

PHASE TRANSITIONS INDUCED BY FEMTOSECOND LASER PULSES

Eric Mazur

Eli Glezer

Li Huang

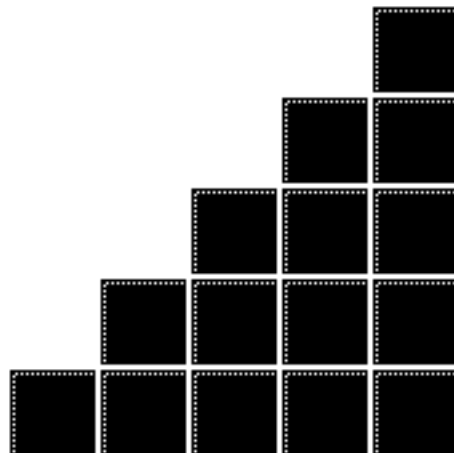
Paul Callan

Harvard University

CUOS Seminar

University of Michigan

19 February 1996



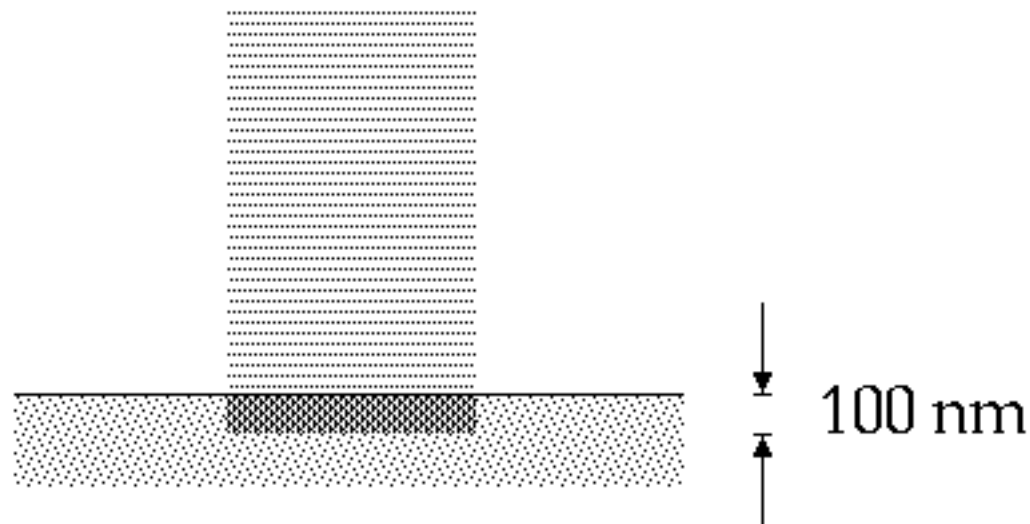
INTRODUCTION

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



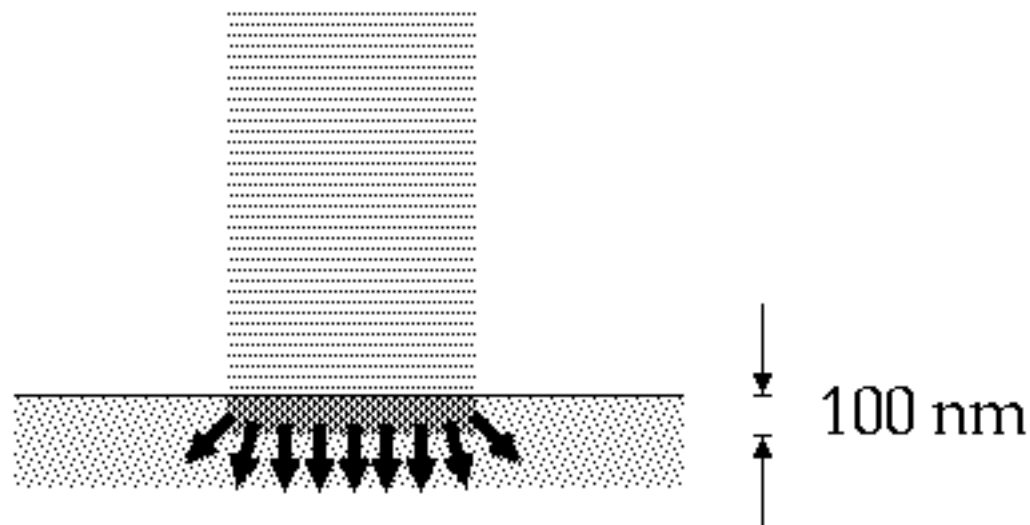
INTRODUCTION

laser deposits energy near surface...



INTRODUCTION

energy rapidly diffuses into bulk...

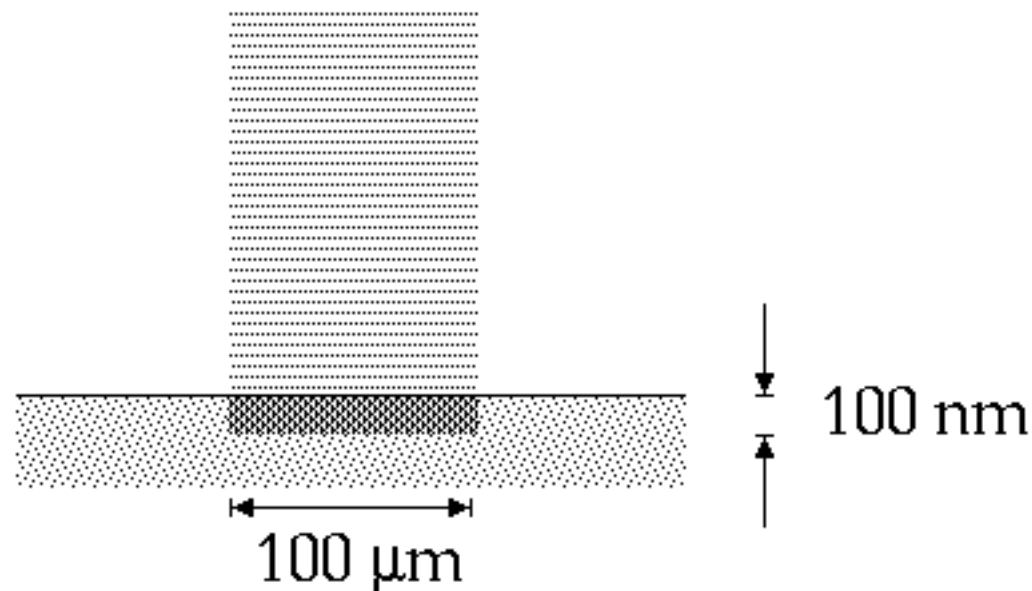


in 1 ns, diffusion length \approx 100 nm



INTRODUCTION

For $t < 1$ ns can ignore diffusion



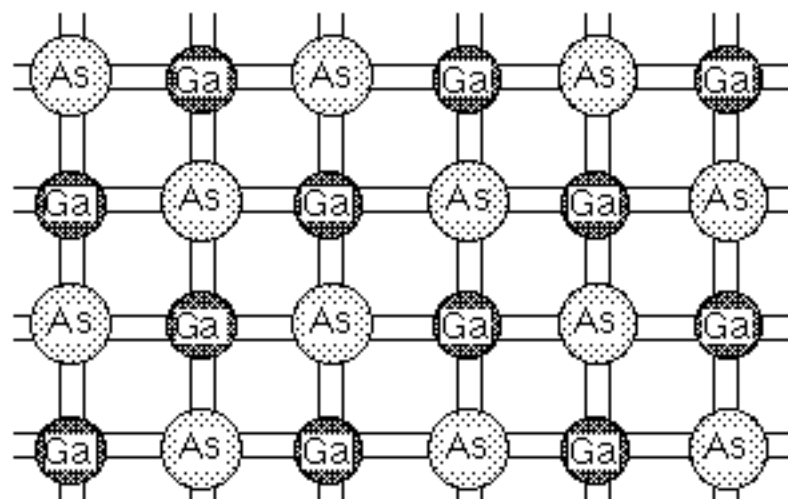
$10\ \mu\text{J}$ into a volume of about $10^{-15}\ \text{m}^3$!



- ① Background
- ② Femtosecond work
- ③ Future



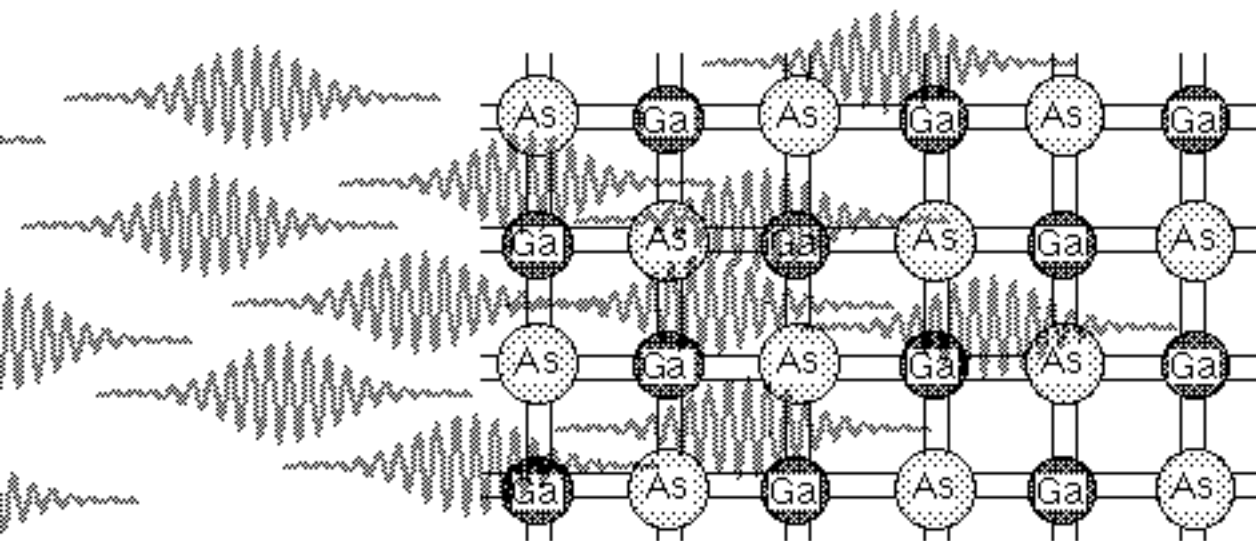
EARLY MODELS



how does light melt a solid?



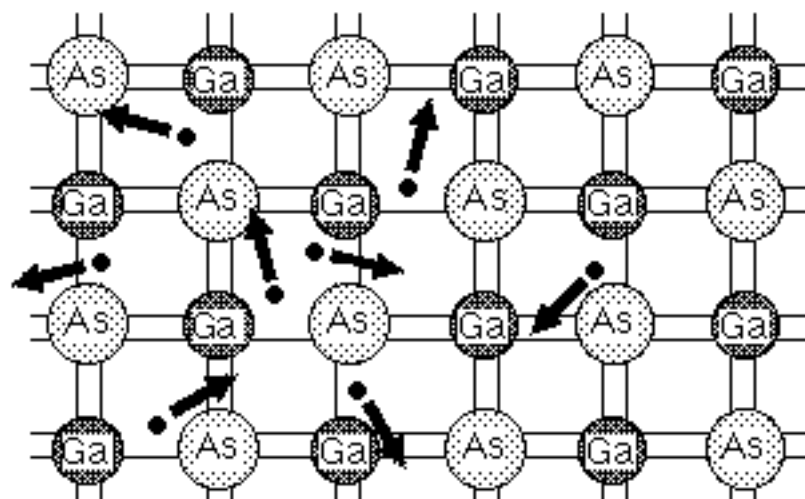
THERMAL MODEL



photons excite valence electrons...



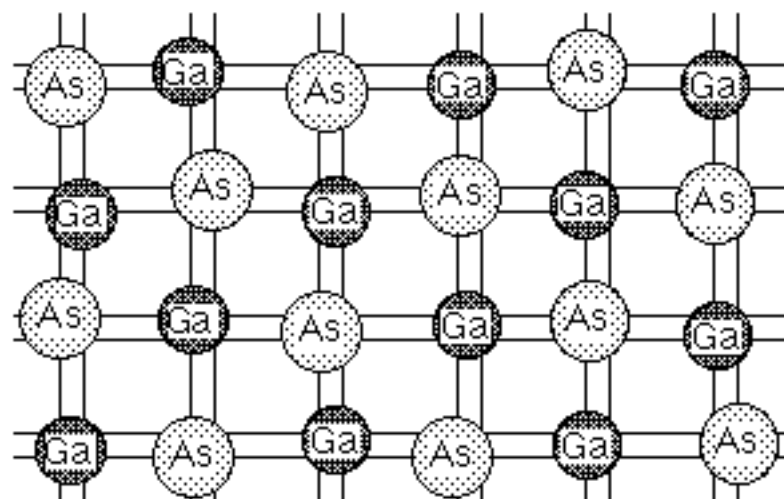
THERMAL MODEL



...and create hot electrons...



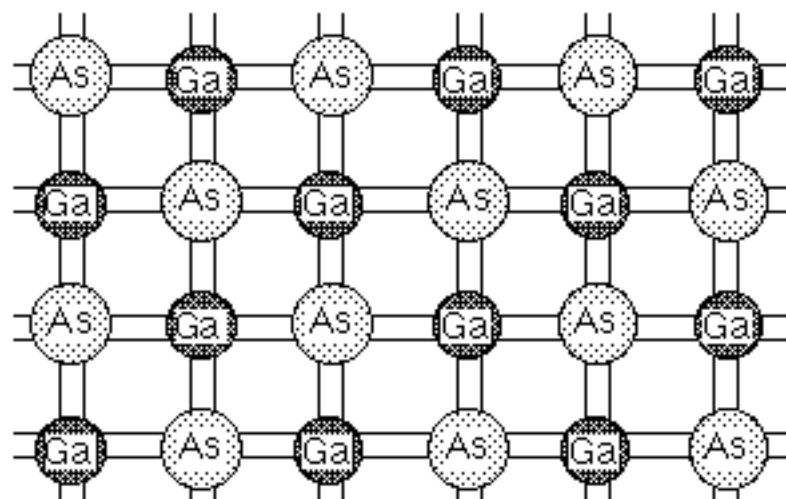
THERMAL MODEL



...which heat lattice



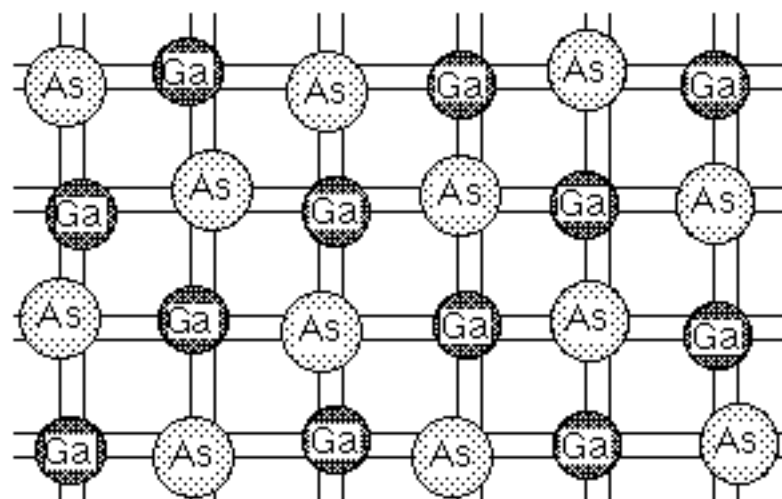
THERMAL MODEL



...which heat lattice



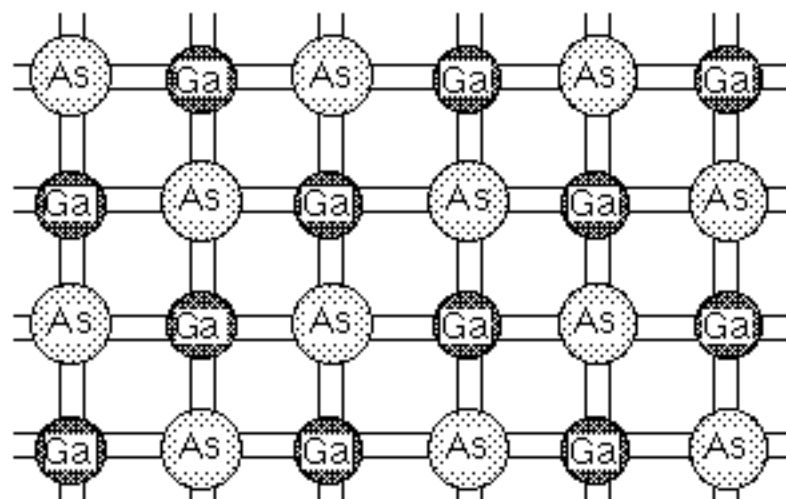
THERMAL MODEL



...which heat lattice



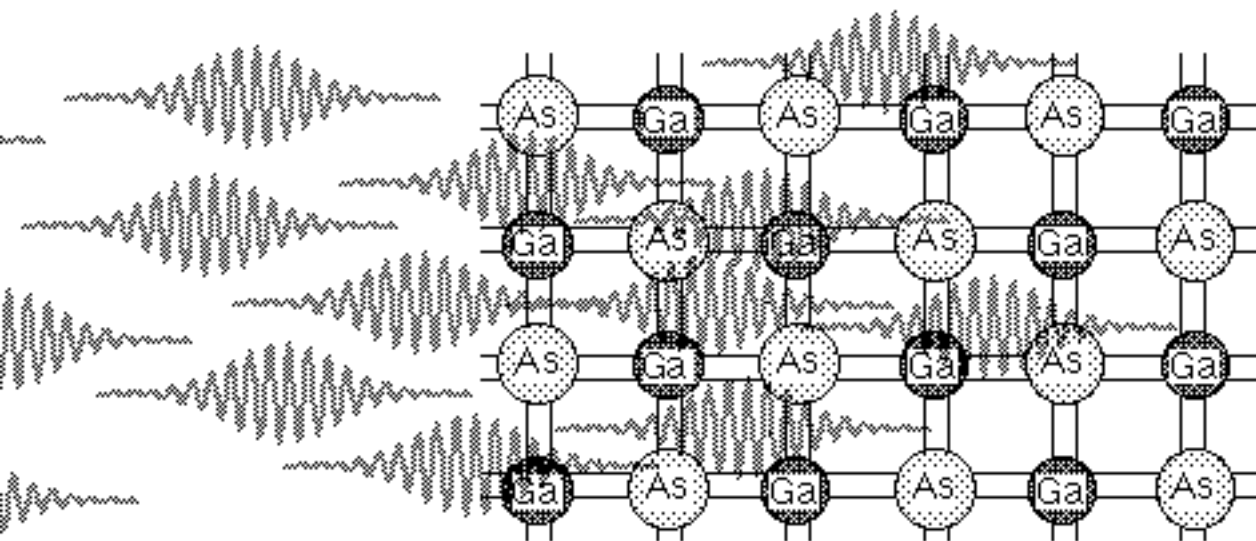
EARLY MODELS



...Or...



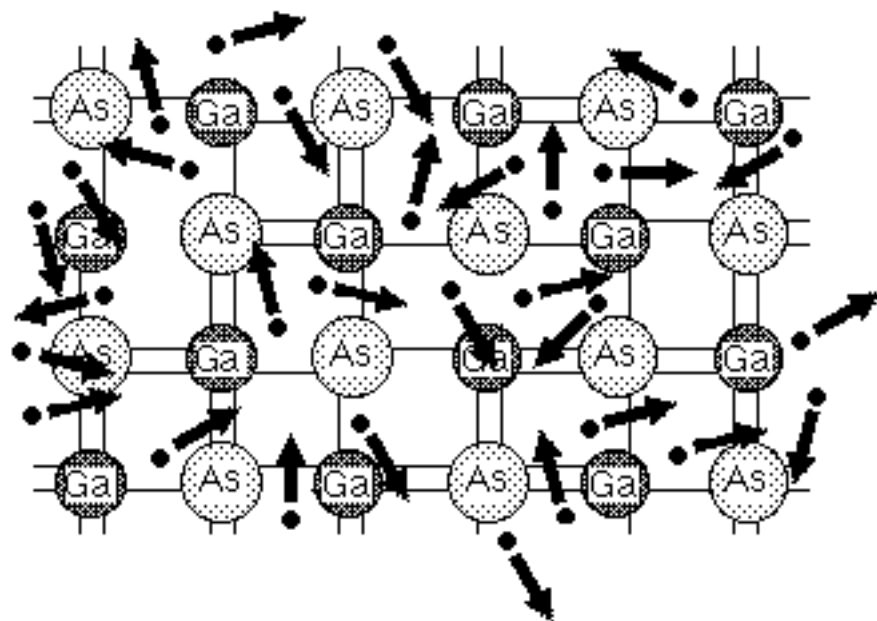
PLASMA ANNEALING



photons excite valence electrons...



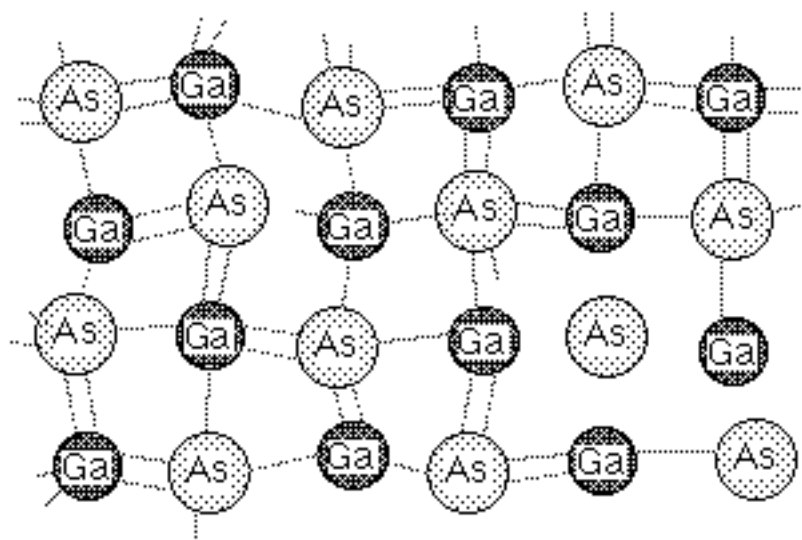
PLASMA ANNEALING



...creating a dense plasma...



