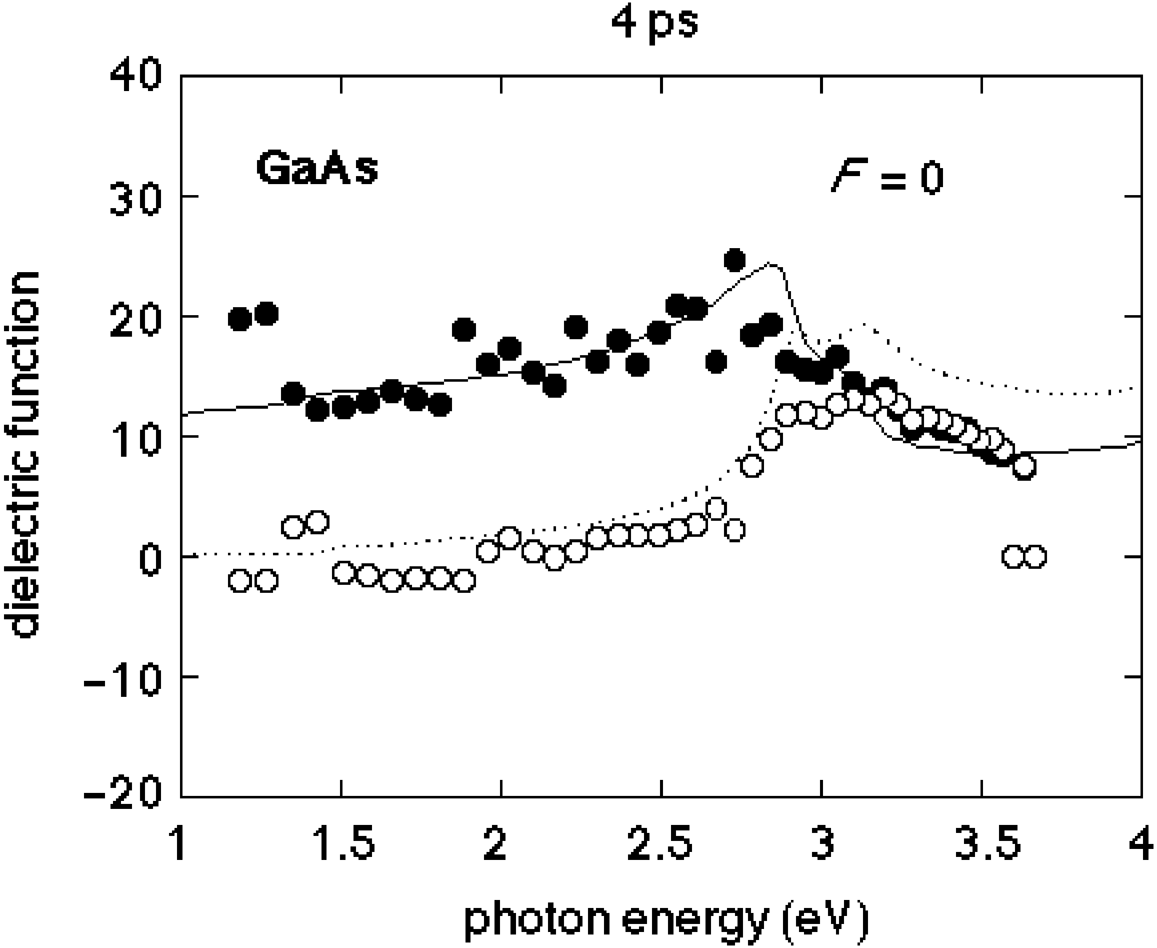

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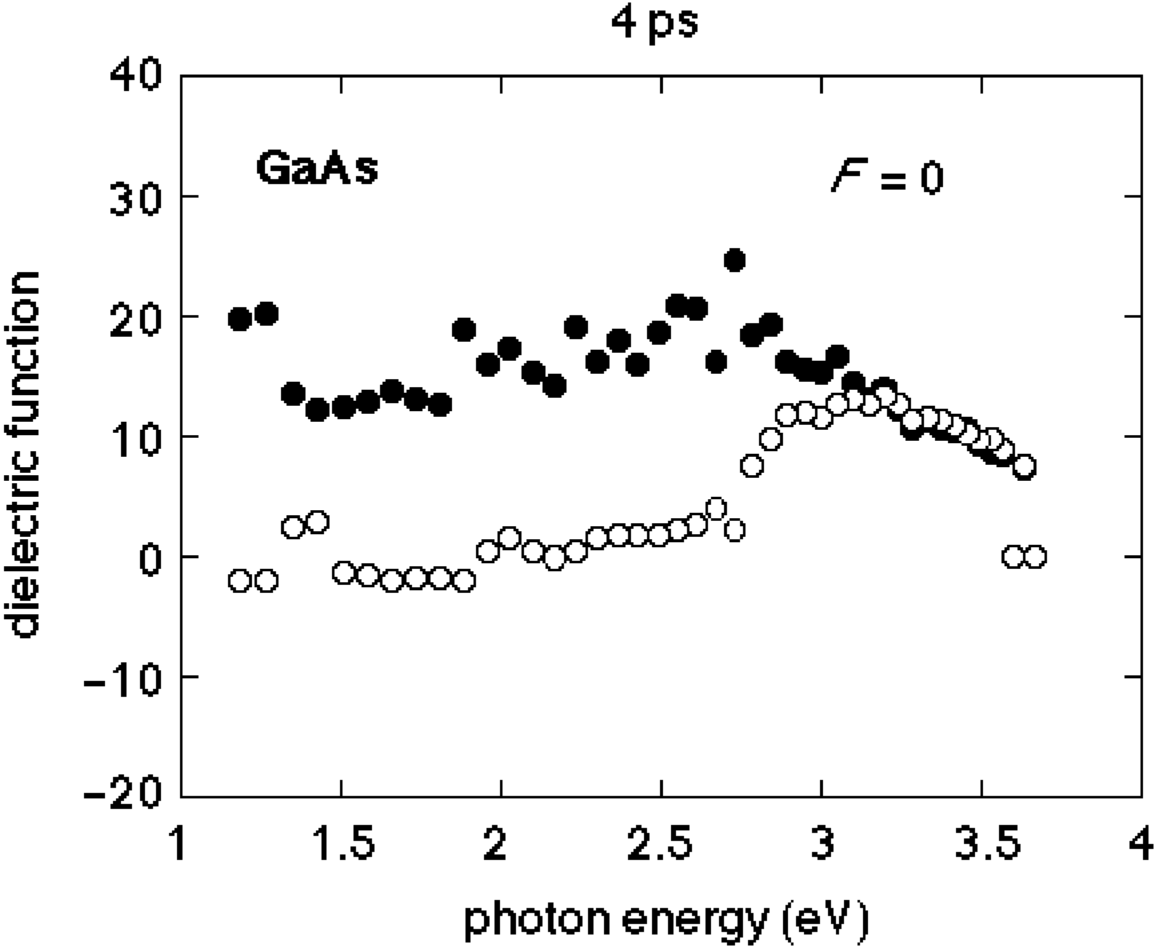