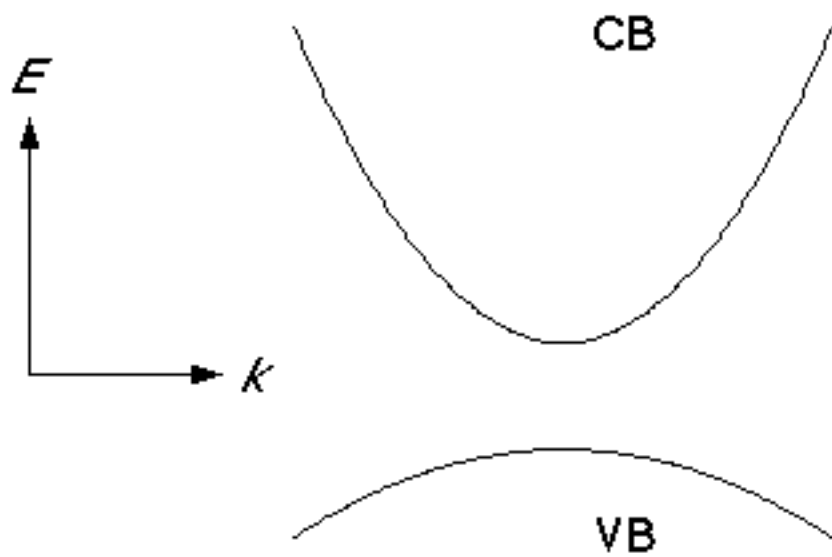
PLASMA ANNEALING



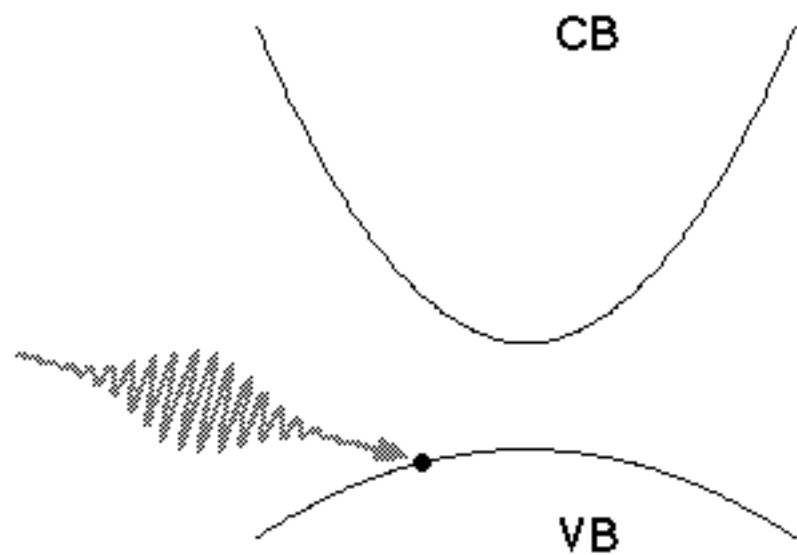
...and breaking bonds



EXCITATION



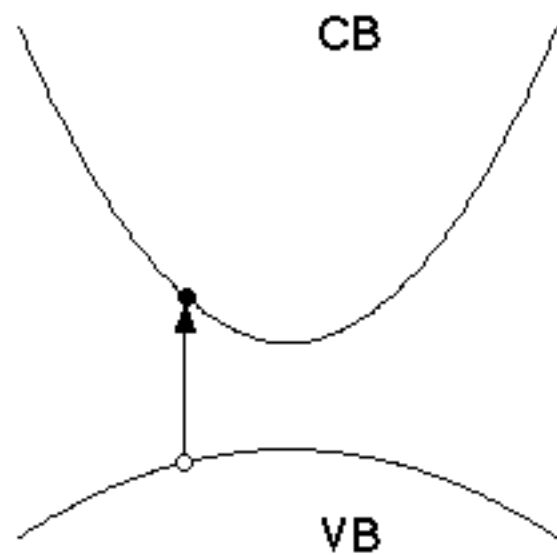
EXCITATION



photon excites valence electron



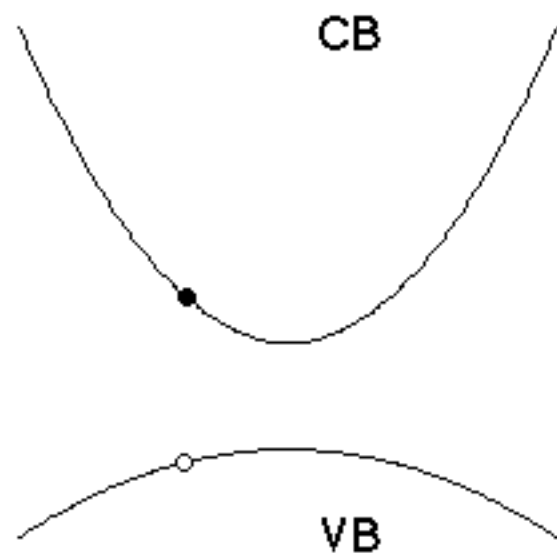
EXCITATION



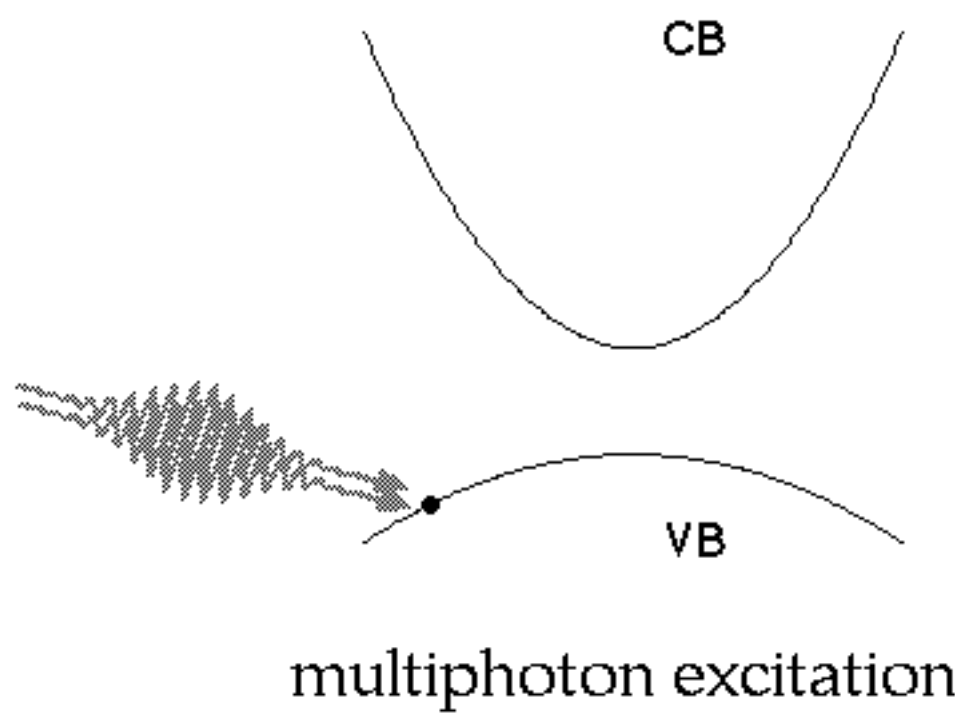
creating an electron-hole pair



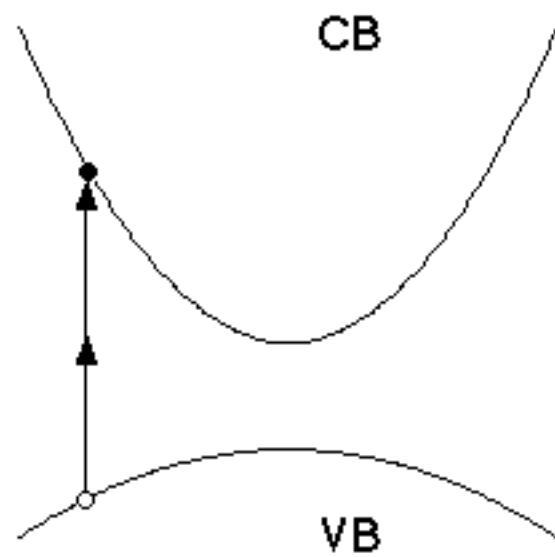
EXCITATION



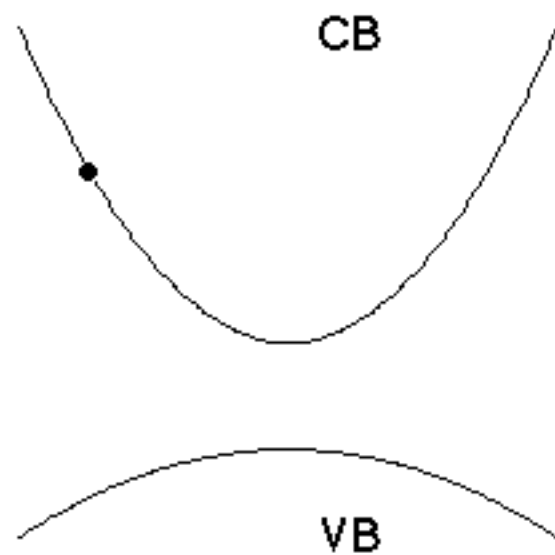
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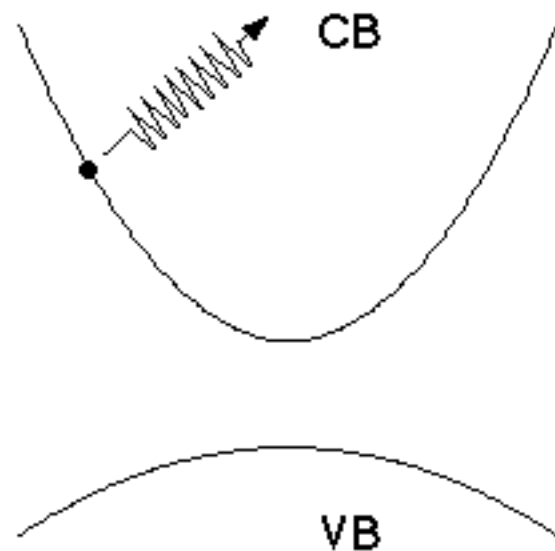
EXCITATION



EXCITATION



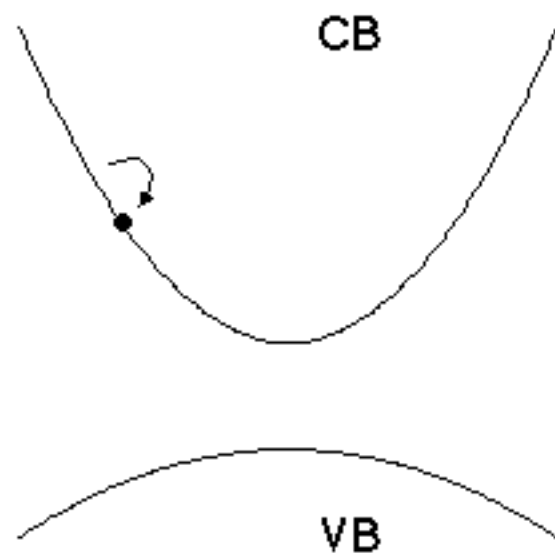
RELAXATION



emission of LO phonons



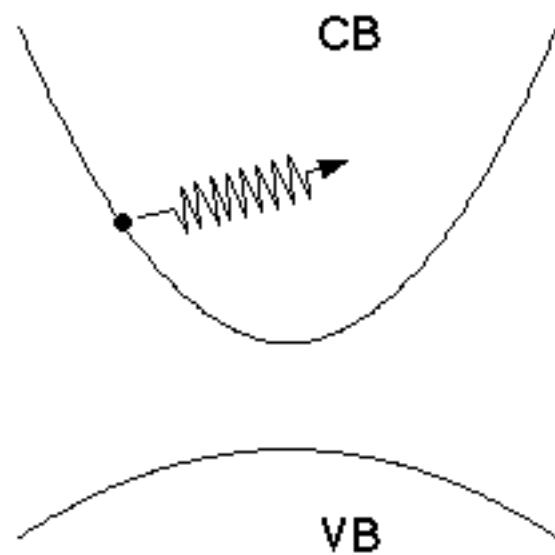
RELAXATION



emission of LO phonons



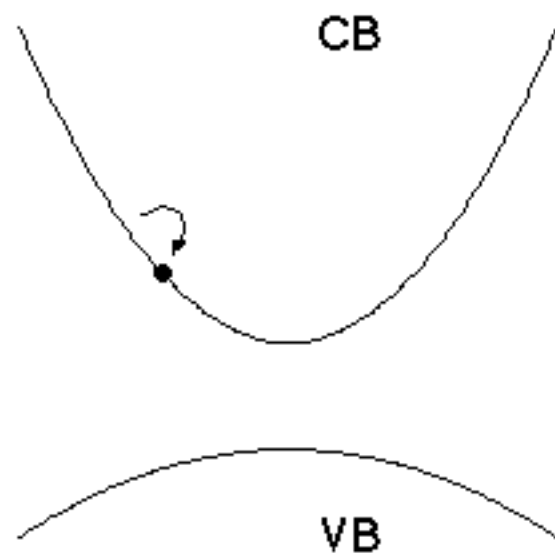
RELAXATION



emission of LO phonons



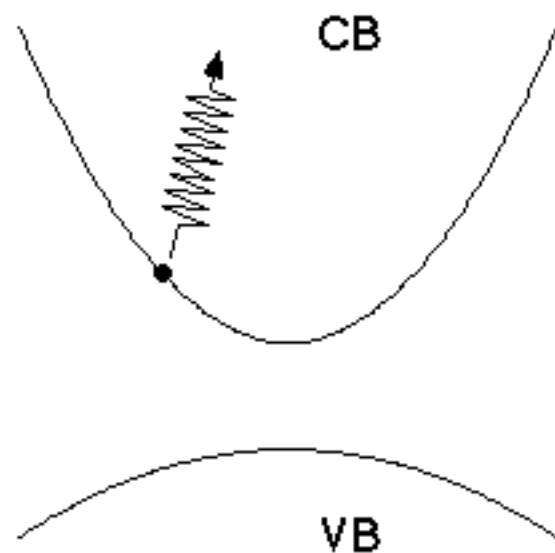
RELAXATION



emission of LO phonons



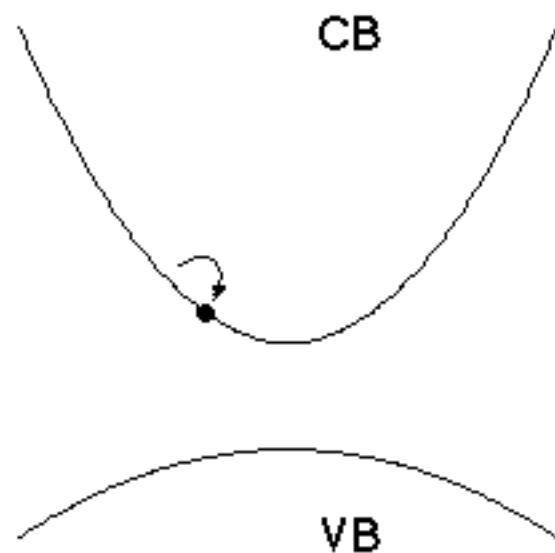
RELAXATION



emission of LO phonons



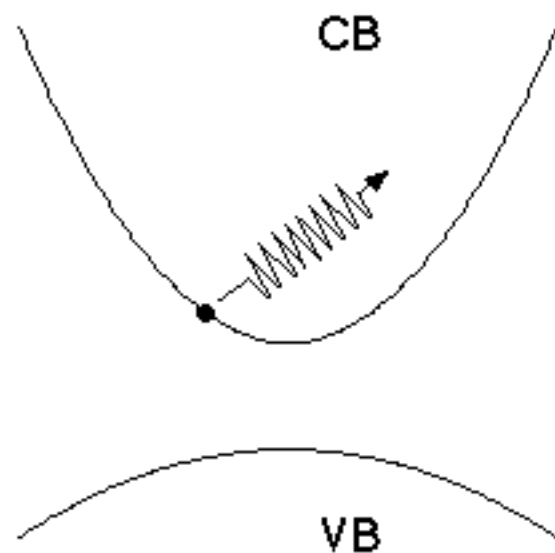
RELAXATION



emission of LO phonons



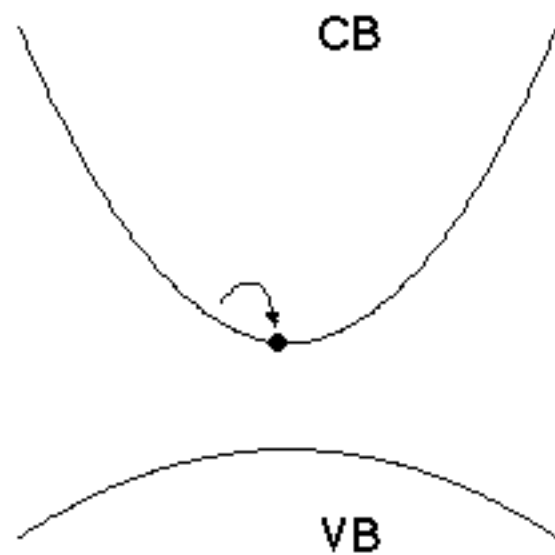
RELAXATION



emission of LO phonons



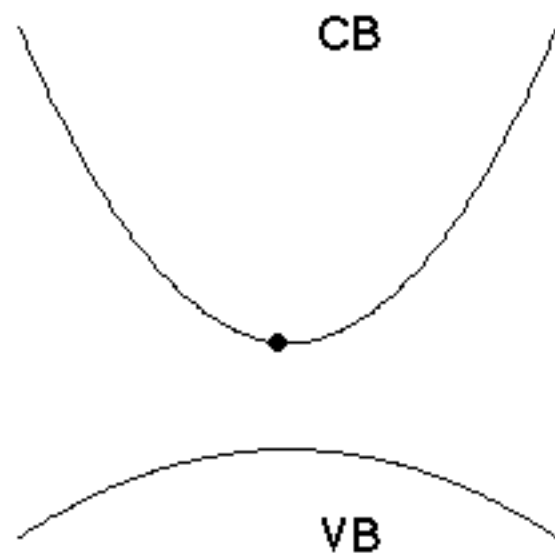
RELAXATION



emission of LO phonons

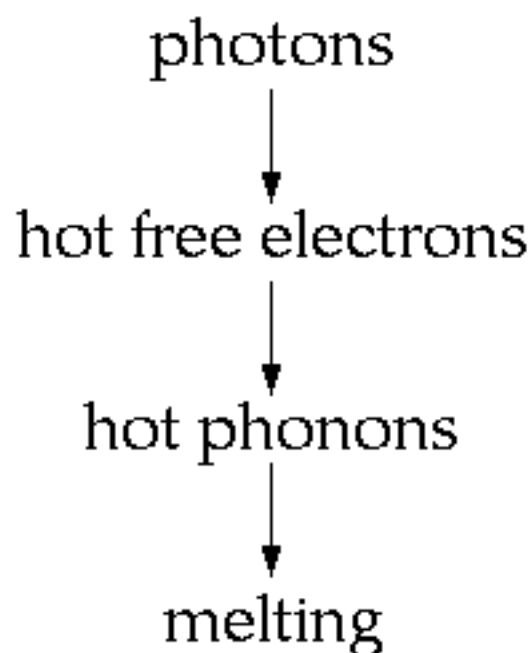


RELAXATION



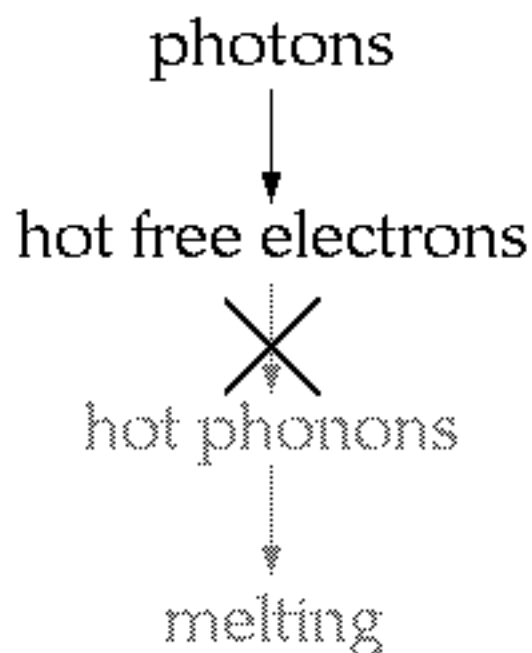
TIME SCALE

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



TIME SCALE

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



- ① Background
- ② Femtosecond work
- ③ Future



FEMTOSECOND WORK

Phonons

How can we drive them?

Beyond phonons

What happens when we break
the limit?



EXCITATION OF PHONONS

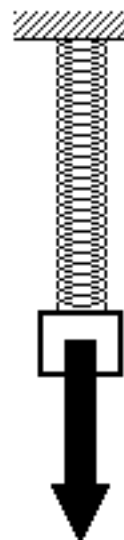
<i>absorption</i>	incoherent heating	$\tau_{e-ph} \approx 2 \text{ ps} \gg T_{ph}$
<i>SRS, CARS</i>	coherent driving	$\tau_{exc} \geq 6 \text{ ps} \gg T_{ph}$
<i>ISRS</i>	impulsive excitation	$\tau_{exc} \approx 0.1 \text{ ps} \leq T_{ph}$



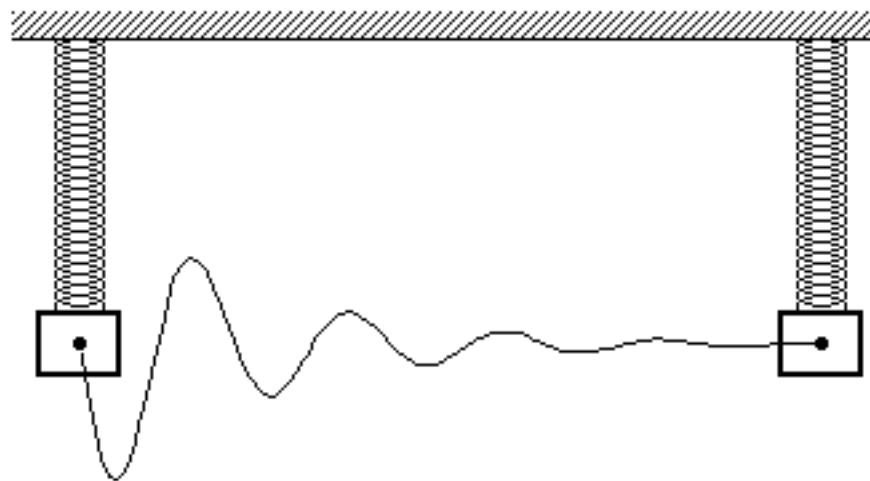
IMPULSIVE RAMAN SCATTERING



IMPULSIVE RAMAN SCATTERING



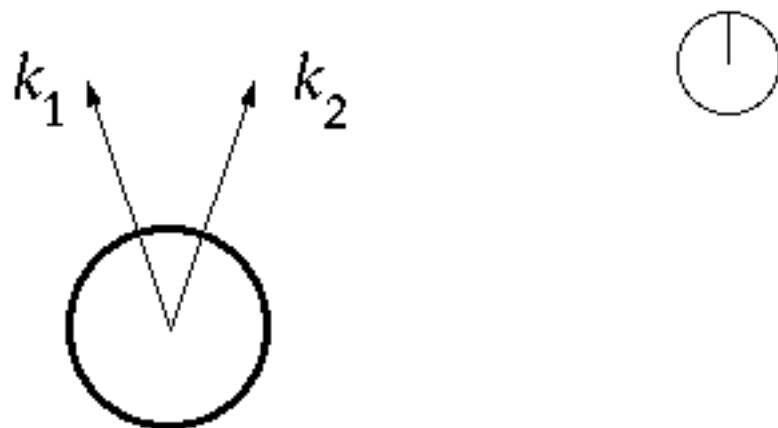
IMPULSIVE RAMAN SCATTERING



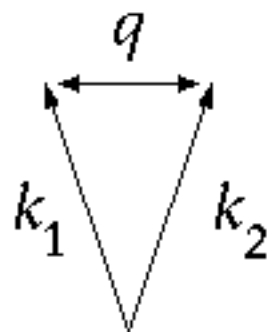
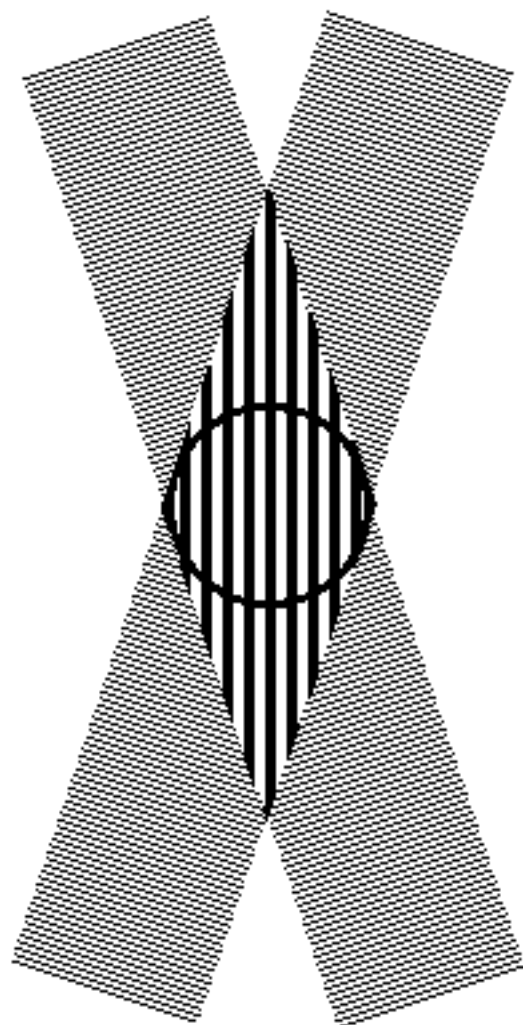
IMPULSIVE RAMAN SCATTERING



IMPULSIVE RAMAN SCATTERING



IMPULSIVE RAMAN SCATTERING



$$k_1 - k_2 = \pm q$$



IMPULSIVE RAMAN SCATTERING



create decaying standing wave...



IMPULSIVE RAMAN SCATTERING

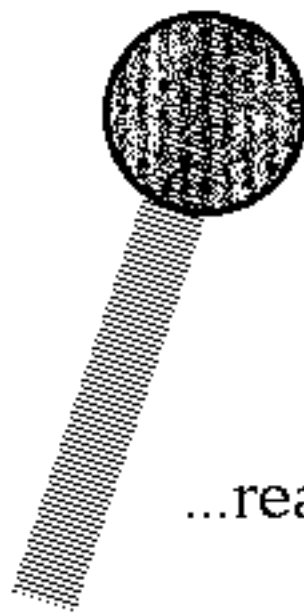


create decaying standing wave...



IMPULSIVE RAMAN SCATTERING

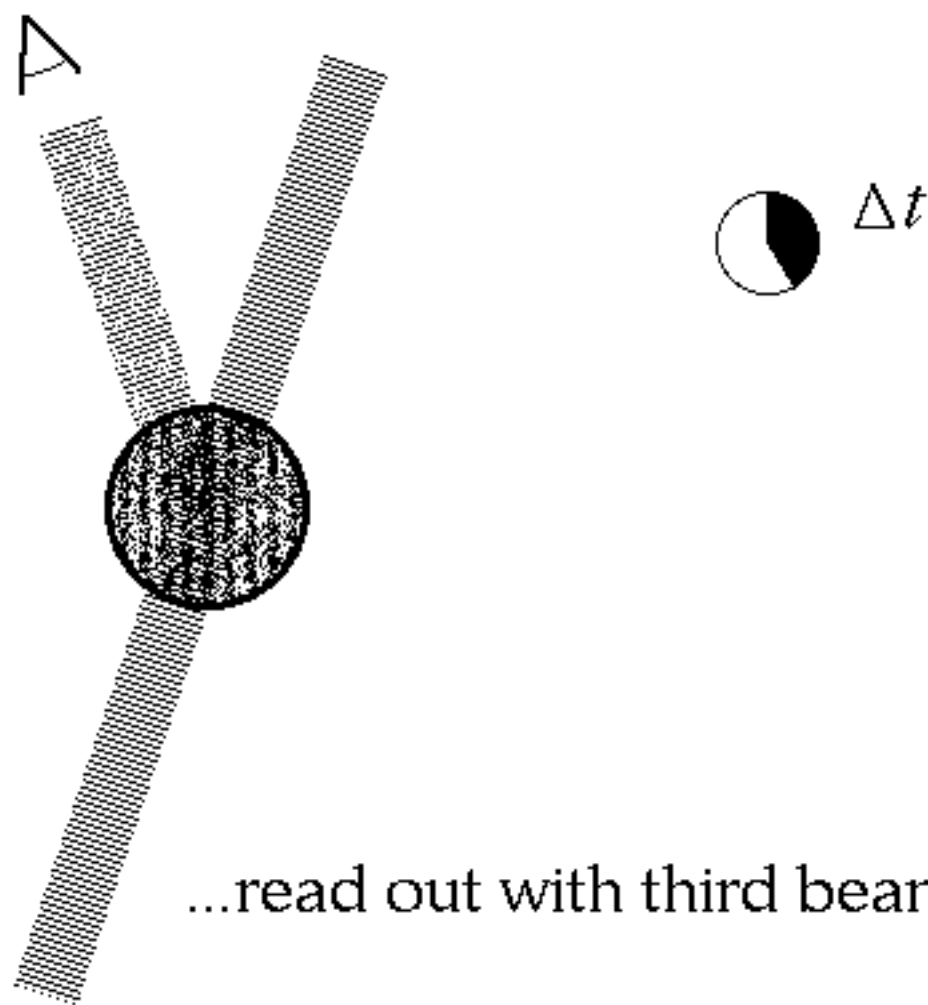
A



...read out with third beam



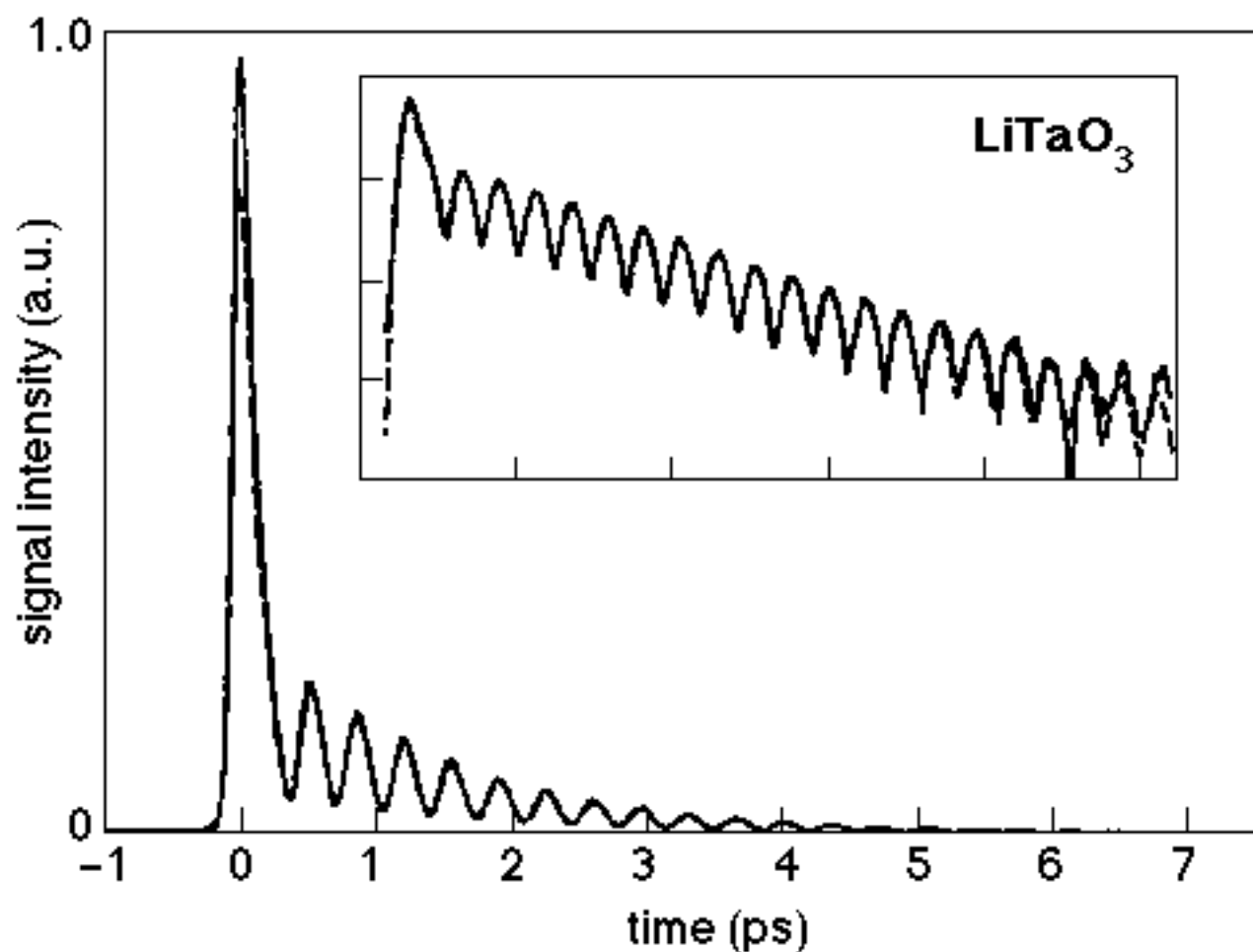
IMPULSIVE RAMAN SCATTERING



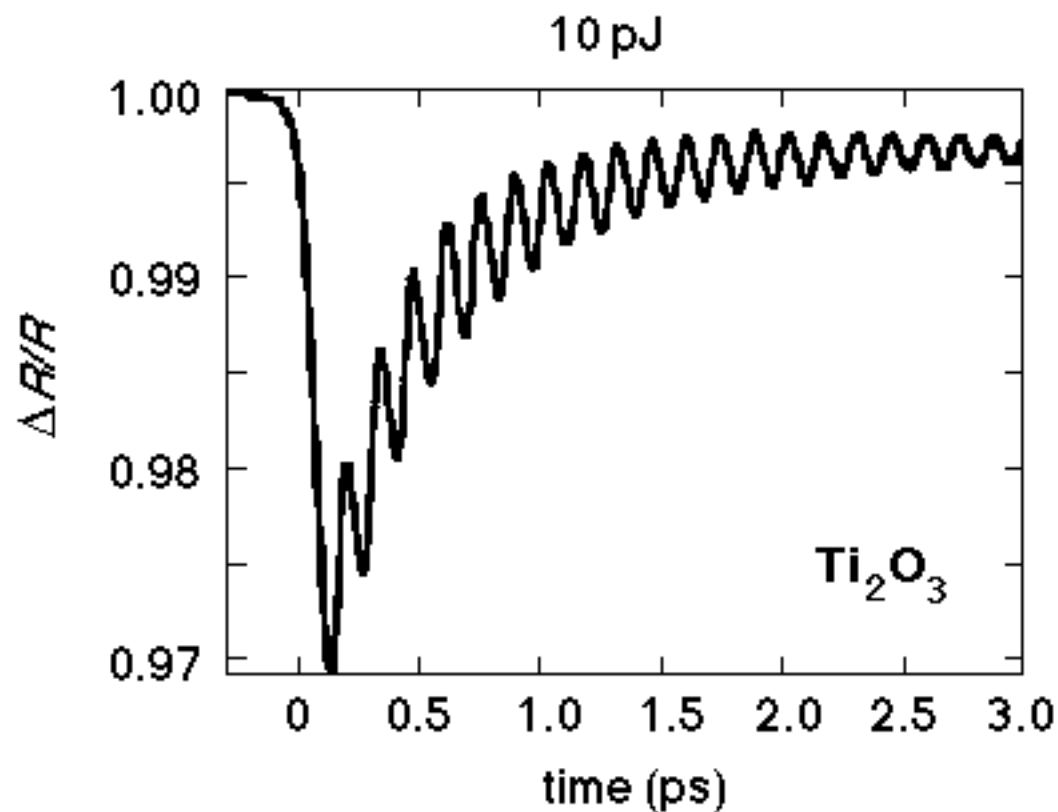
...read out with third beam



IMPULSIVE RAMAN SCATTERING



LARGE AMPLITUDE OSCILLATIONS

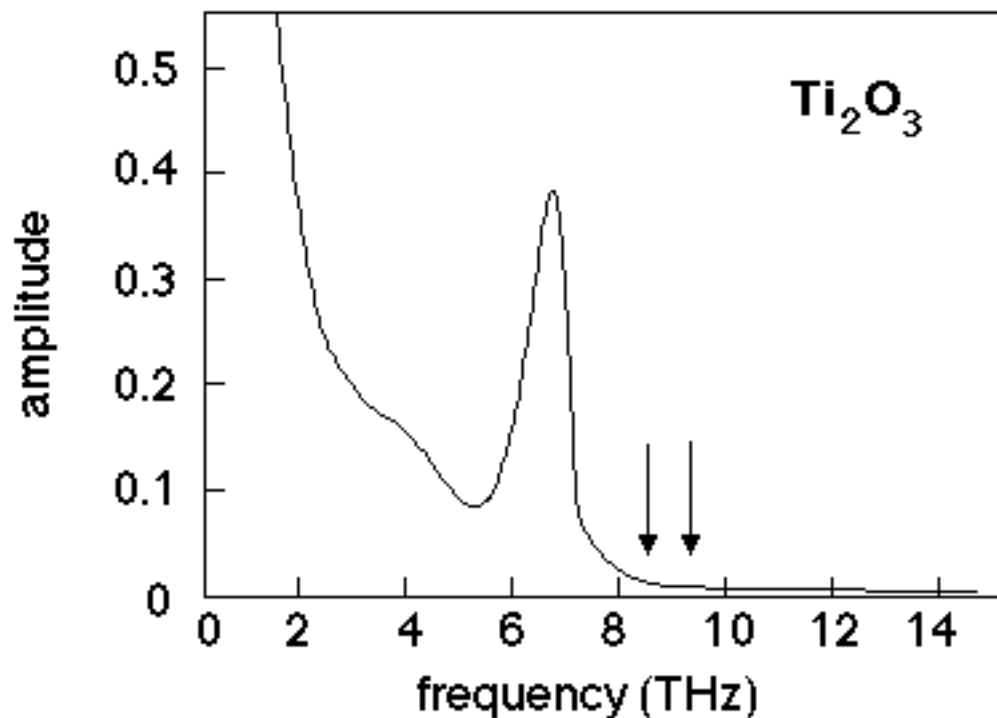


T.K. Cheng *et al.*, *Appl. Phys. Lett.* **57** (1990) 1004

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



LARGE AMPLITUDE OSCILLATIONS

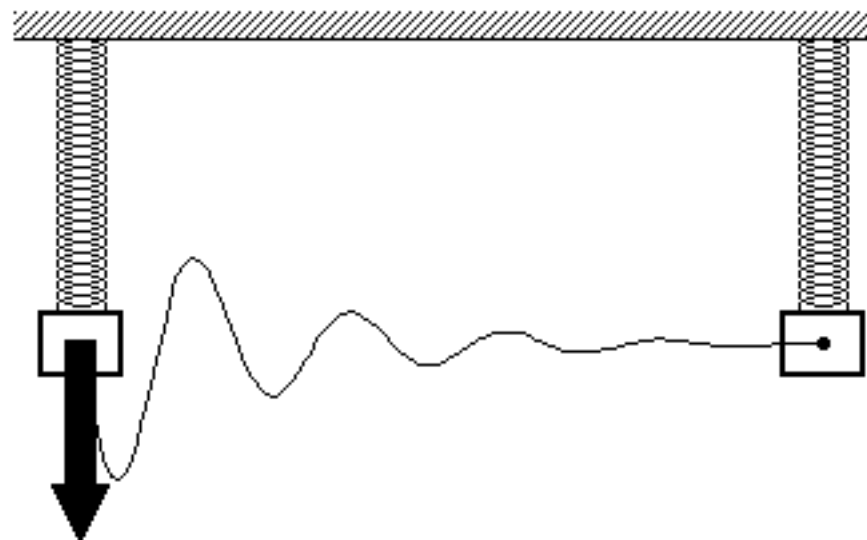


T.K. Cheng *et al.*, *Appl. Phys. Lett.* **57** (1990) 1004

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



IMPULSIVE RAMAN SCATTERING



- need to match q
- sinusoidal oscillation



DISPLACIVE EXCITATION



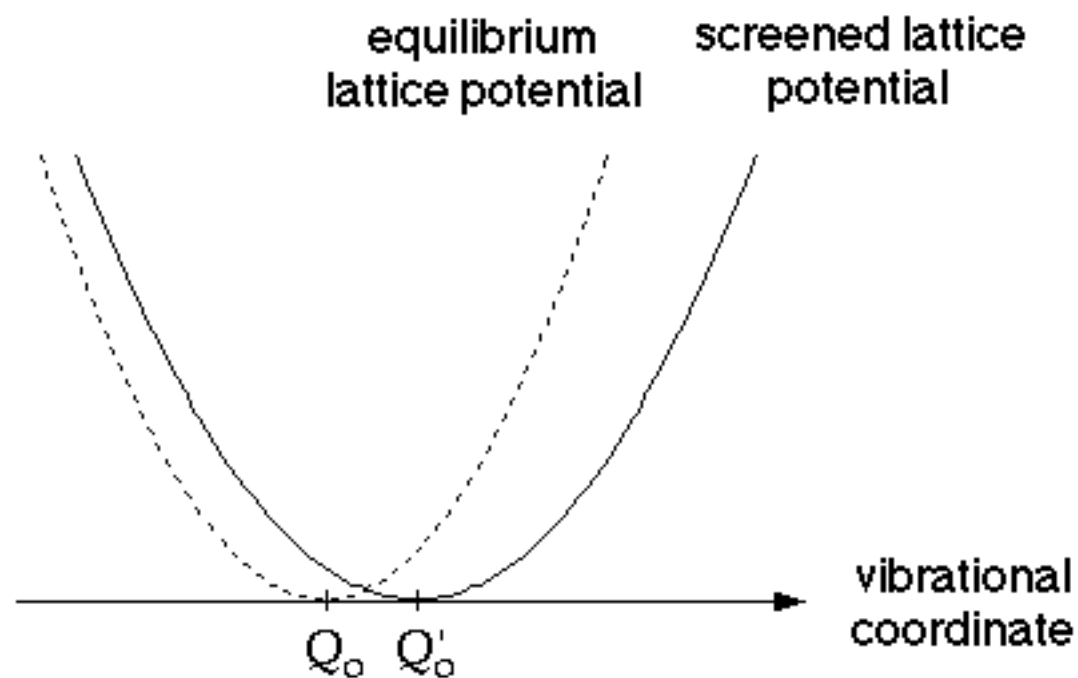
DISPLACIVE EXCITATION



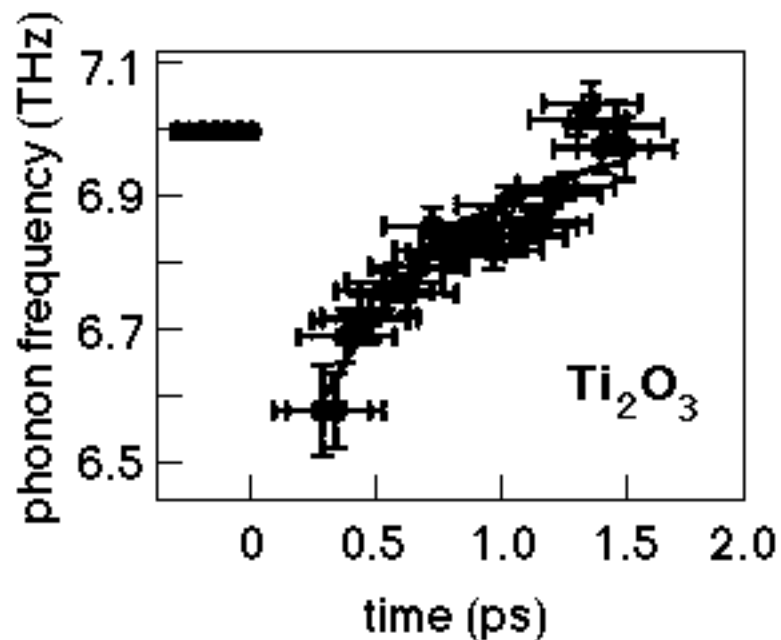
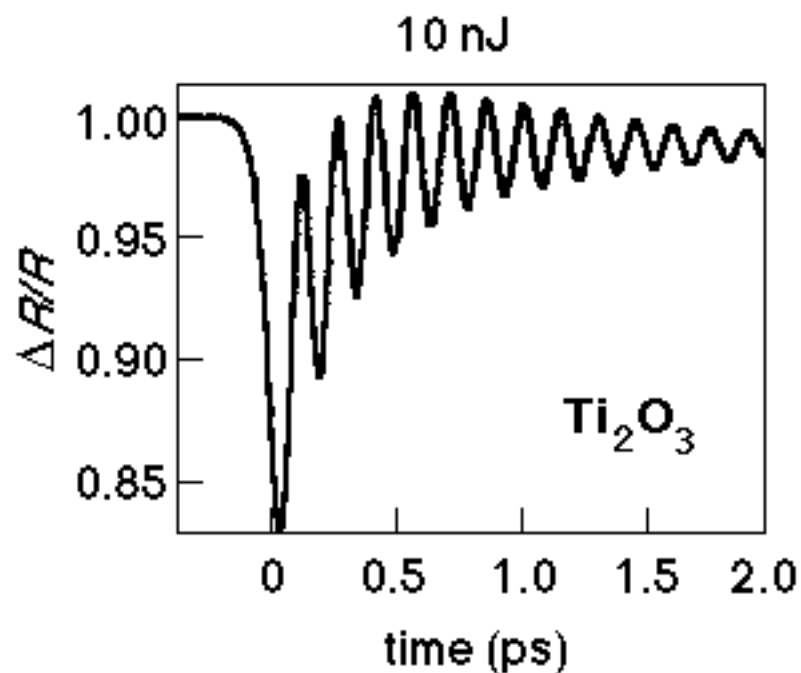
DISPLACIVE EXCITATION



DISPLACIVE EXCITATION

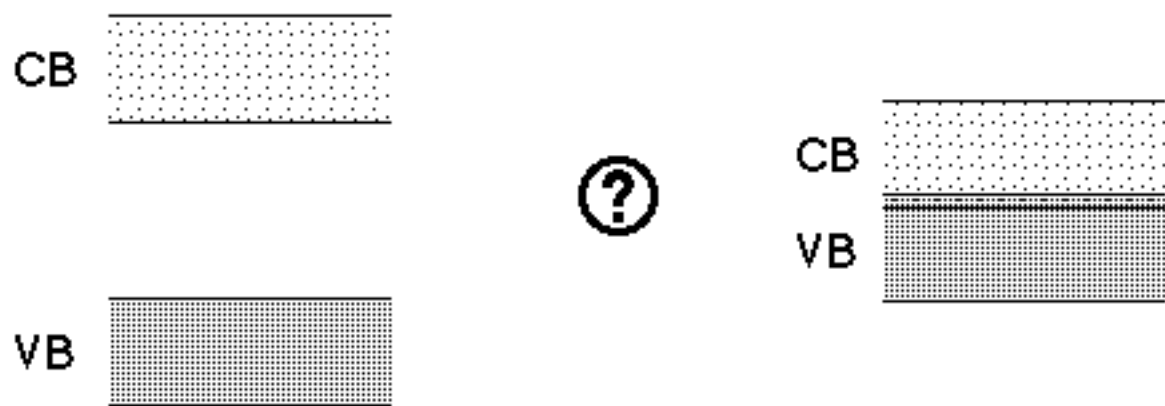


DISPLACIVE EXCITATION



BEYOND PHONONS

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



BEYOND PHONONS

intrinsic properties

measured properties

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



$$R$$

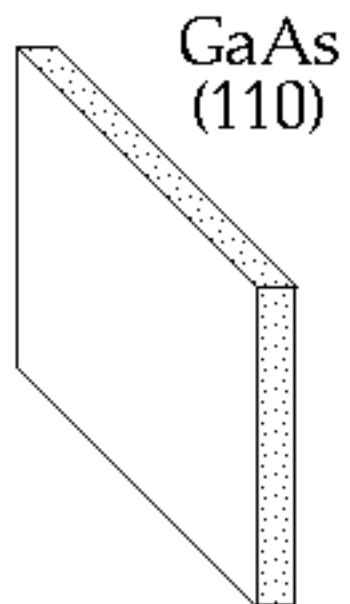
$$\begin{aligned} \epsilon', \epsilon''(\omega) \\ \epsilon', \epsilon''(2\omega) \\ \chi(2) \end{aligned}$$