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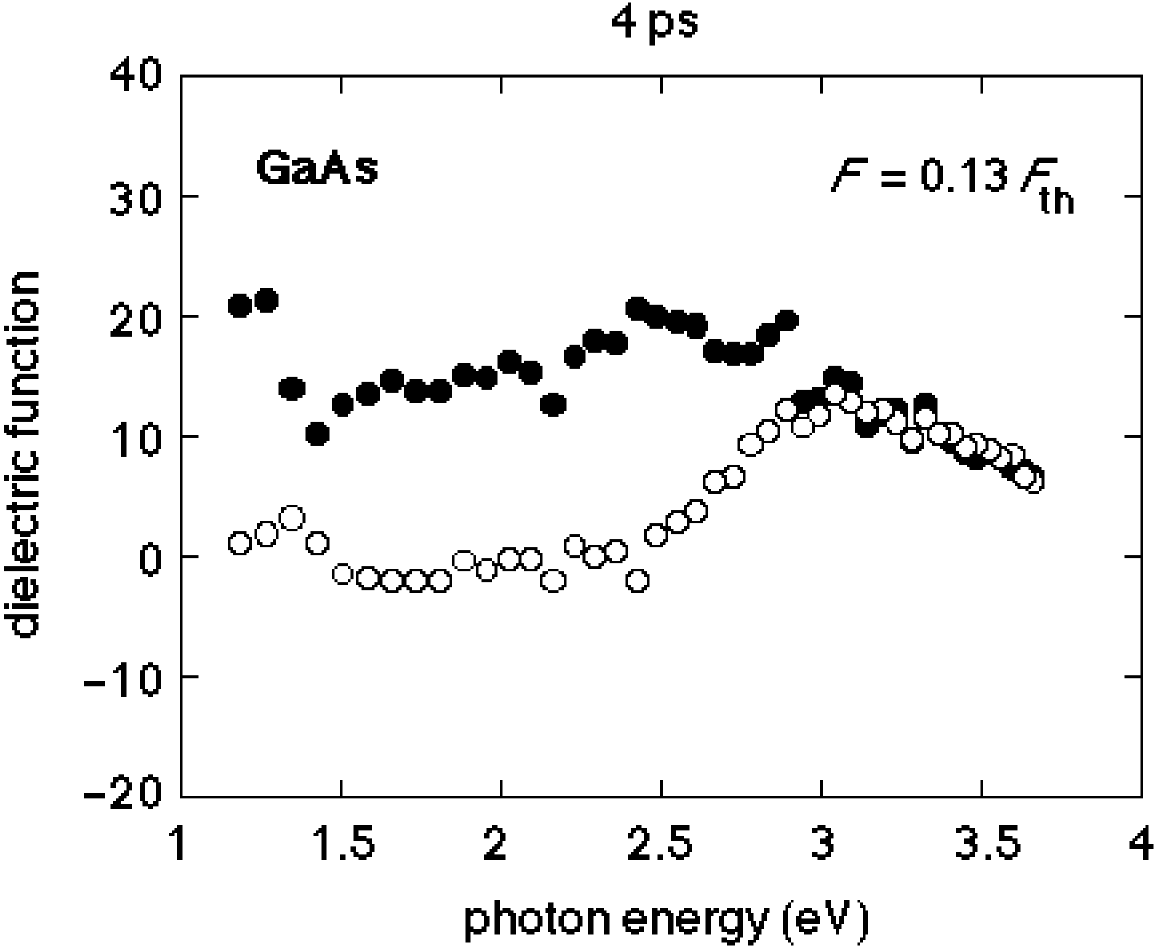