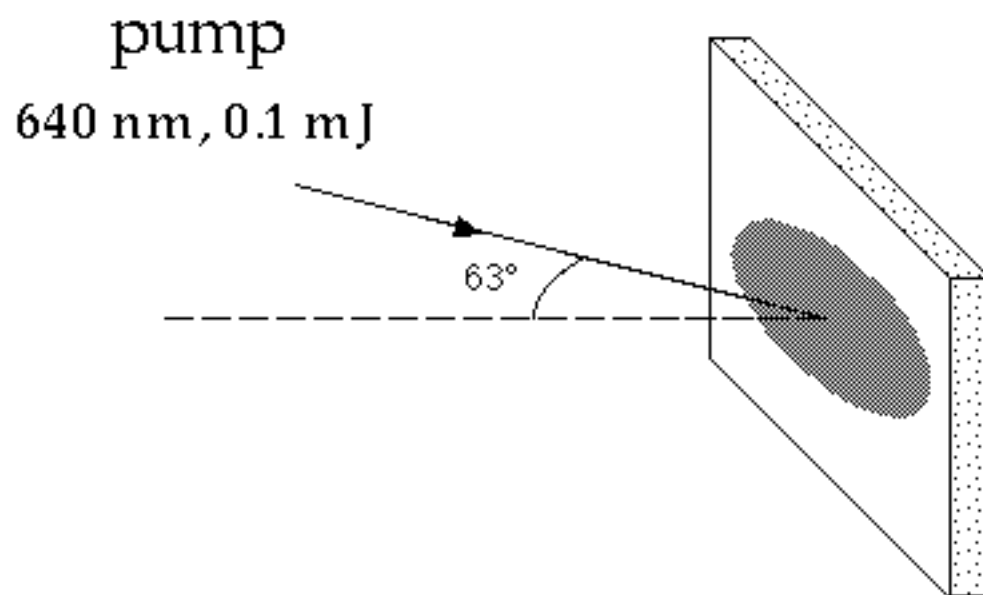
SHG



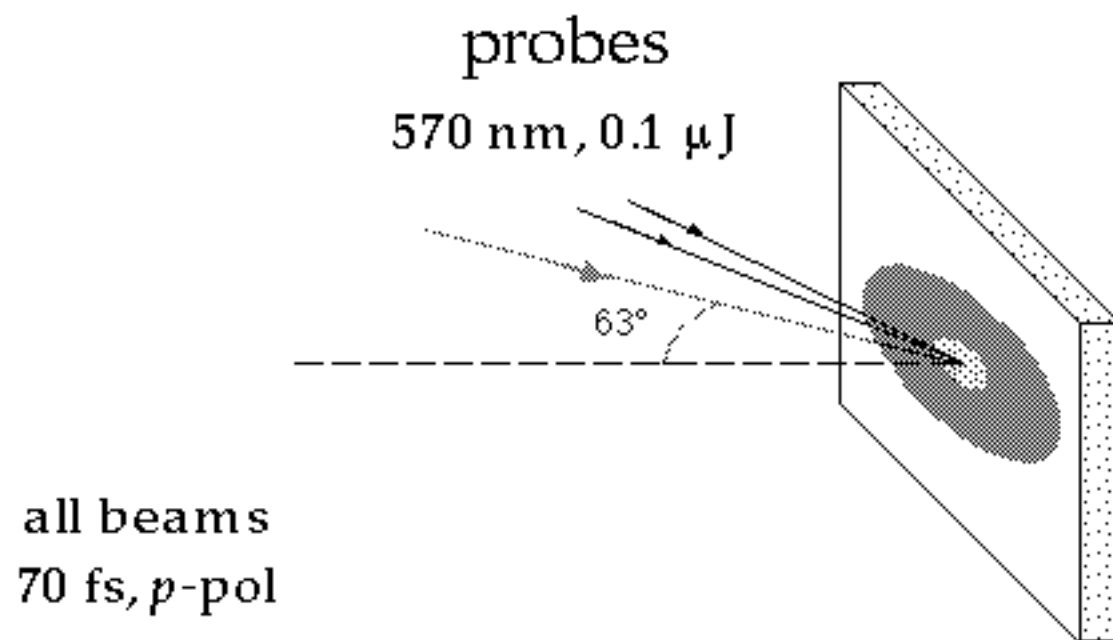
2.2 eV DIELECTRIC CONSTANT



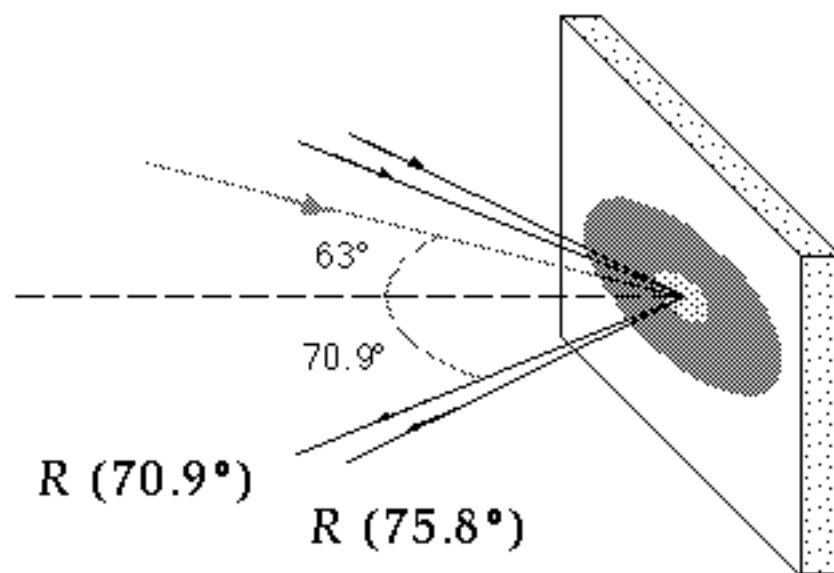
2.2 eV DIELECTRIC CONSTANT



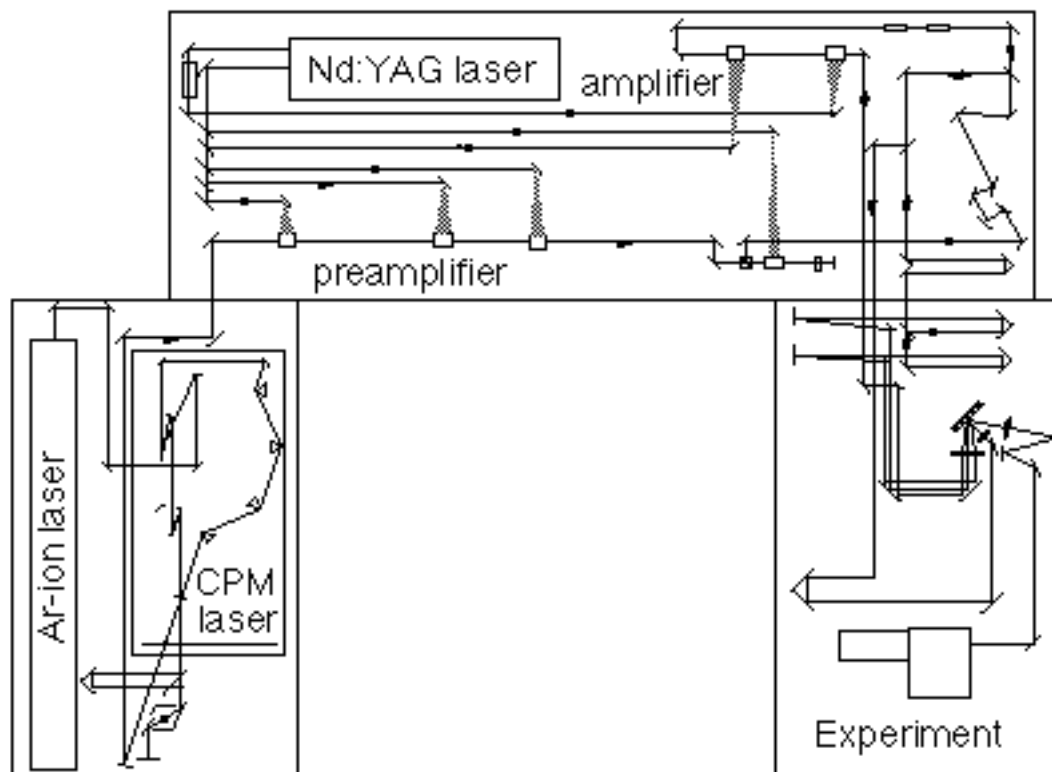
2.2 eV DIELECTRIC CONSTANT



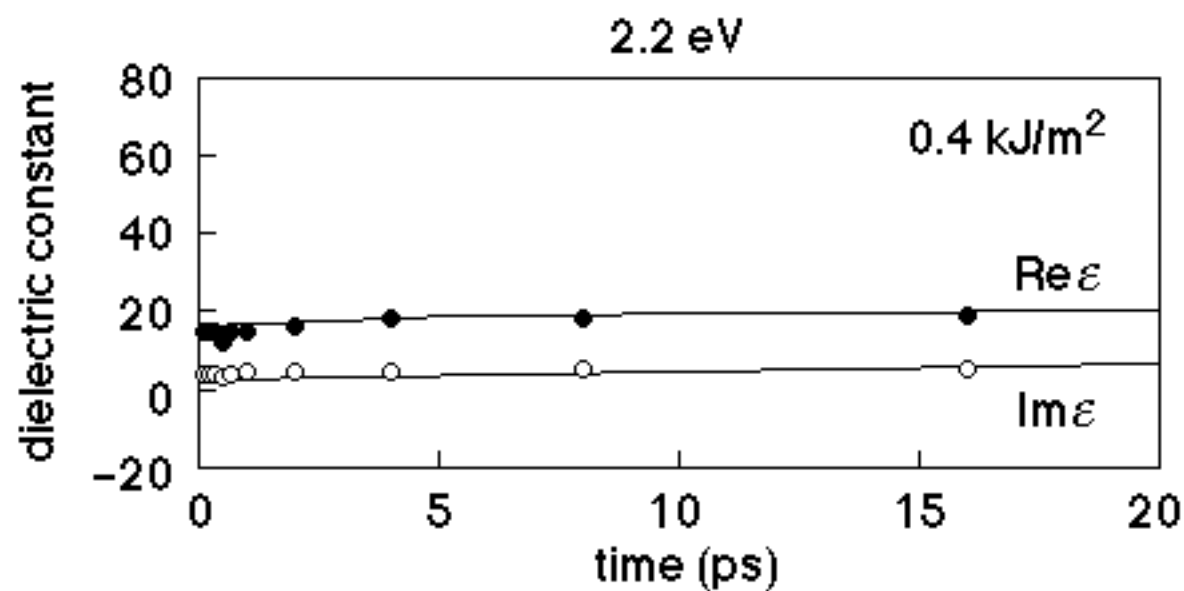
2.2 eV DIELECTRIC CONSTANT



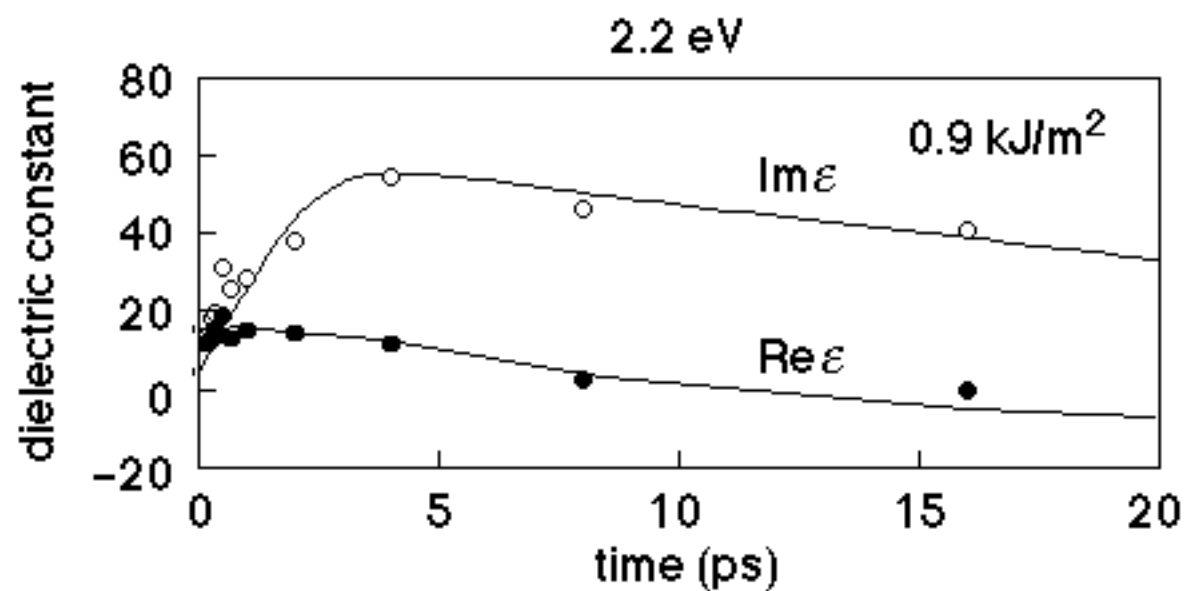
SETUP



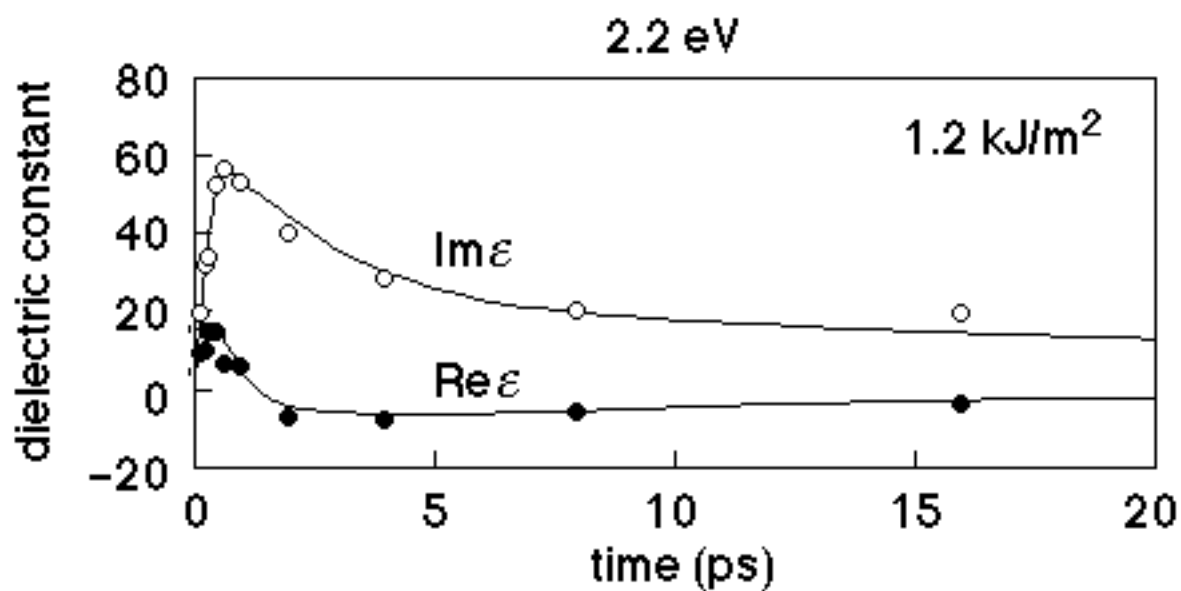
TIME DEPENDENCE



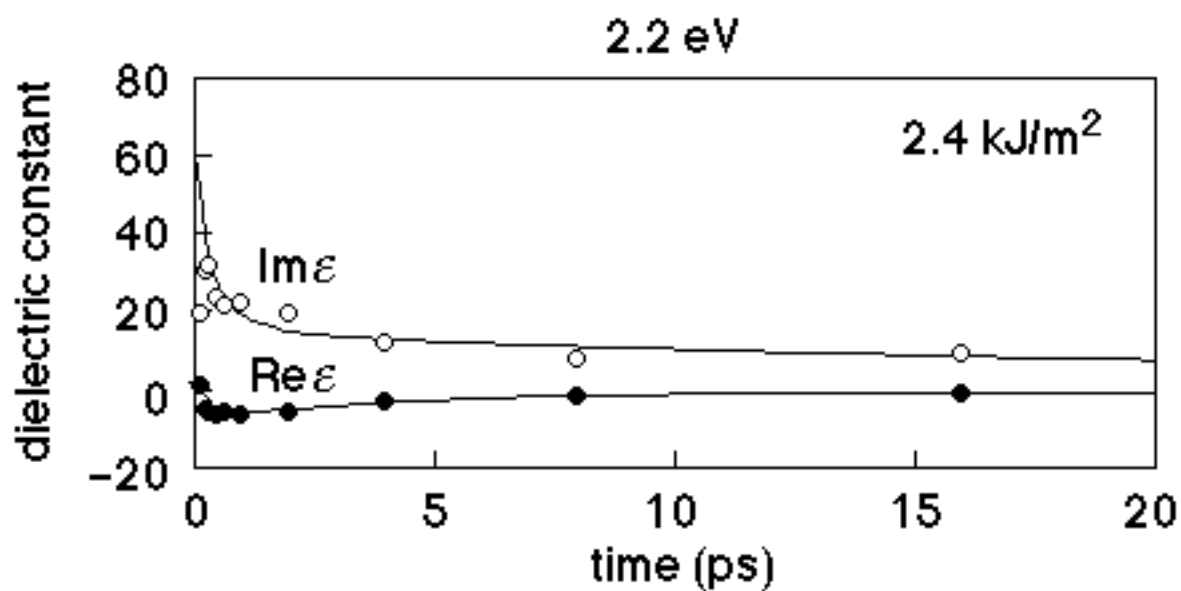
TIME DEPENDENCE



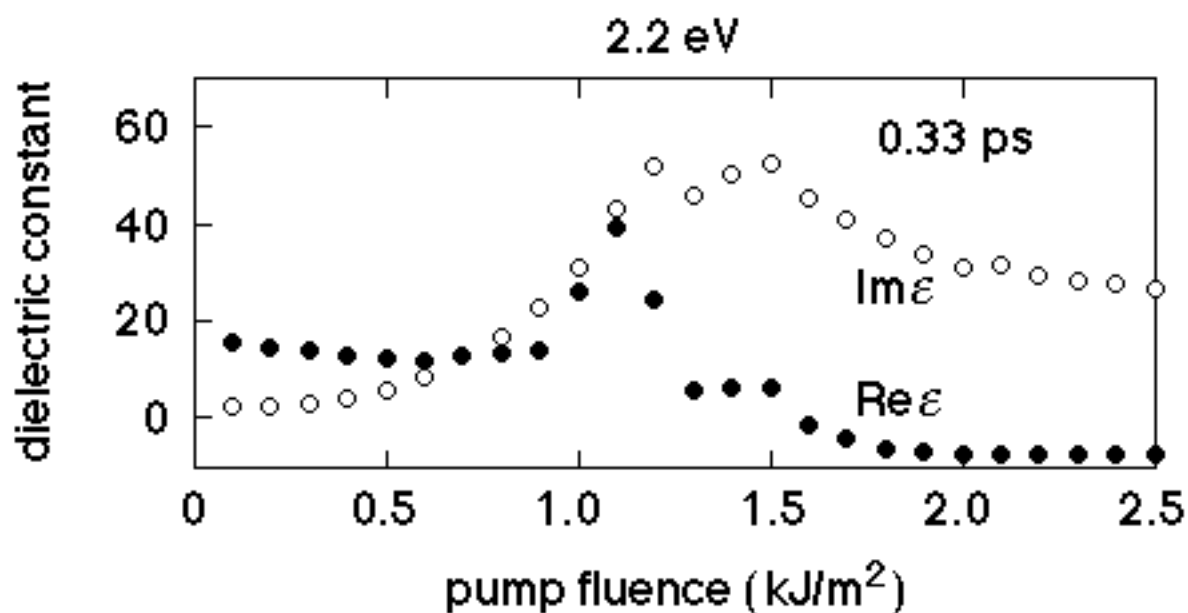
TIME DEPENDENCE



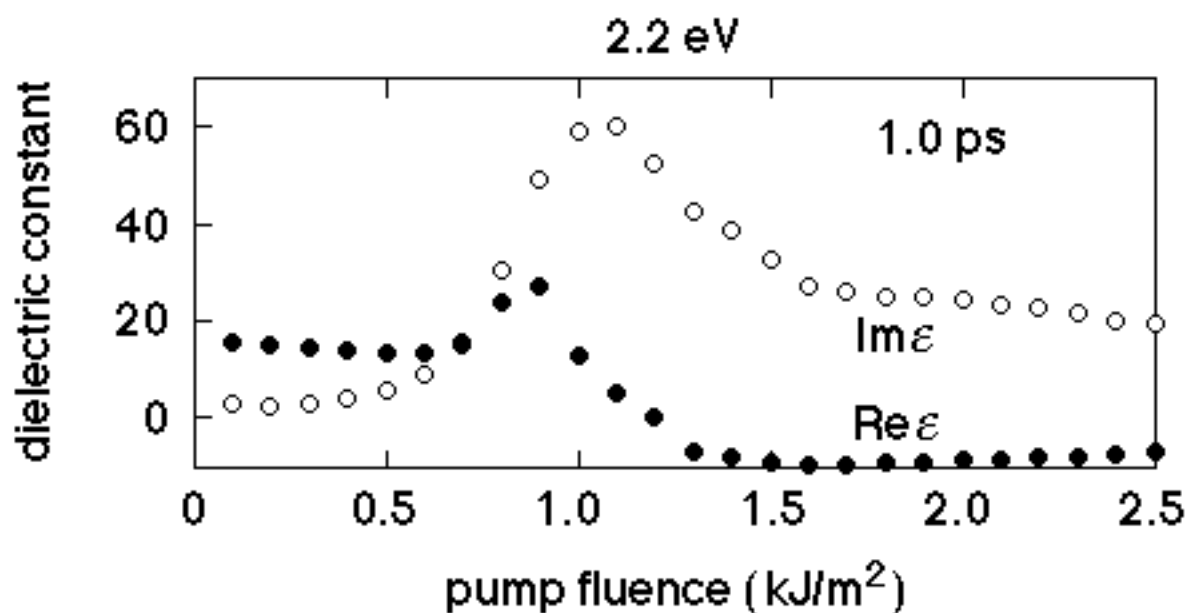
TIME DEPENDENCE



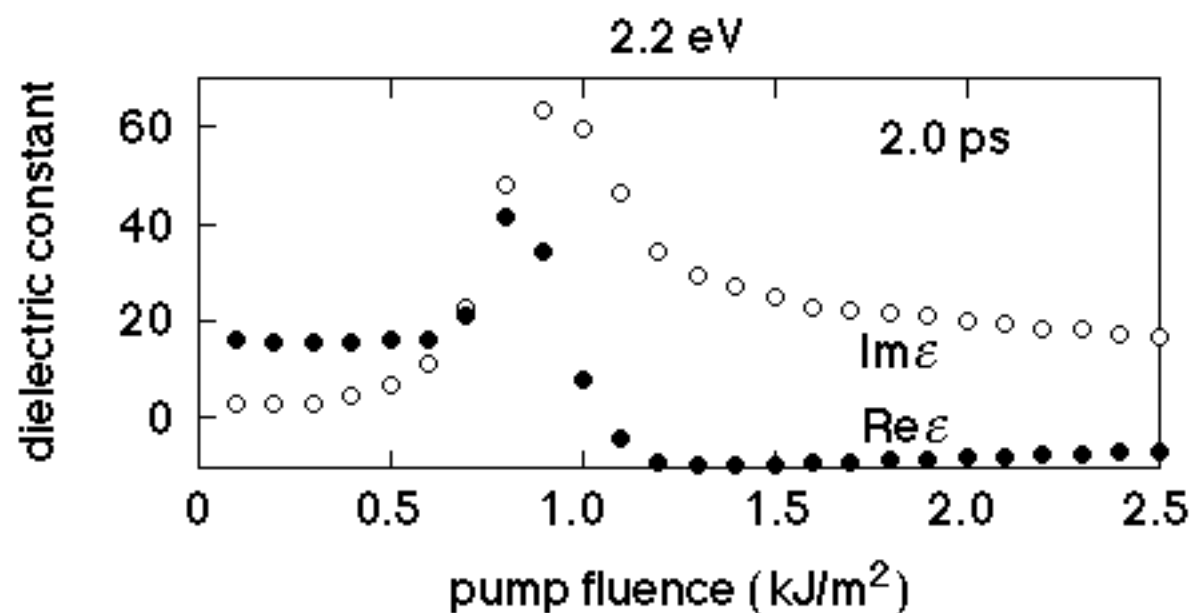
FLUENCE DEPENDENCE



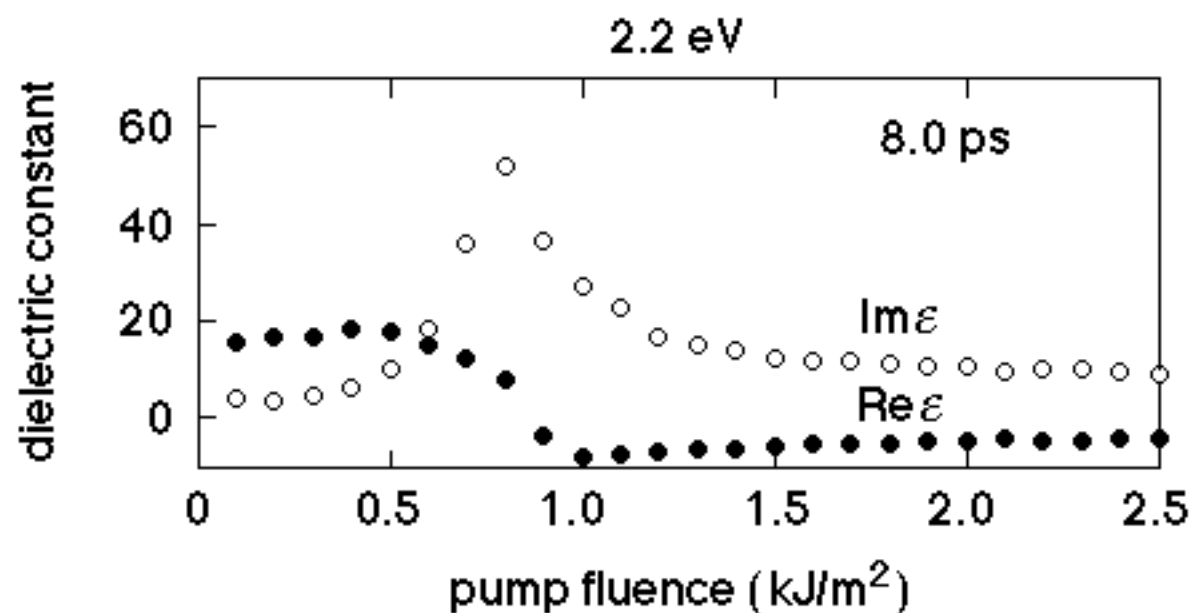
FLUENCE DEPENDENCE



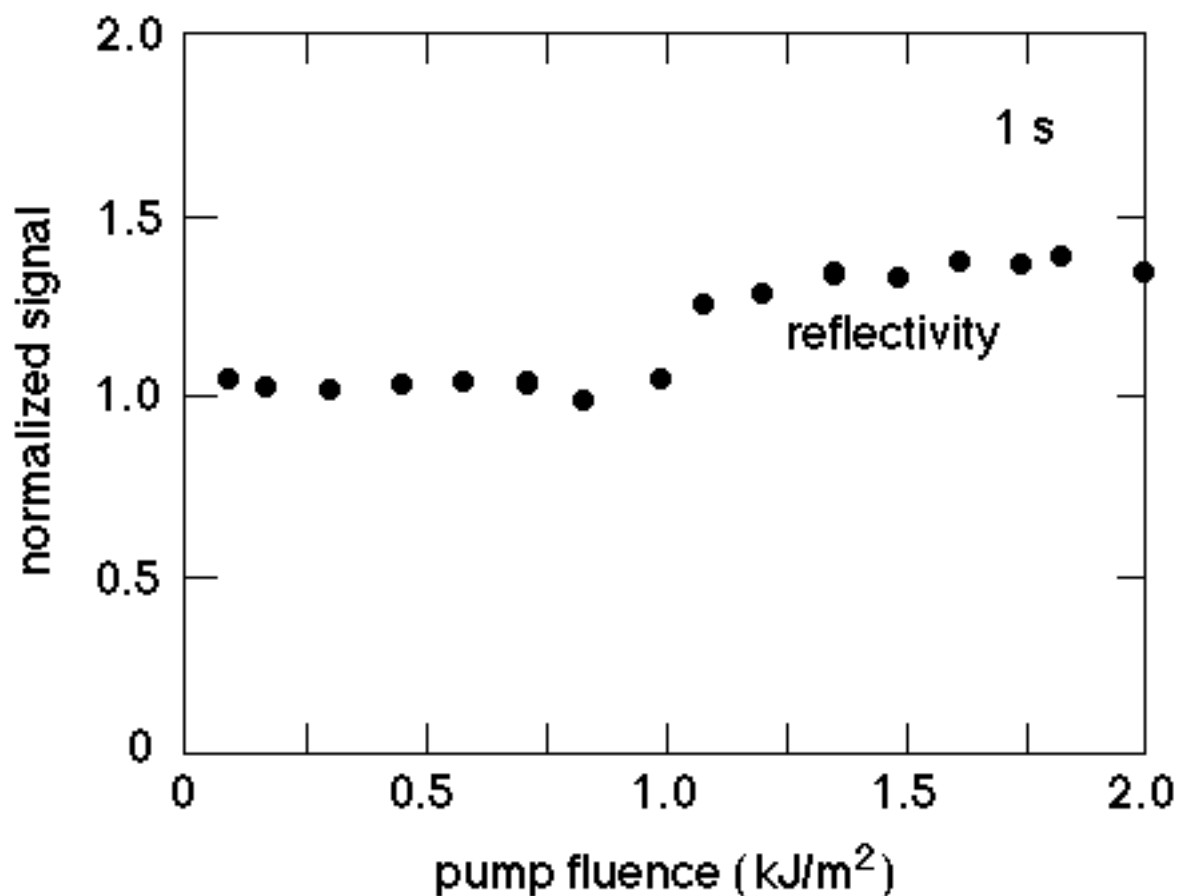
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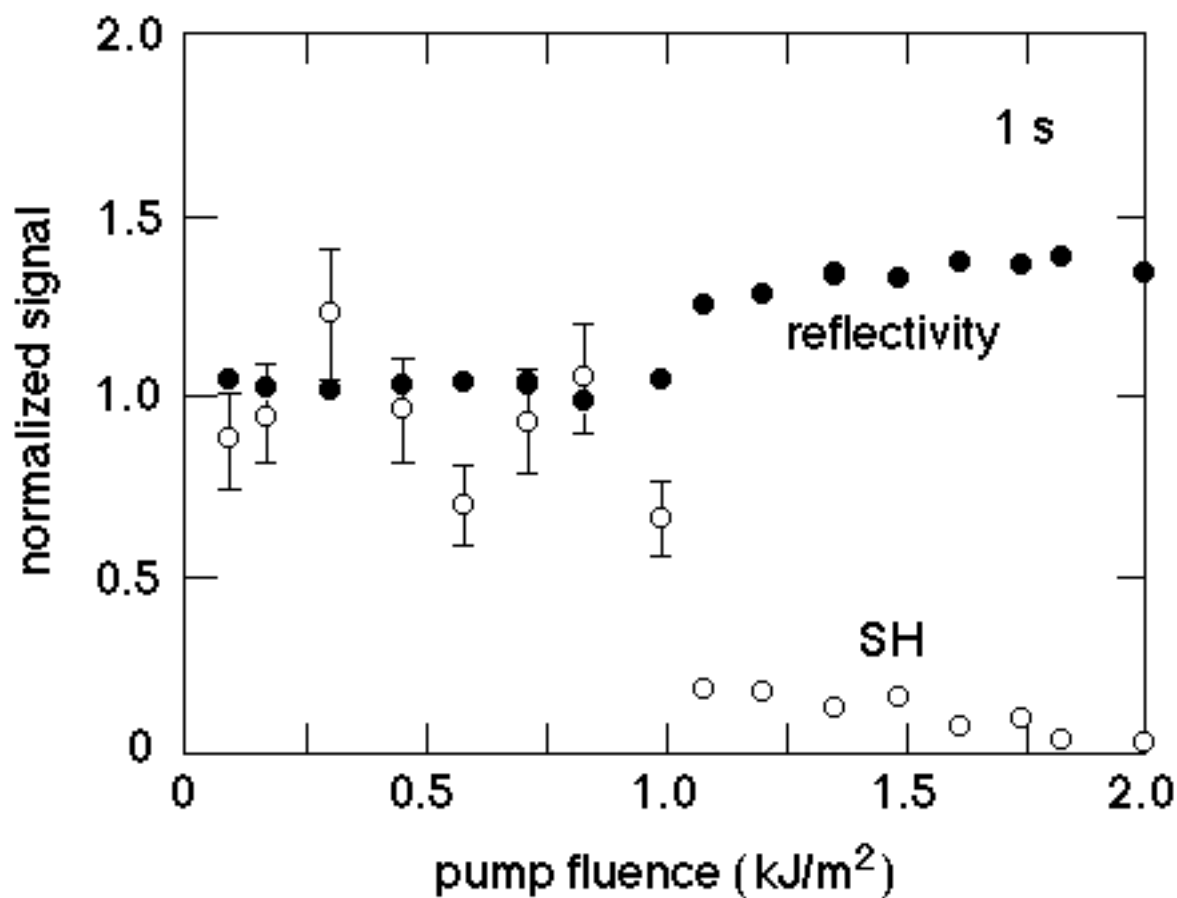
FLUENCE DEPENDENCE



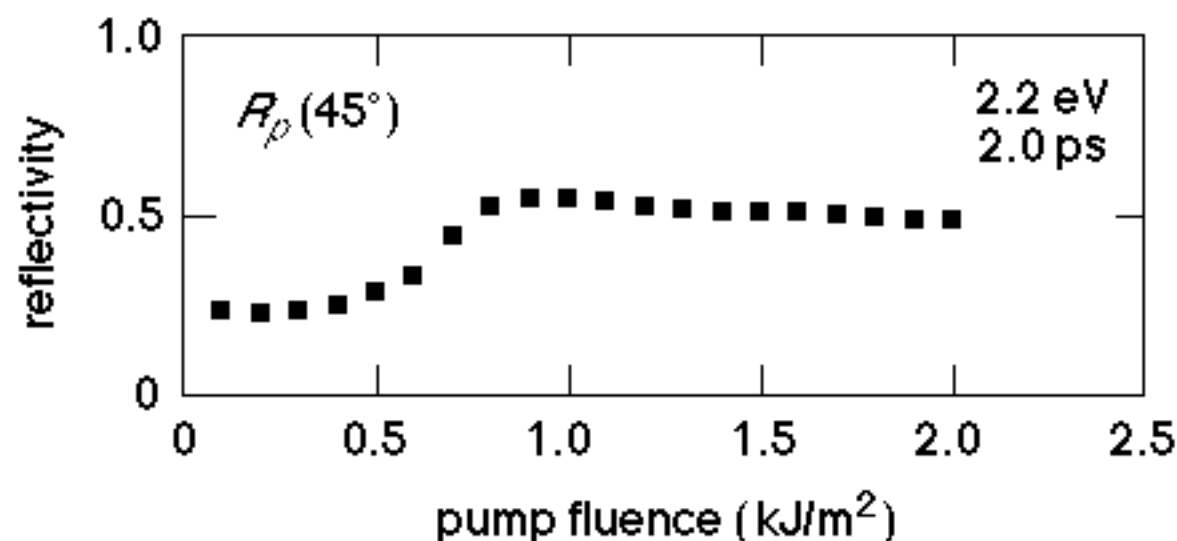
REVERSIBLE vs. IRREVERSIBLE



REVERSIBLE vs. IRREVERSIBLE



SIMPLE MODEL



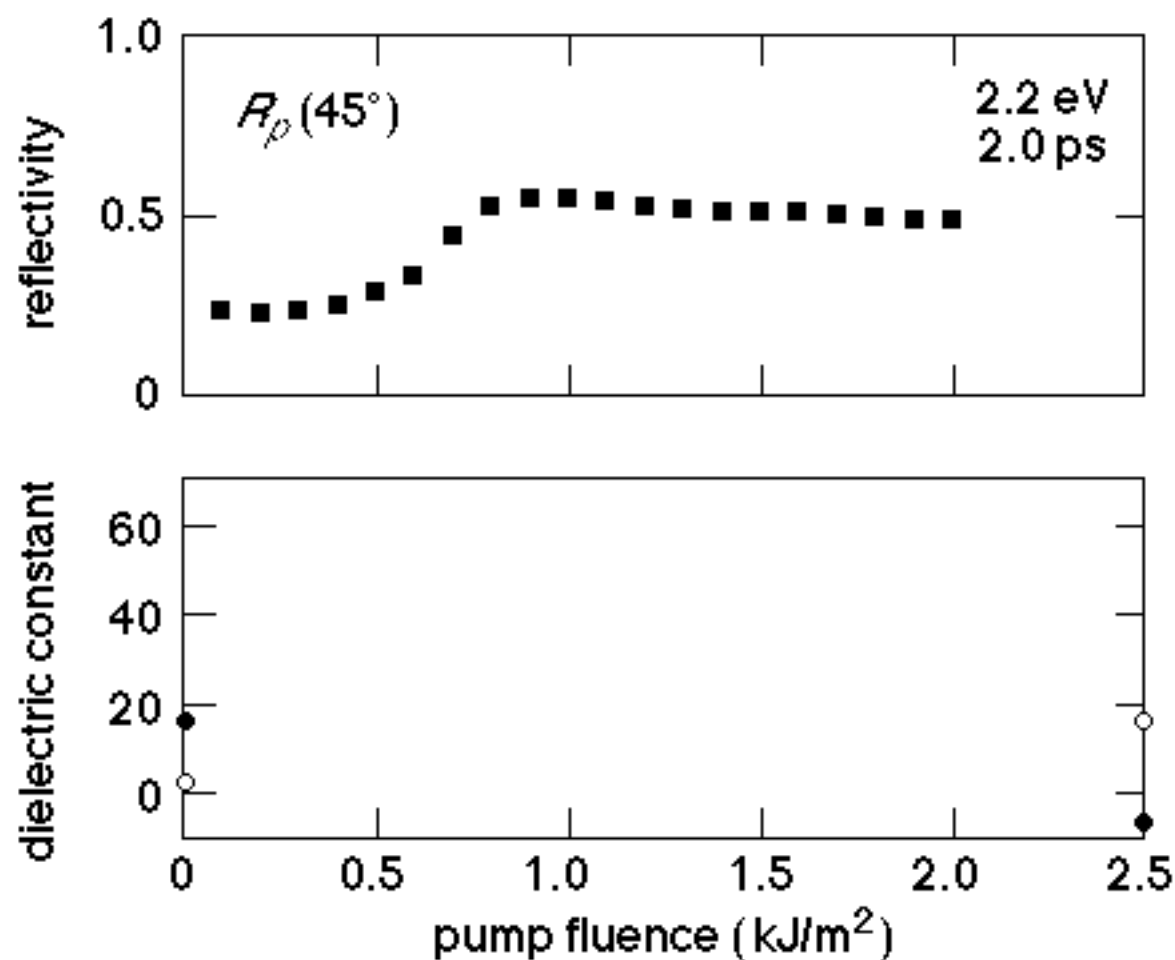
Drude model:

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

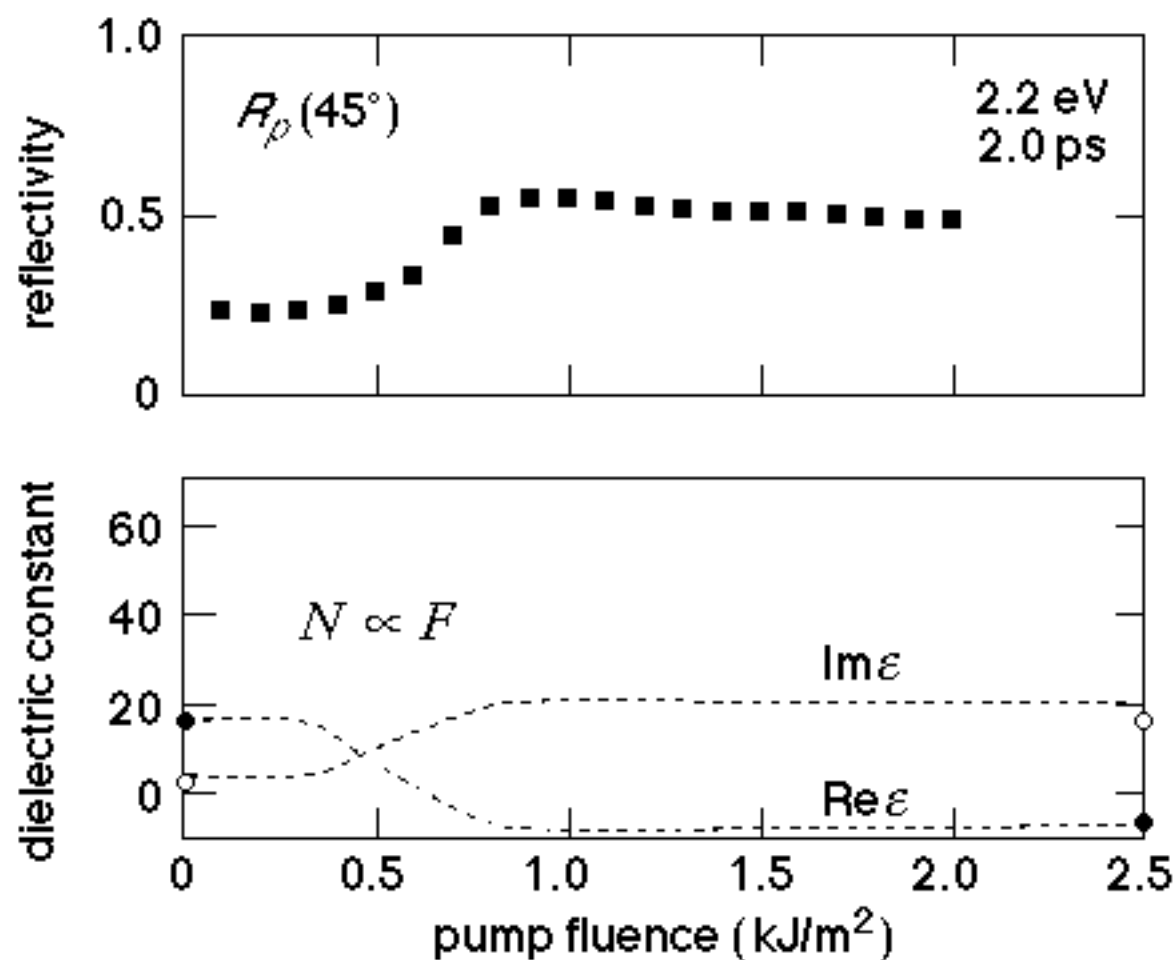
$$\text{Im}(\epsilon) = \frac{Ne^2}{m\epsilon_0} \frac{\gamma\omega}{\omega^4 + \gamma^2\omega^2}$$



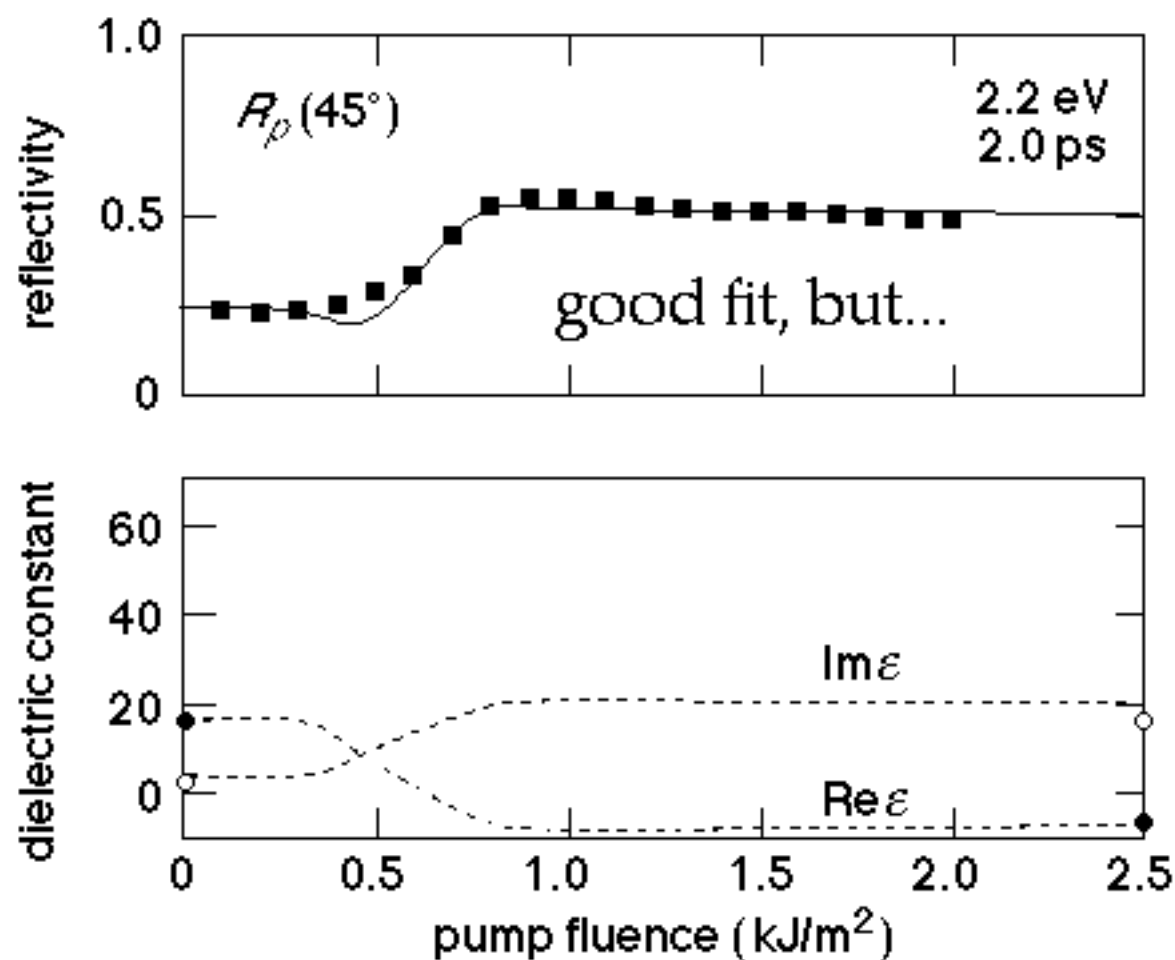
SIMPLE MODEL



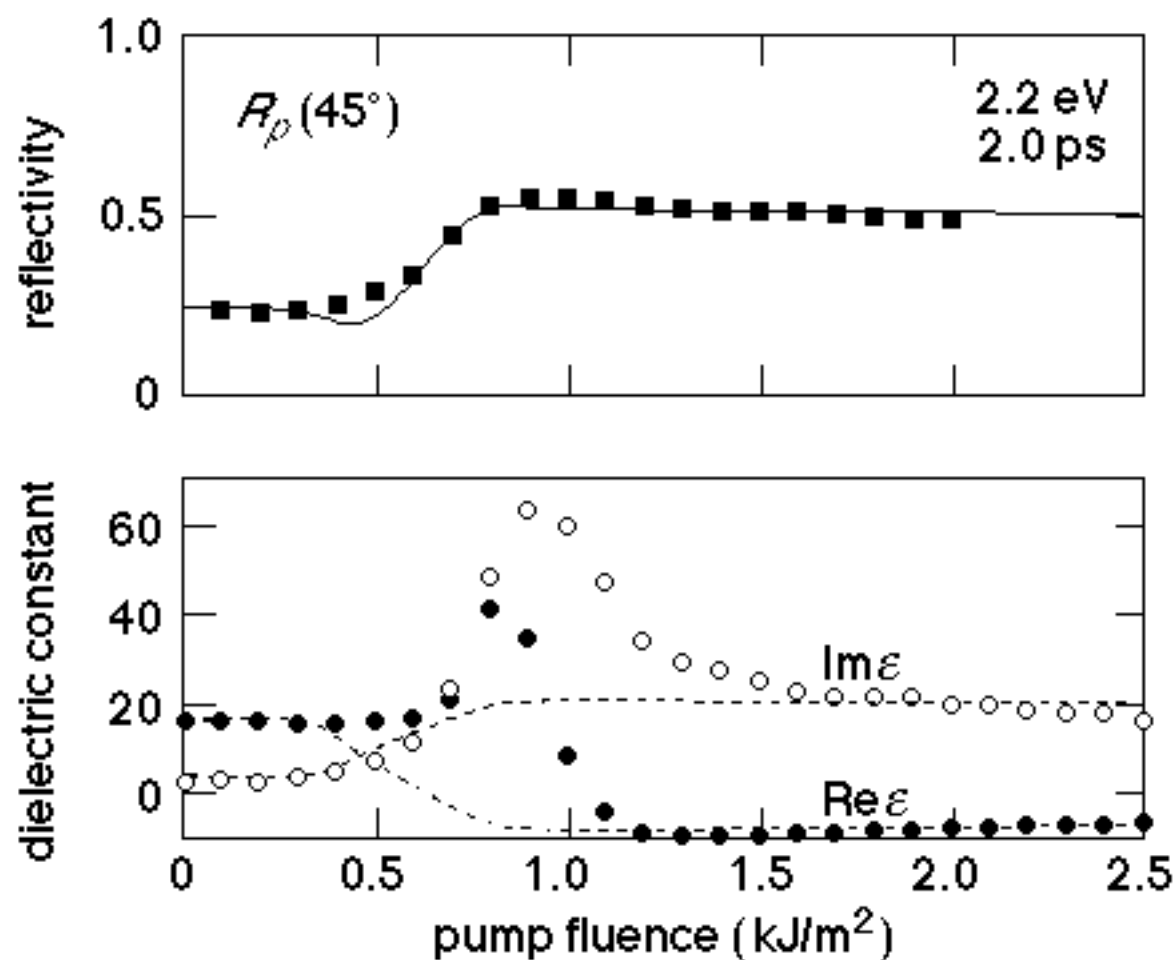
SIMPLE MODEL



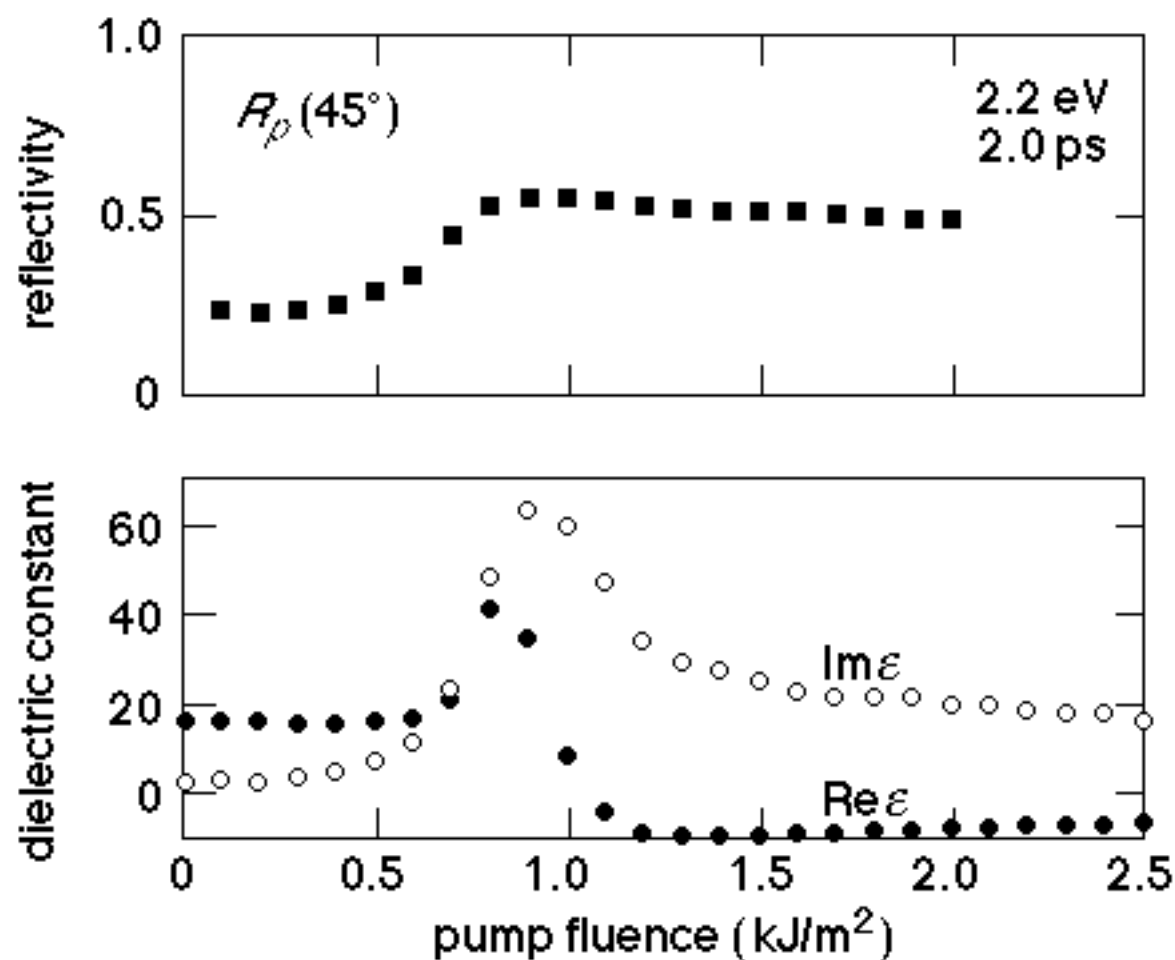
SIMPLE MODEL



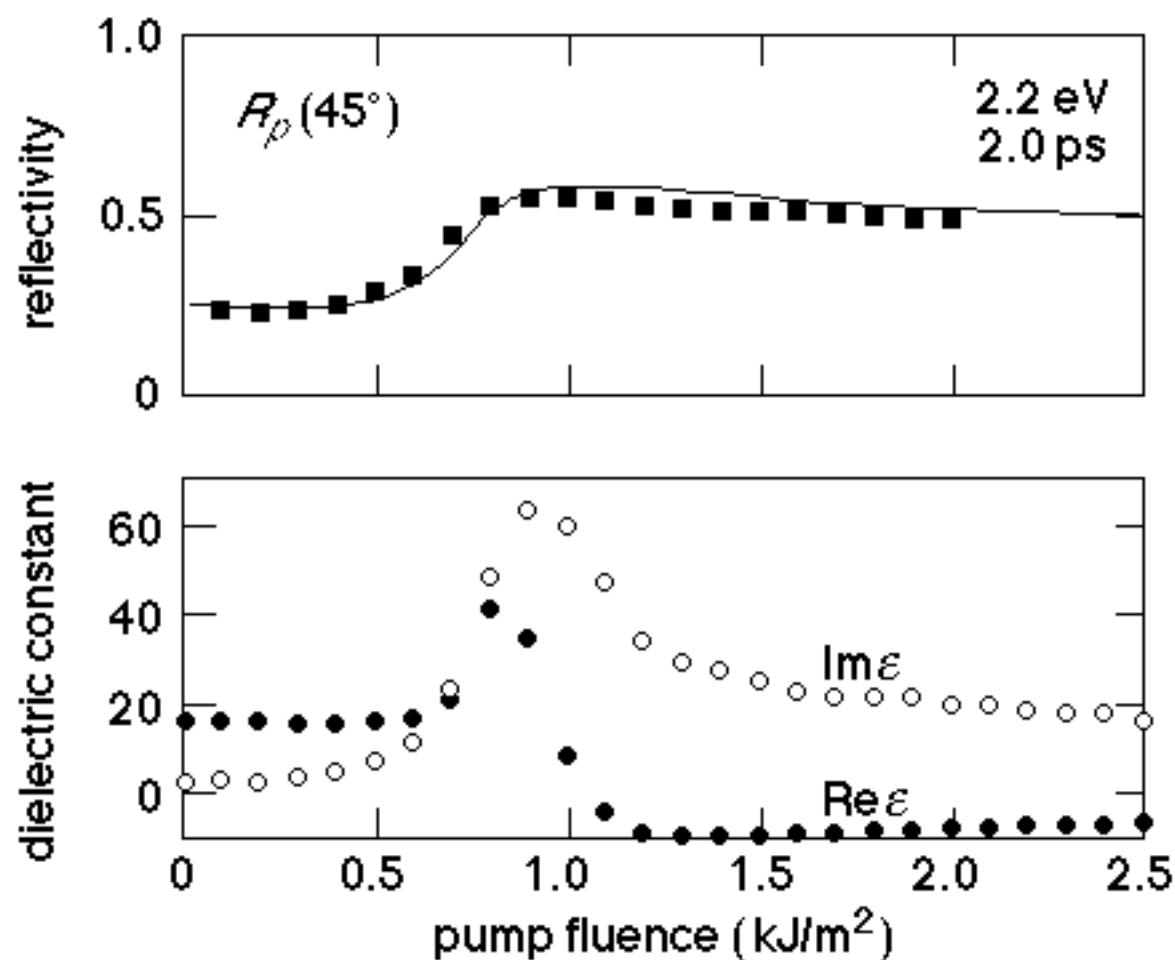
SIMPLE MODEL



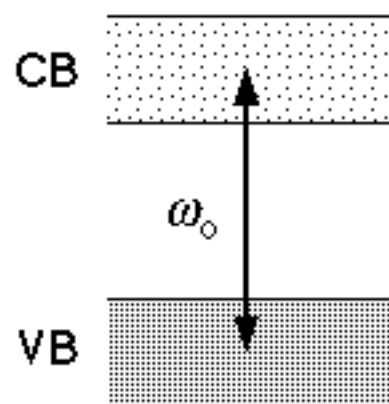
SIMPLE MODEL



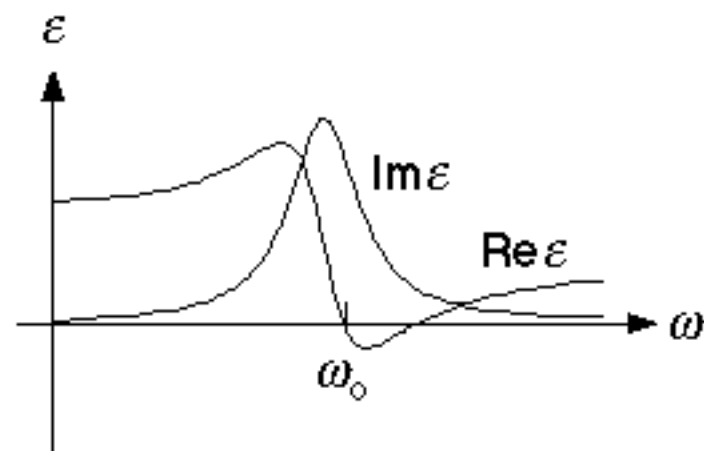
SIMPLE MODEL



WHAT IS HAPPENING?