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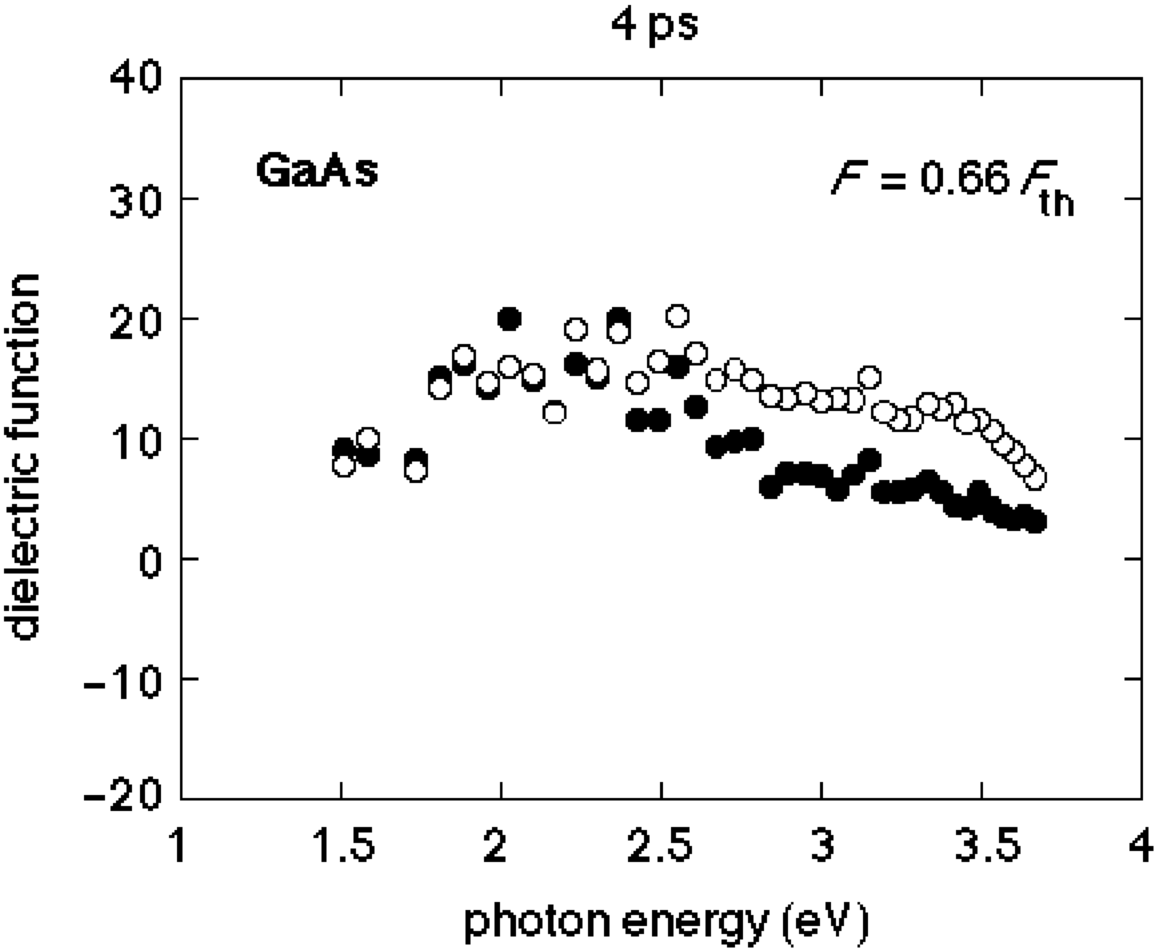