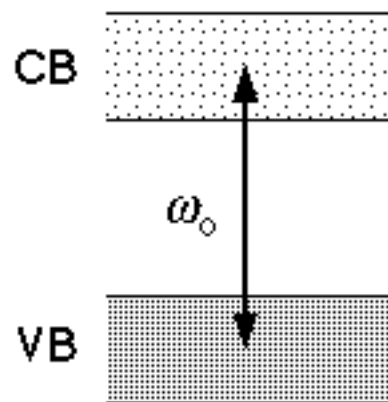
semiconductor



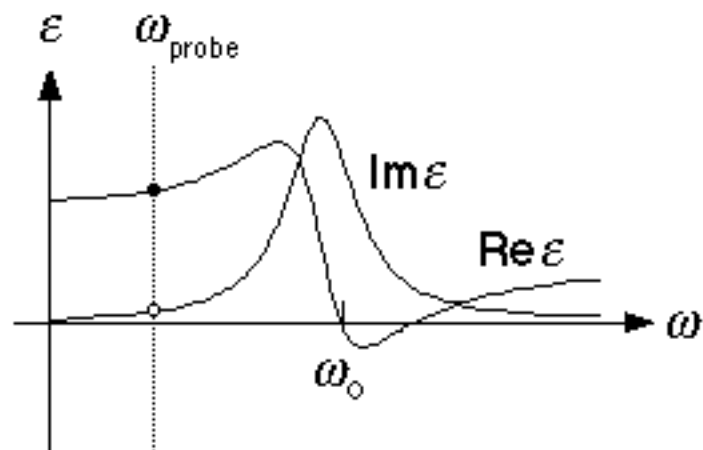
$$\omega_0 \approx 4.5 \text{ eV}$$



WHAT IS HAPPENING?



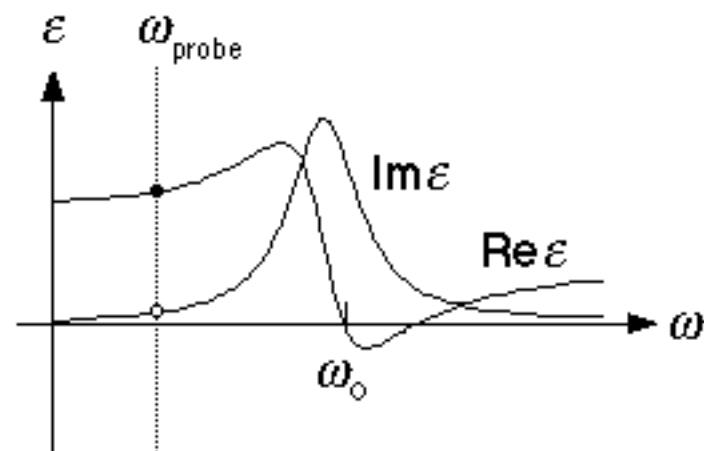
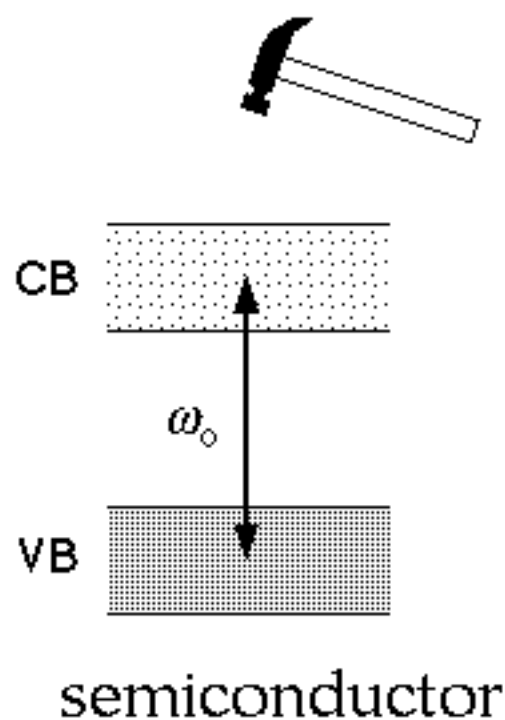
semiconductor



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



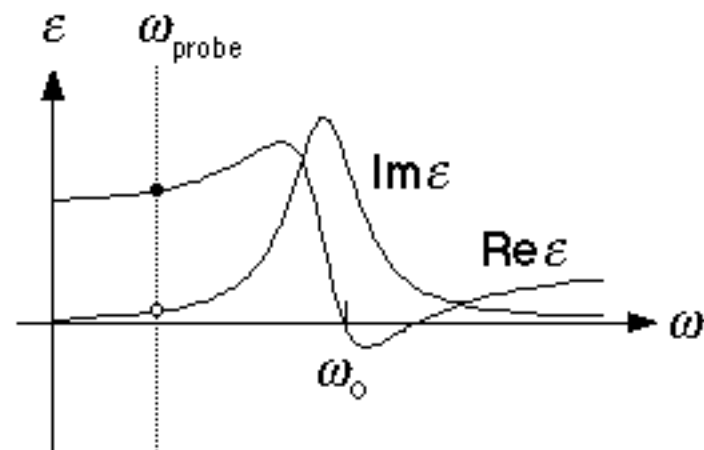
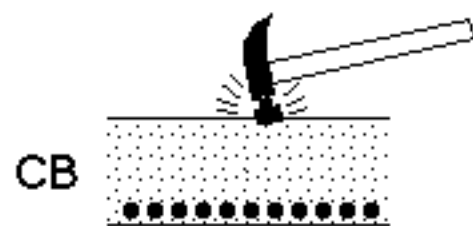
WHAT IS HAPPENING?



pump pulse excites
valence electrons...



WHAT IS HAPPENING?

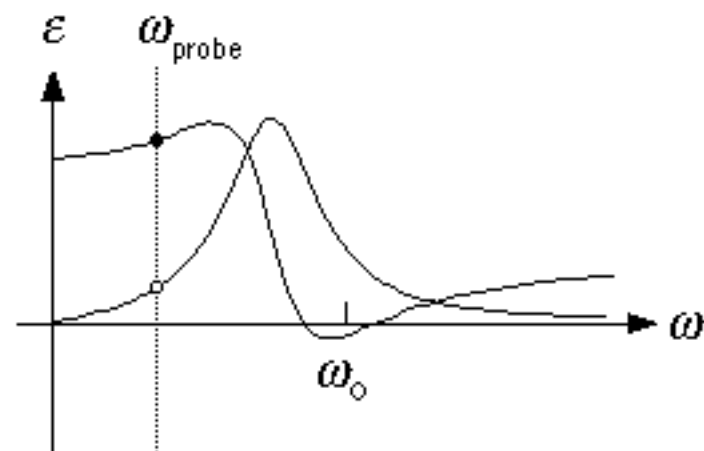
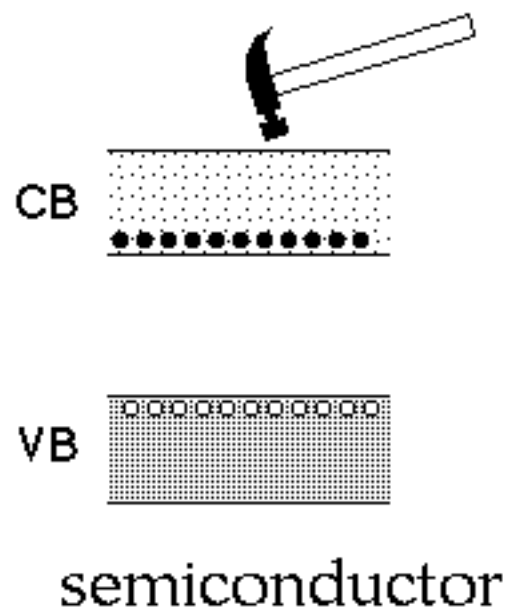


semiconductor

pump pulse excites
valence electrons...



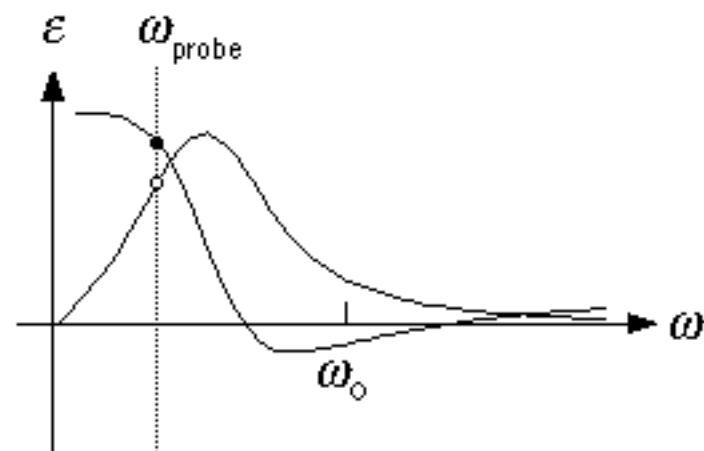
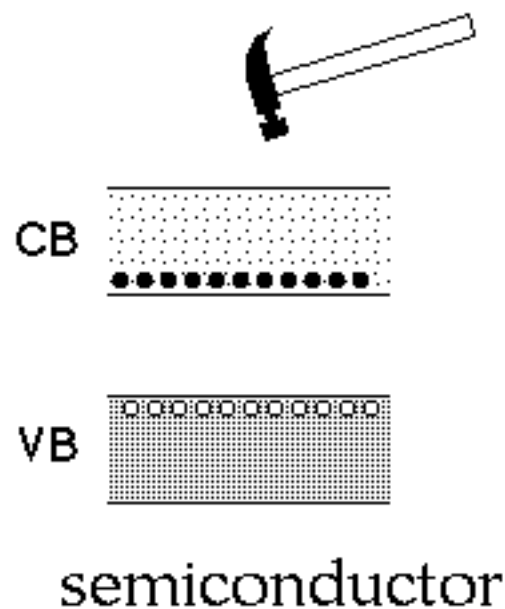
WHAT IS HAPPENING?



...and induces change
in bandstructure



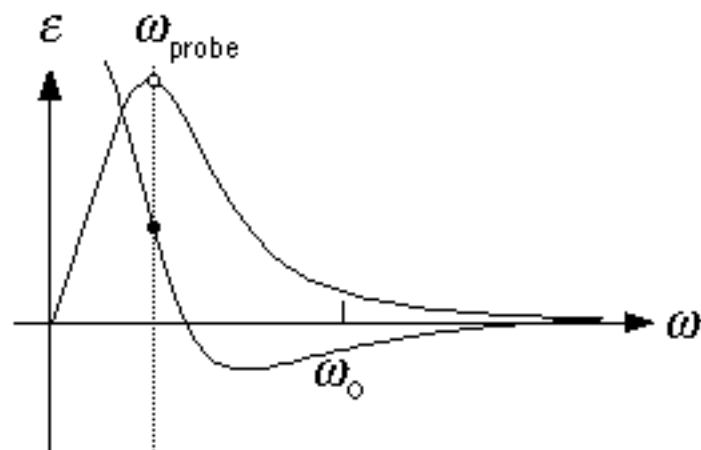
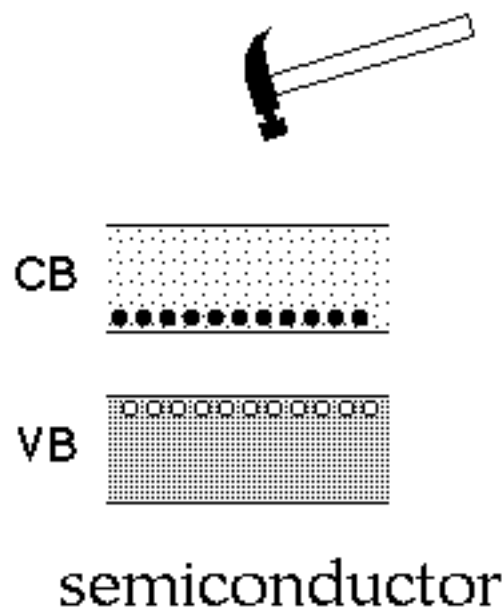
WHAT IS HAPPENING?



...and induces change
in bandstructure



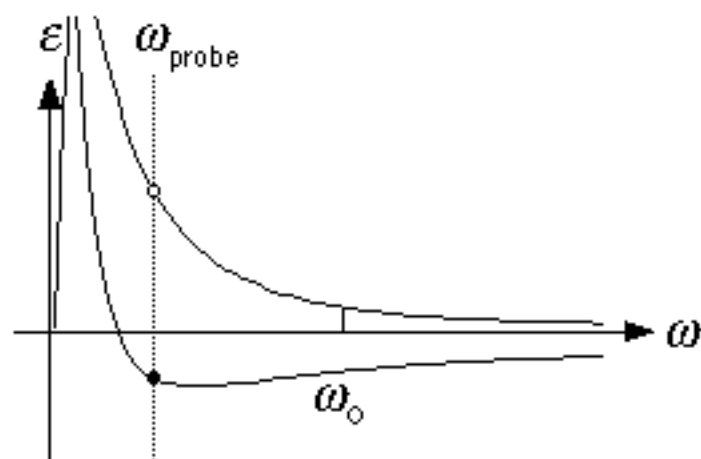
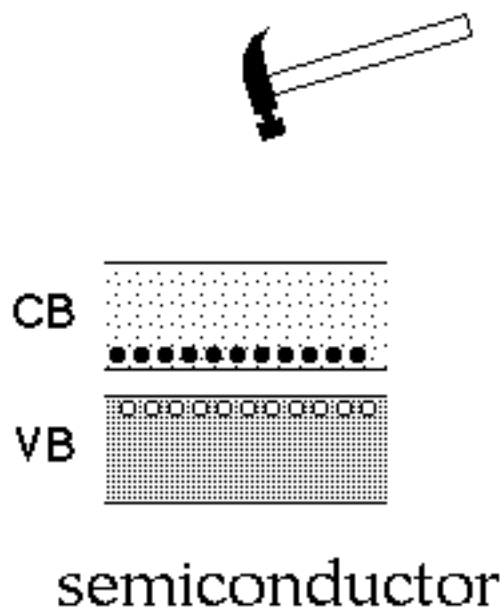
WHAT IS HAPPENING?



...and induces change
in bandstructure



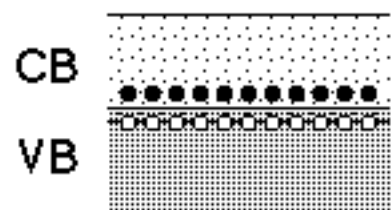
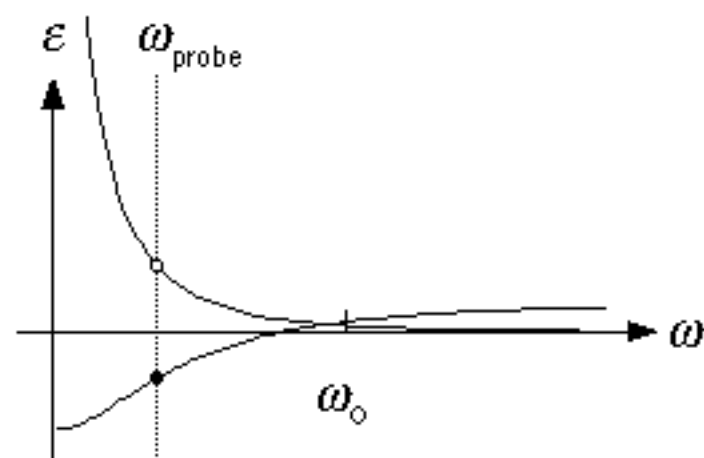
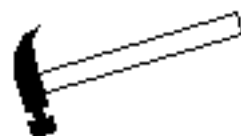
WHAT IS HAPPENING?



...and induces change
in bandstructure



WHAT IS HAPPENING?

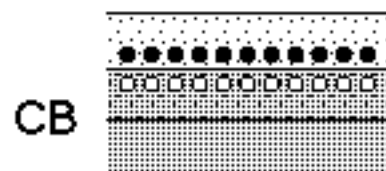
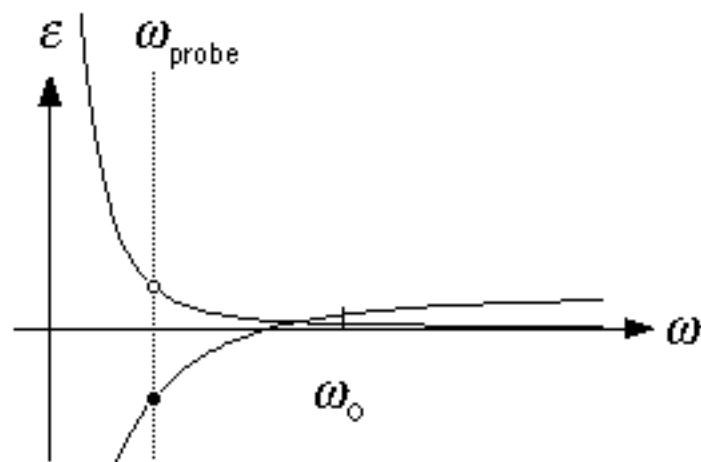
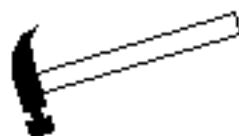


semimetal

...and induces change
in bandstructure



WHAT IS HAPPENING?

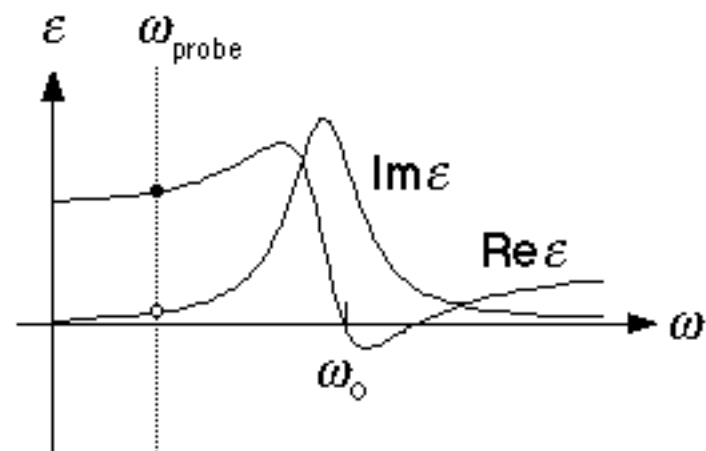
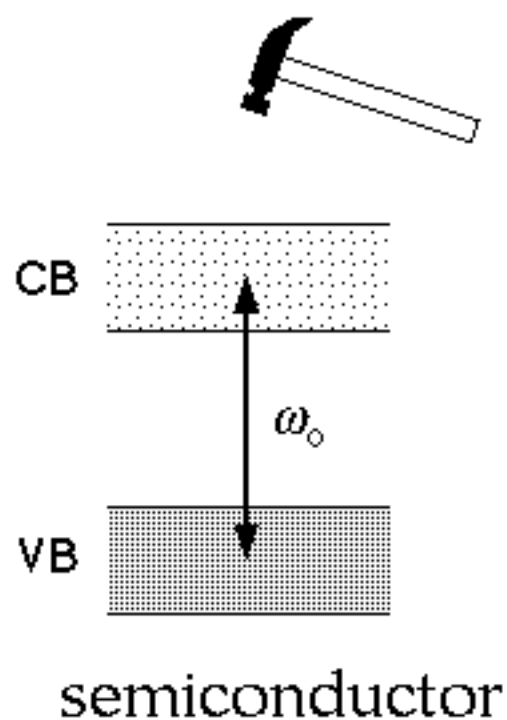


metal

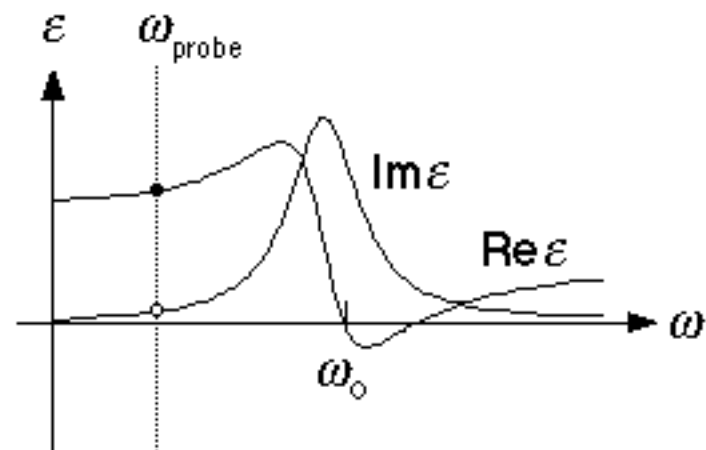
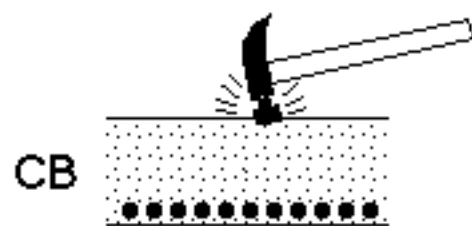
...and induces change
in bandstructure



WHAT IS HAPPENING?



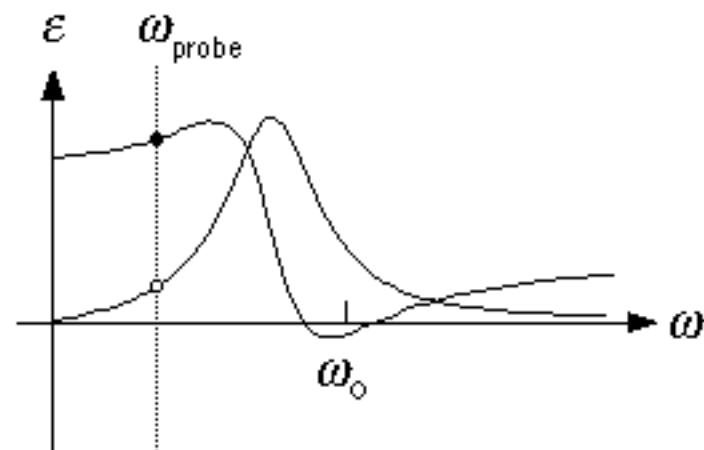
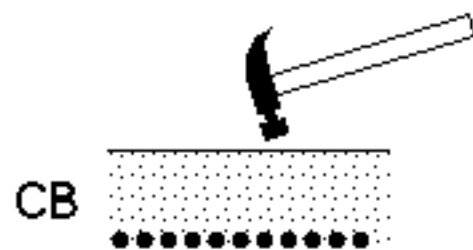
WHAT IS HAPPENING?



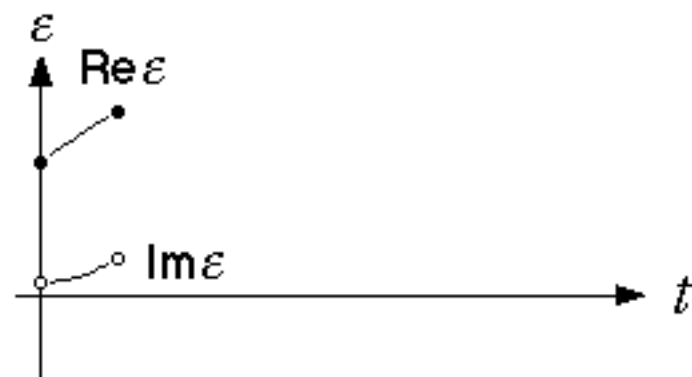
semiconductor



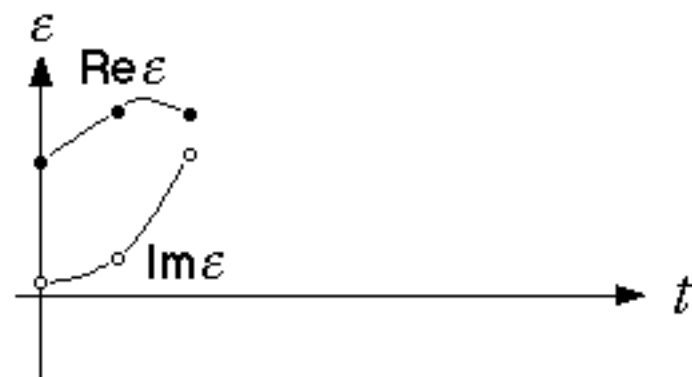
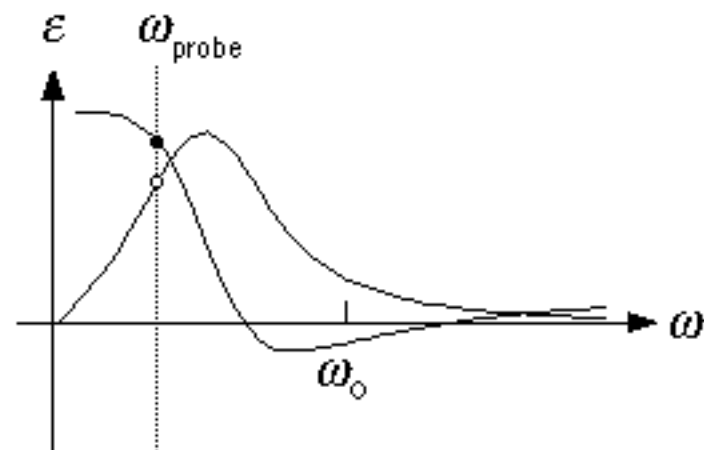
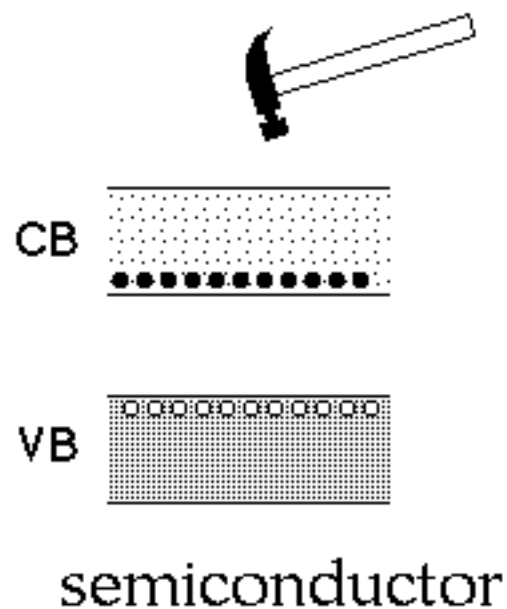
WHAT IS HAPPENING?



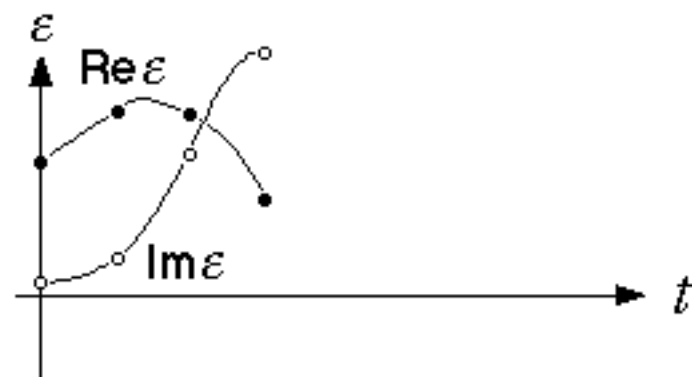
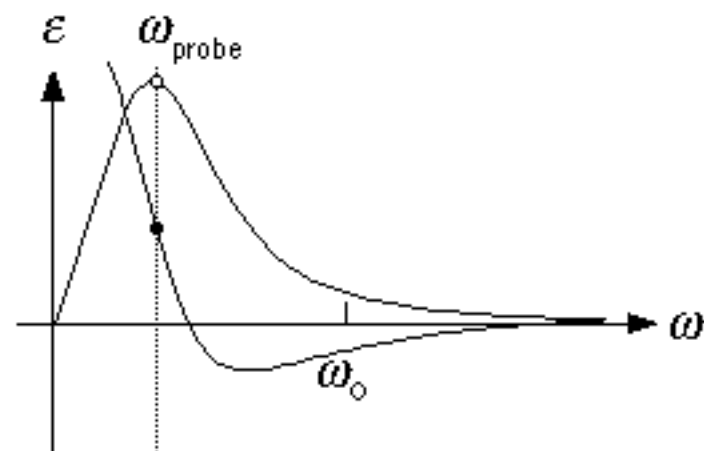
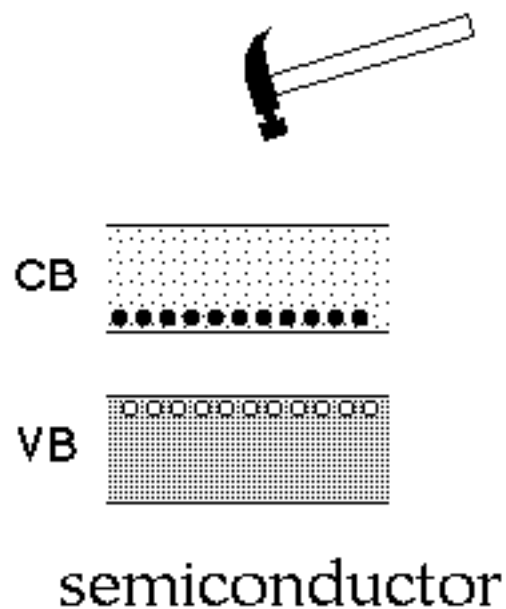
semiconductor



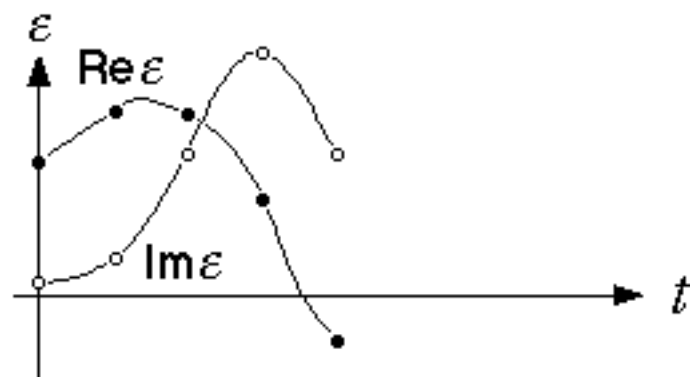
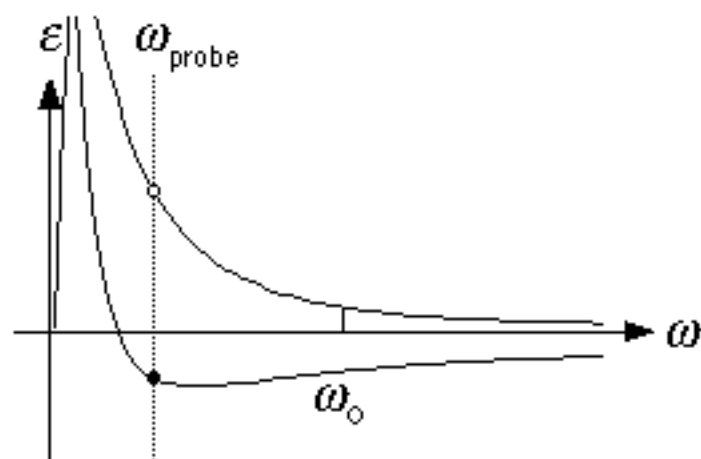
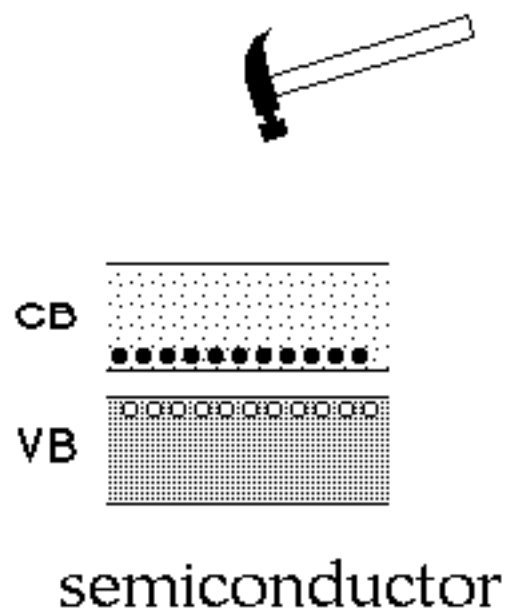
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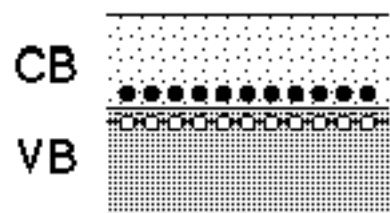
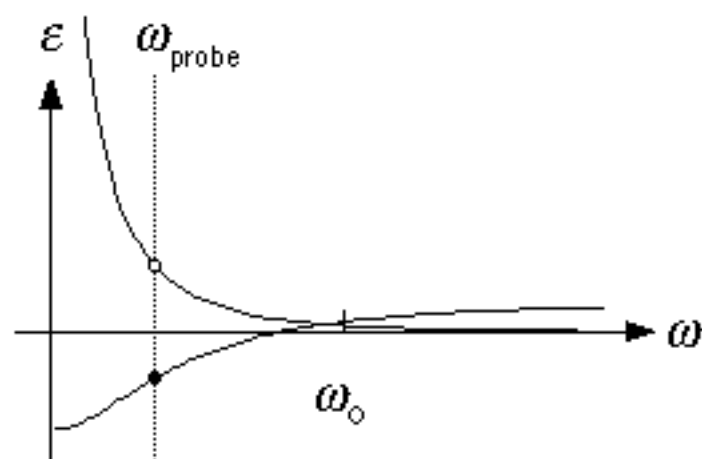
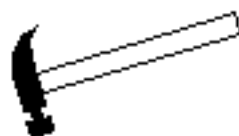
WHAT IS HAPPENING?



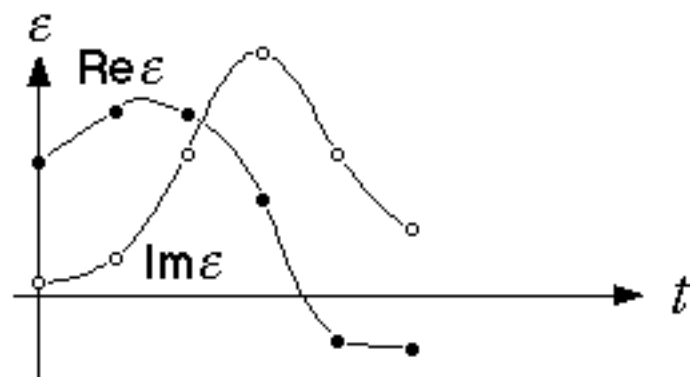
WHAT IS HAPPENING?



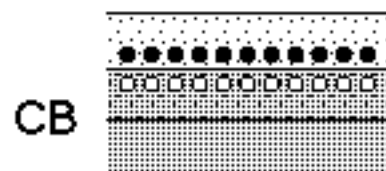
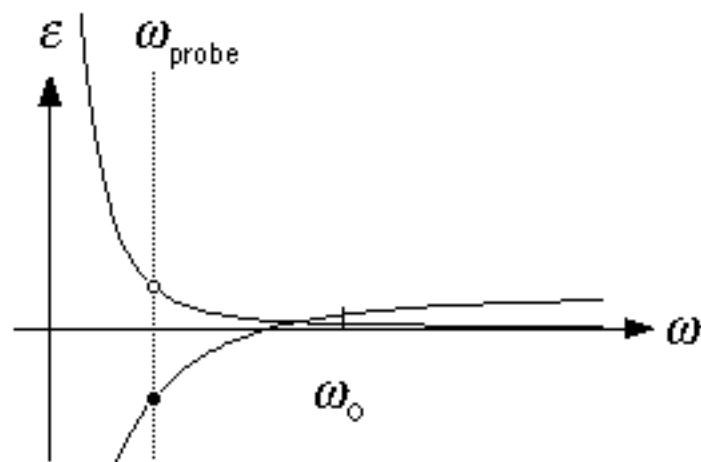
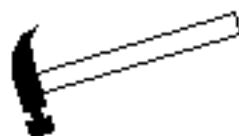
WHAT IS HAPPENING?



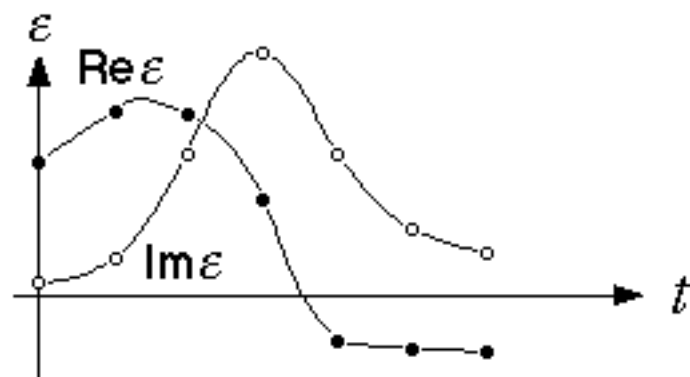
semimetal



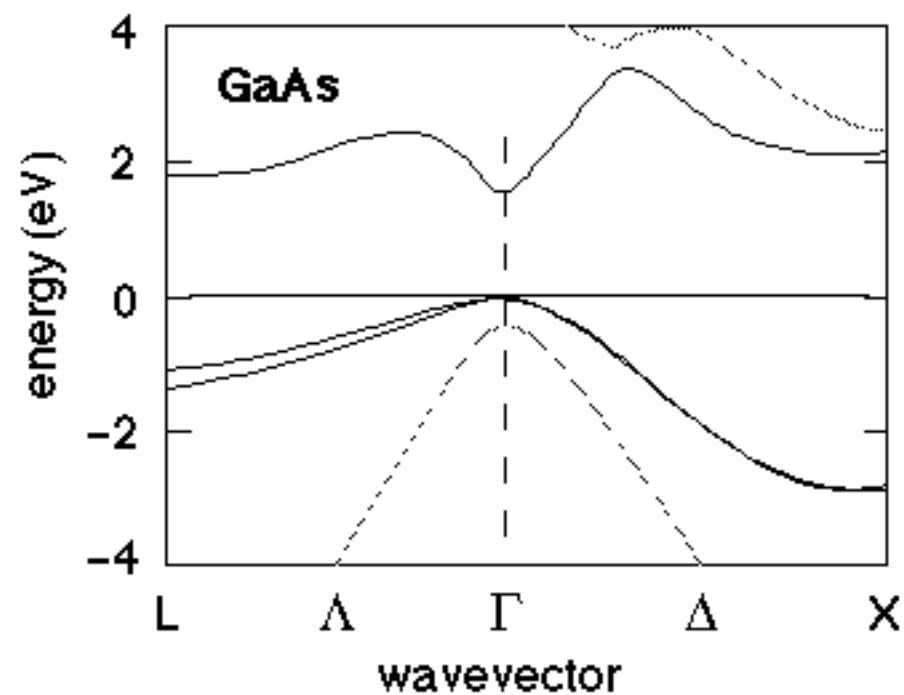
WHAT IS HAPPENING?