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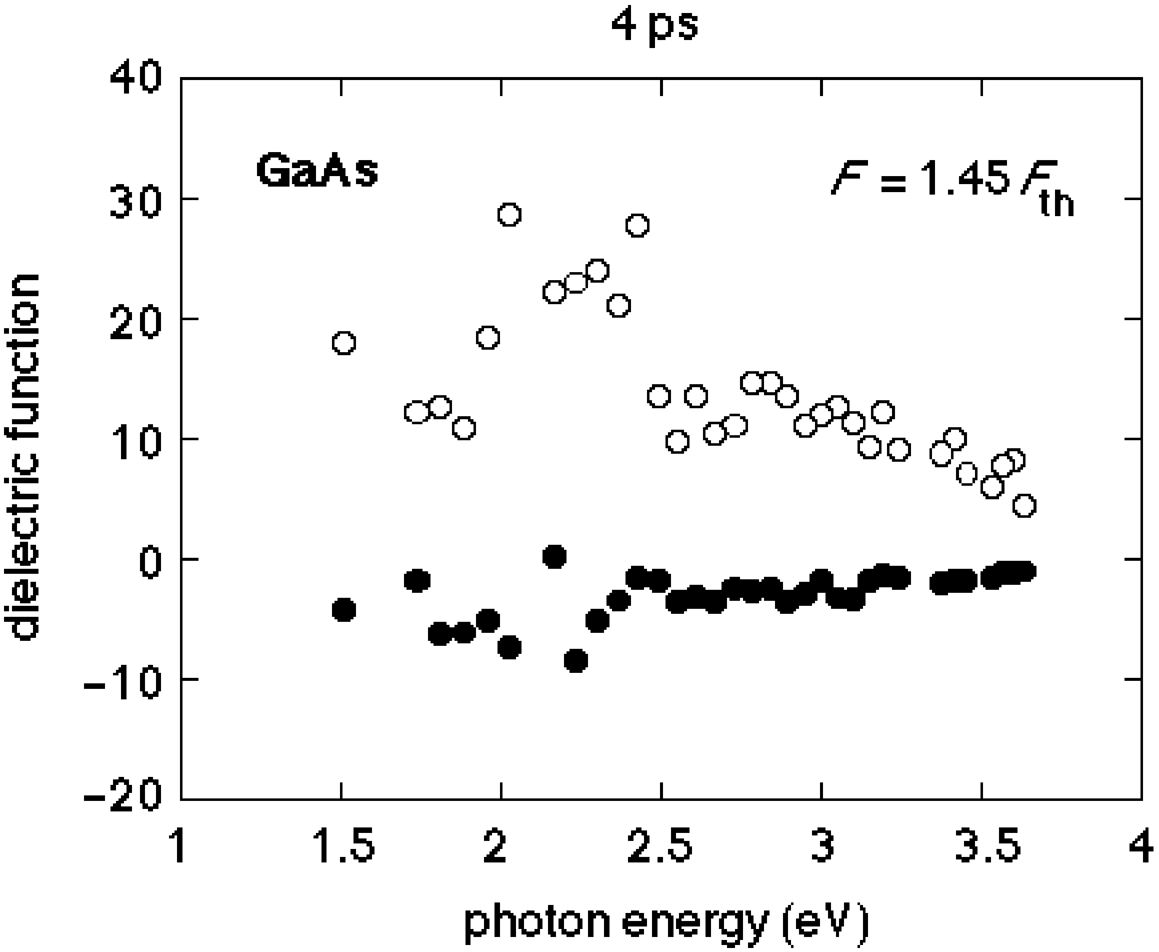

CONCLUSIONS

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

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NSF DMR 89-20490

http://mazur-www.harvard.edu