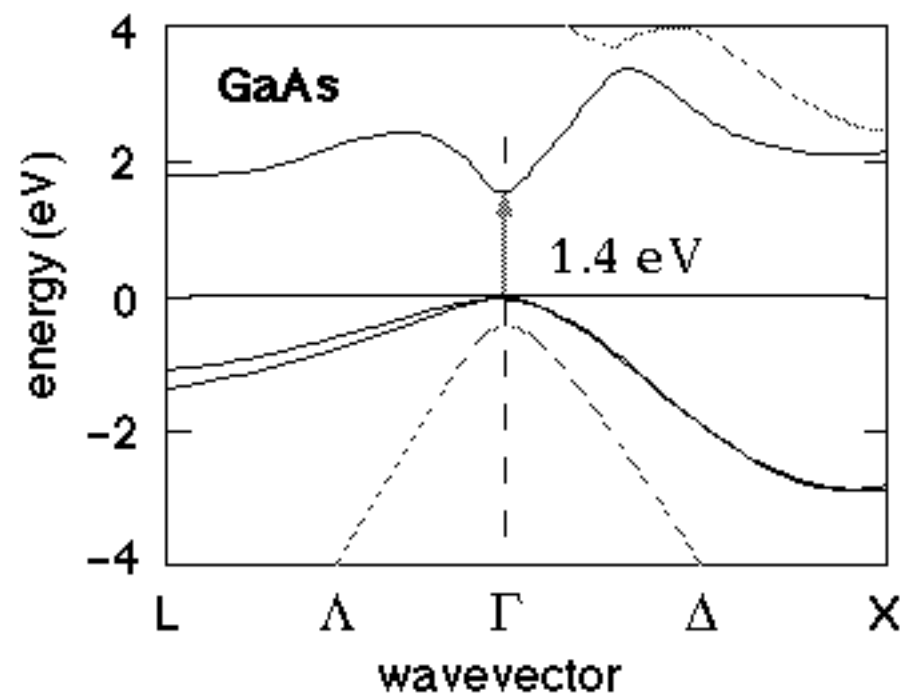
metal



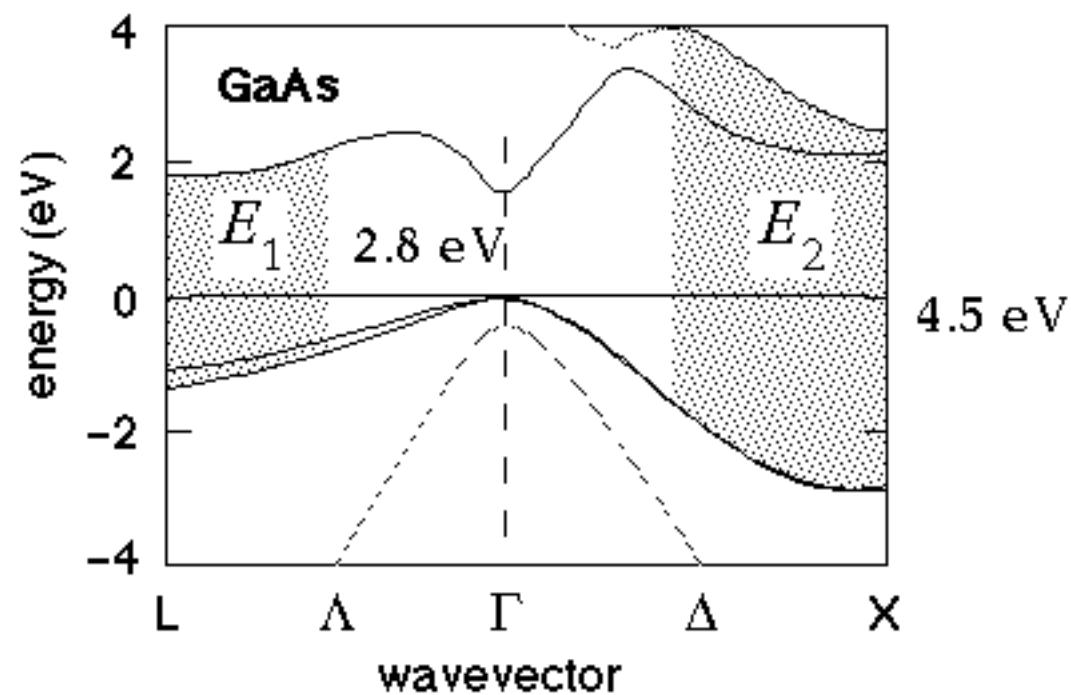
GaAs BANDSTRUCTURE



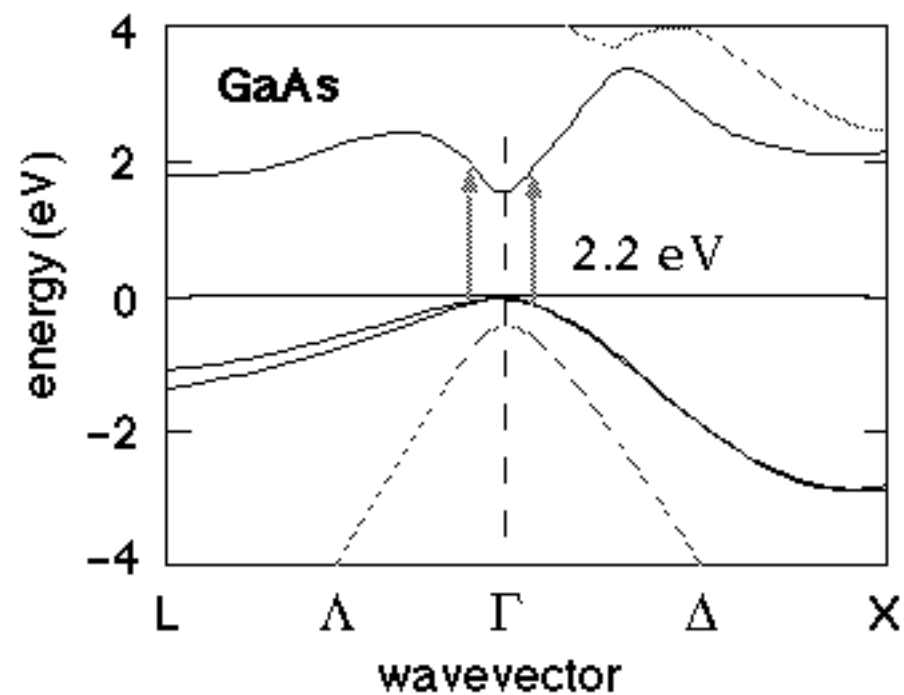
GaAs BANDSTRUCTURE



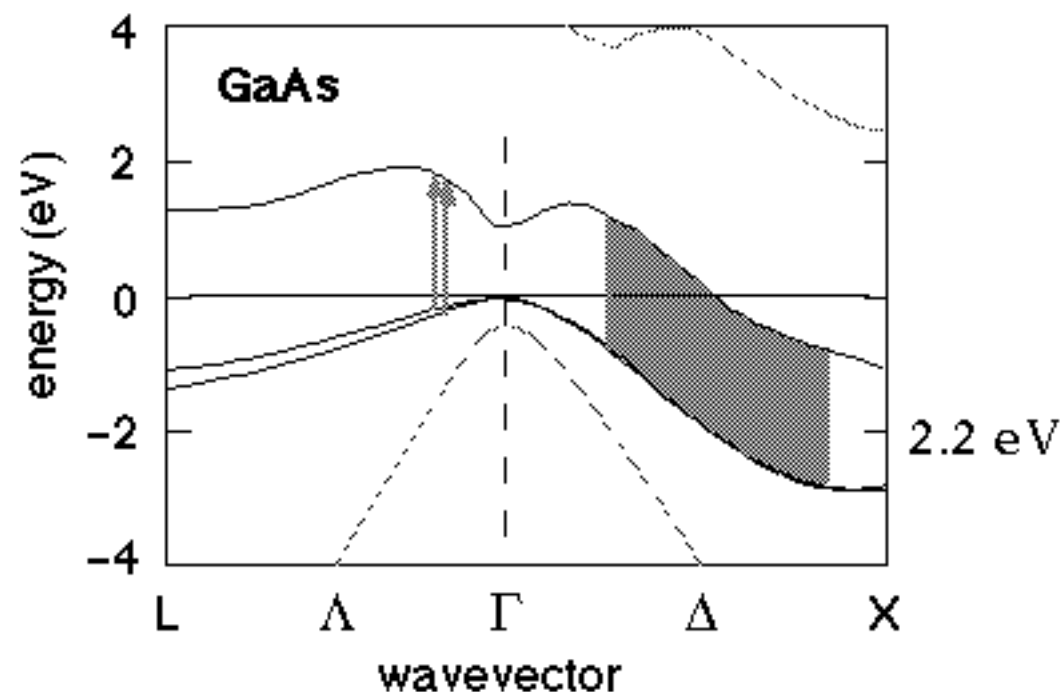
GaAs BANDSTRUCTURE



GaAs BANDSTRUCTURE



GaAs BANDSTRUCTURE

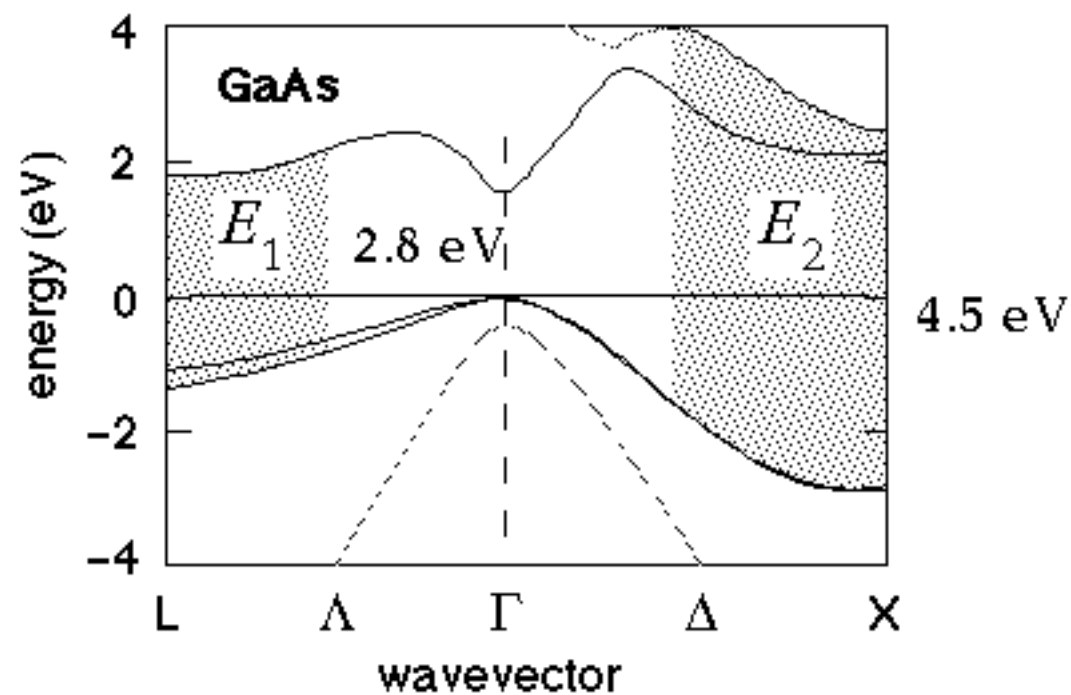


S. Froyen *et al.*, *Phys Rev. B* **28** (1983) 3258

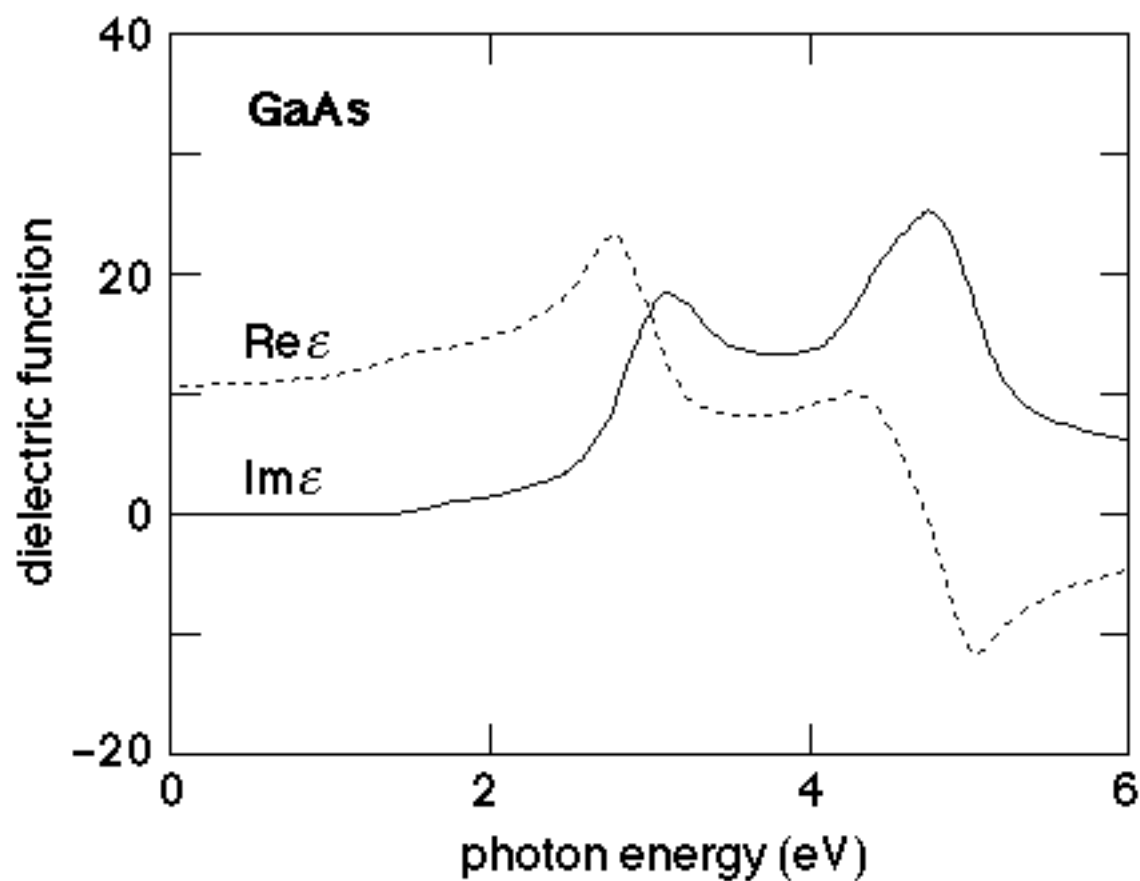
D.H. Kim *et al.*, *Sol. State. Comm.* **89** (1994) 119



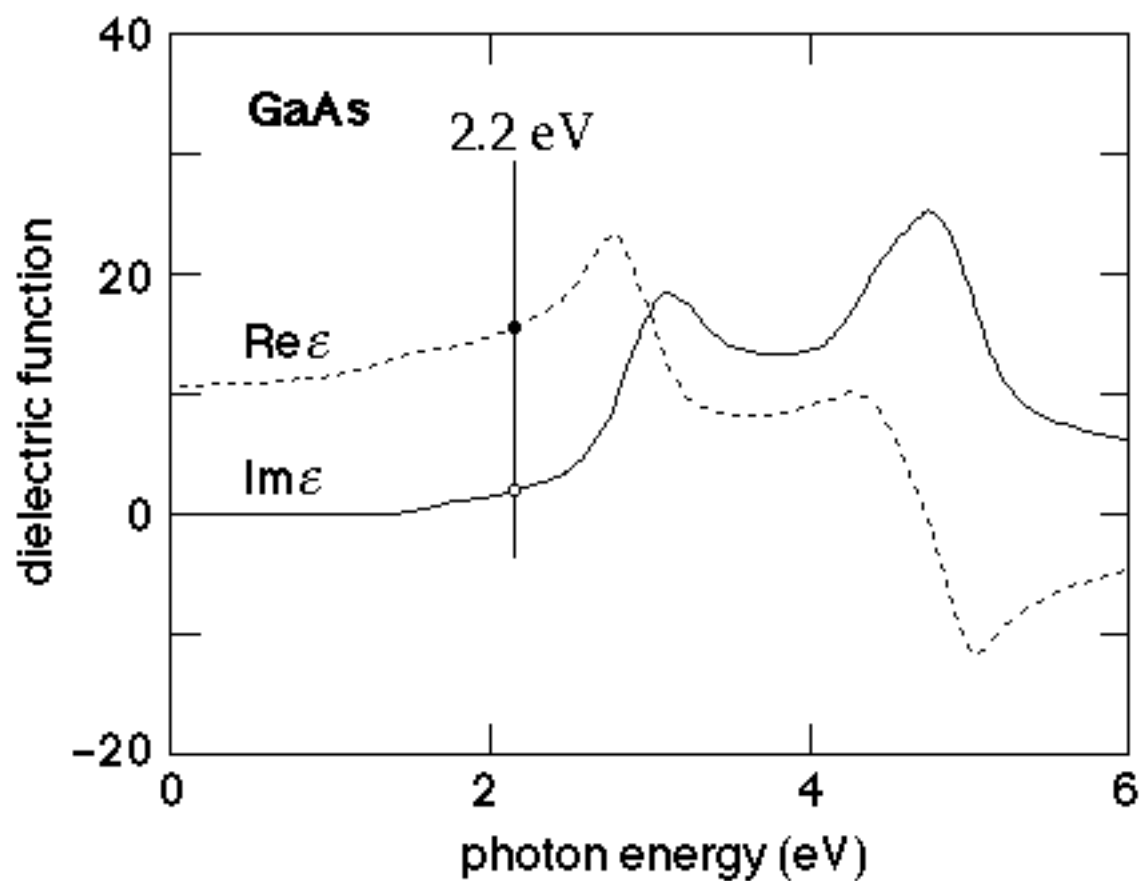
GaAs BANDSTRUCTURE



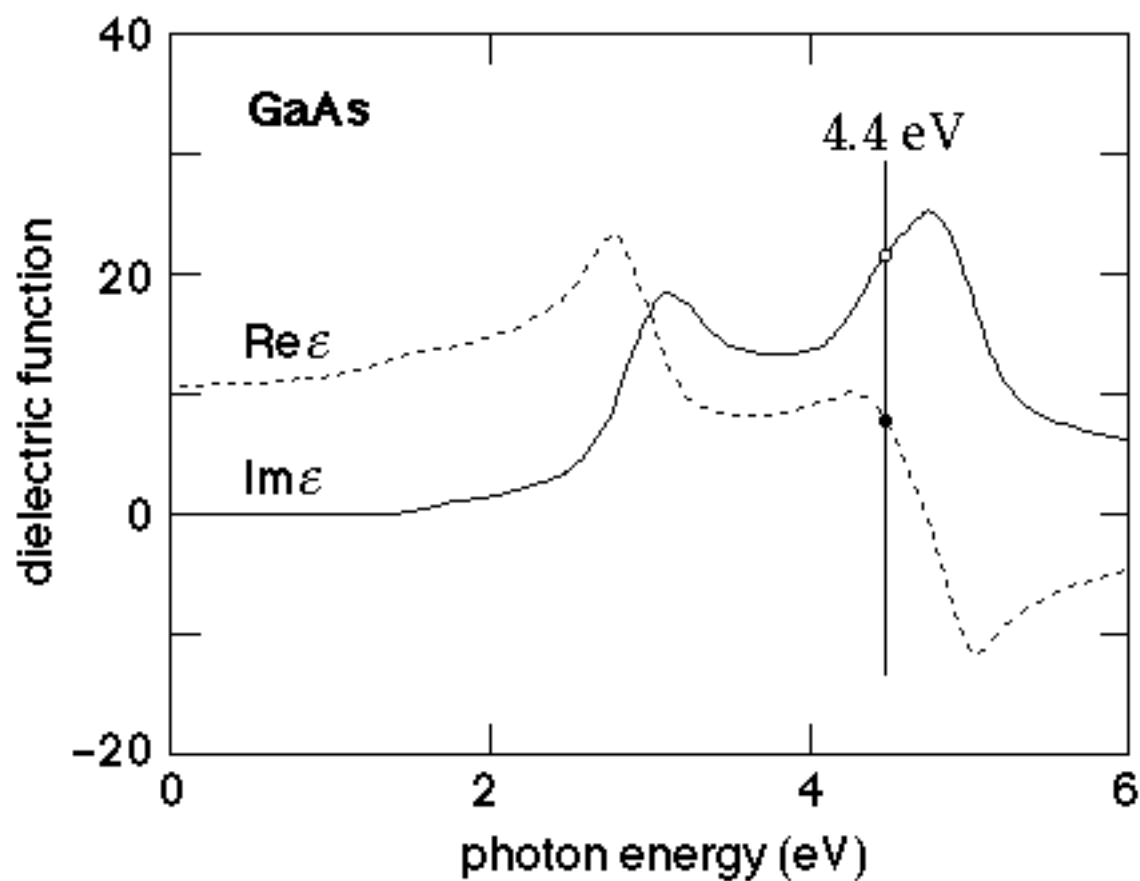
GaAs DIELECTRIC FUNCTION



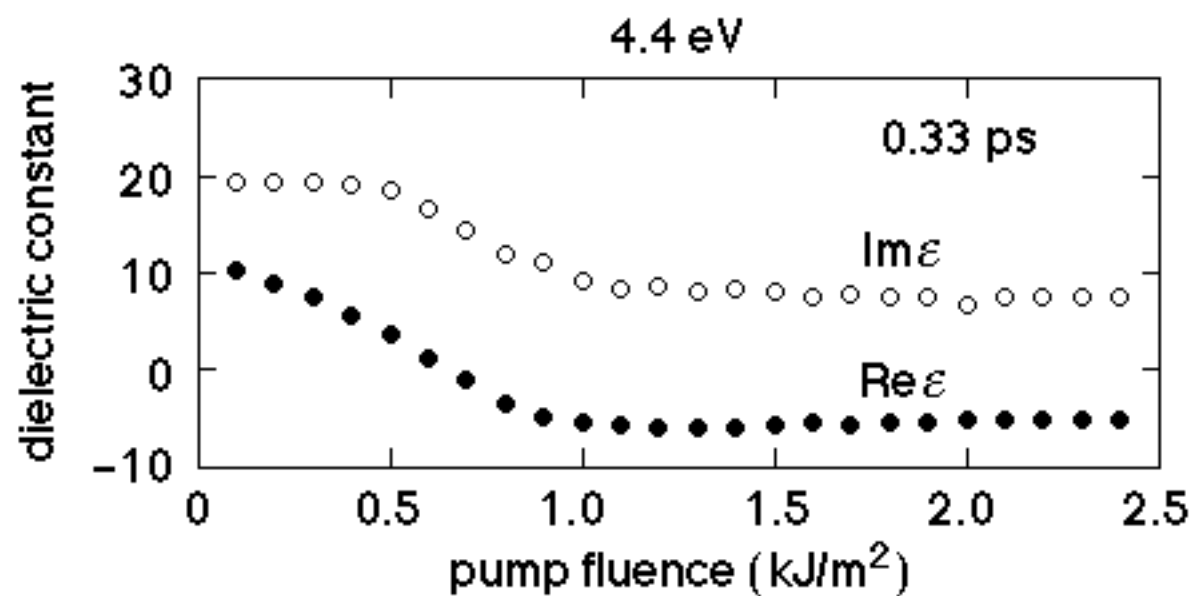
GaAs DIELECTRIC FUNCTION



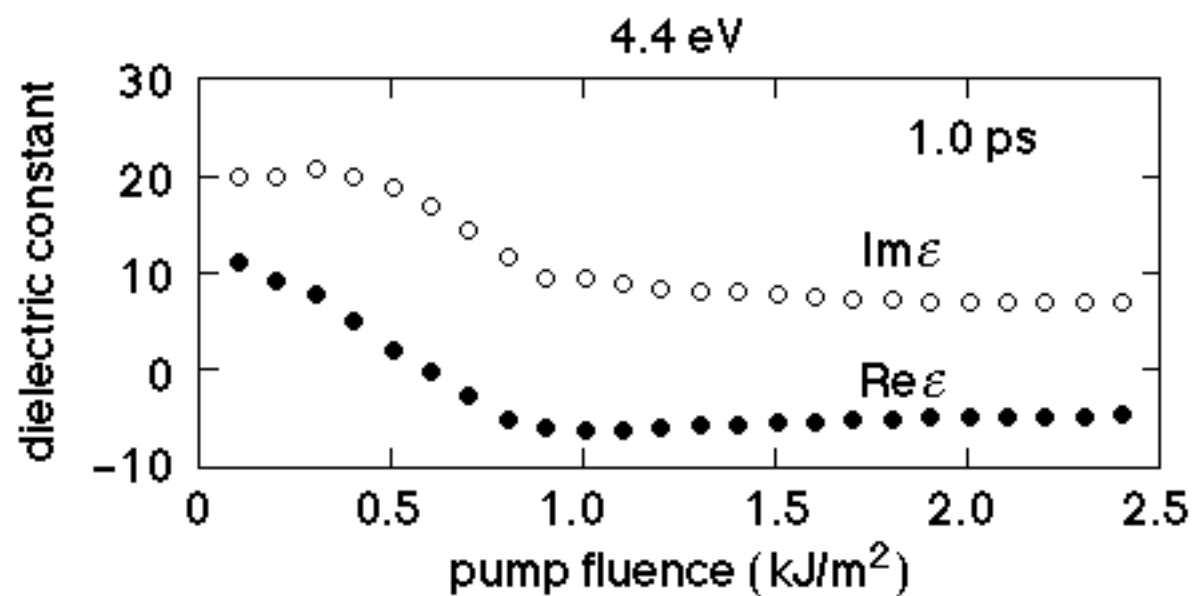
GaAs DIELECTRIC FUNCTION



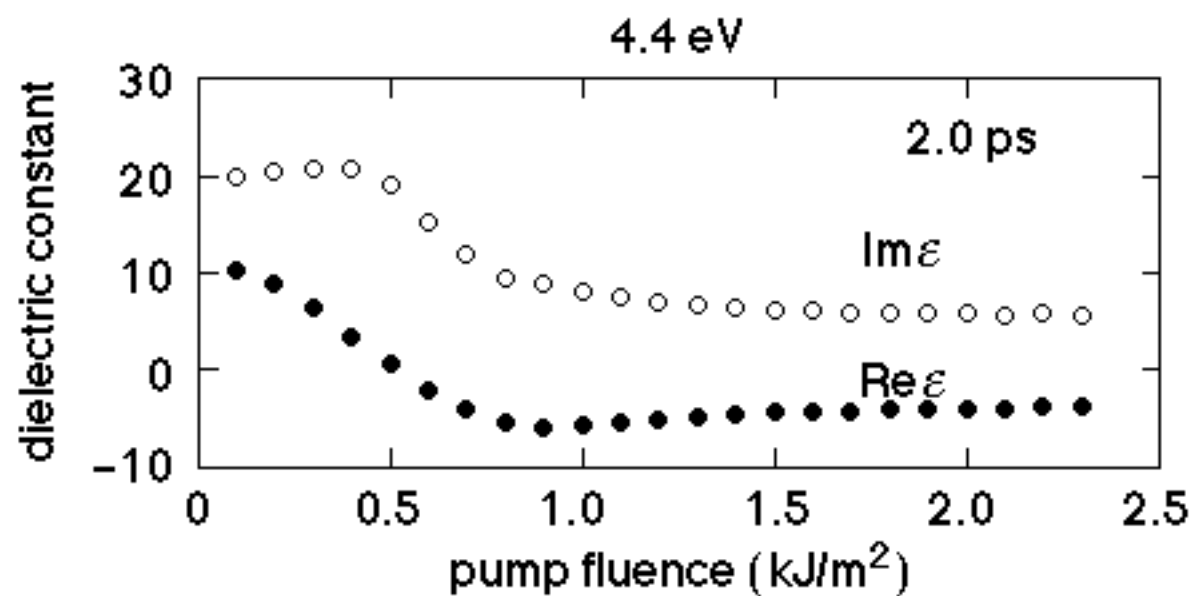
FLUENCE DEPENDENCE



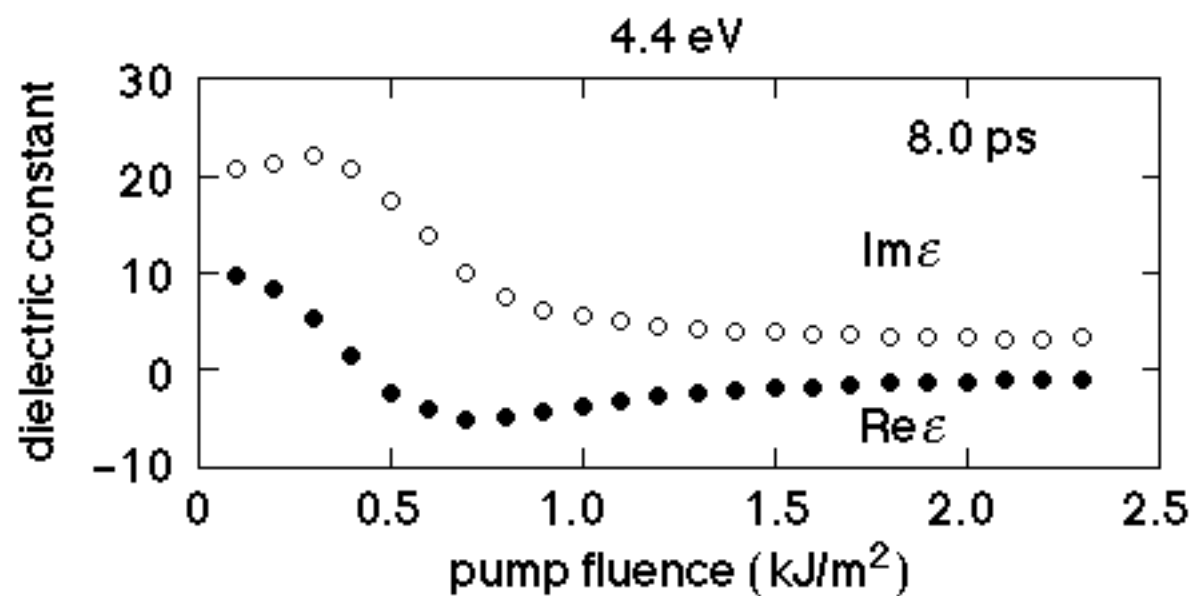
FLUENCE DEPENDENCE



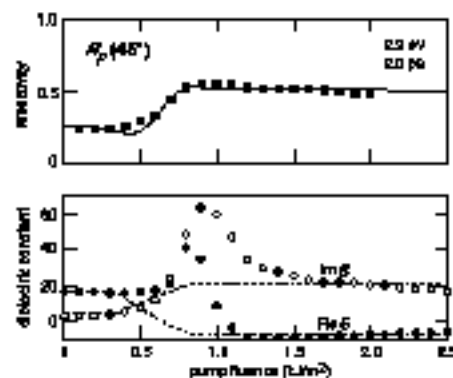
FLUENCE DEPENDENCE



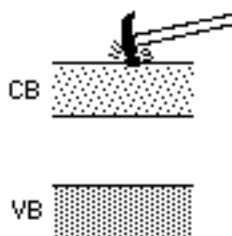
FLUENCE DEPENDENCE



SUMMARY



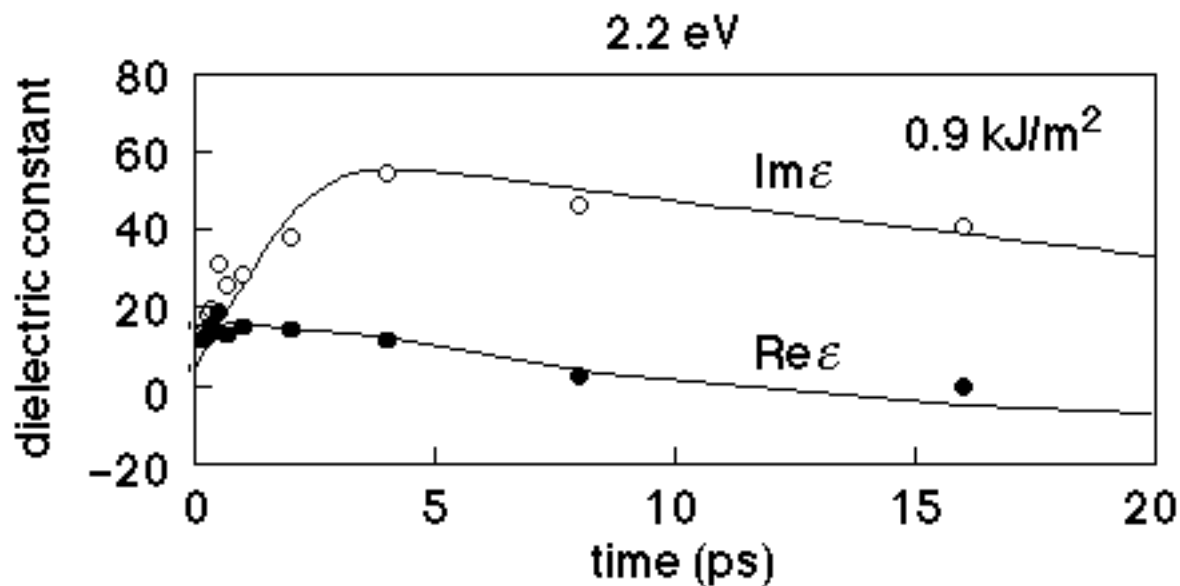
single-angle reflectivity not a good indicator of what happens



laser pulse causes bandgap collapse



CAUSE OF COLLAPSE



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

structural change?



Field strength at second harmonic:

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



FEMTOSECOND WORK

Field strength at second harmonic:

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

where:

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

so:



FEMTOSECOND WORK

Field strength at second harmonic:

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

where:

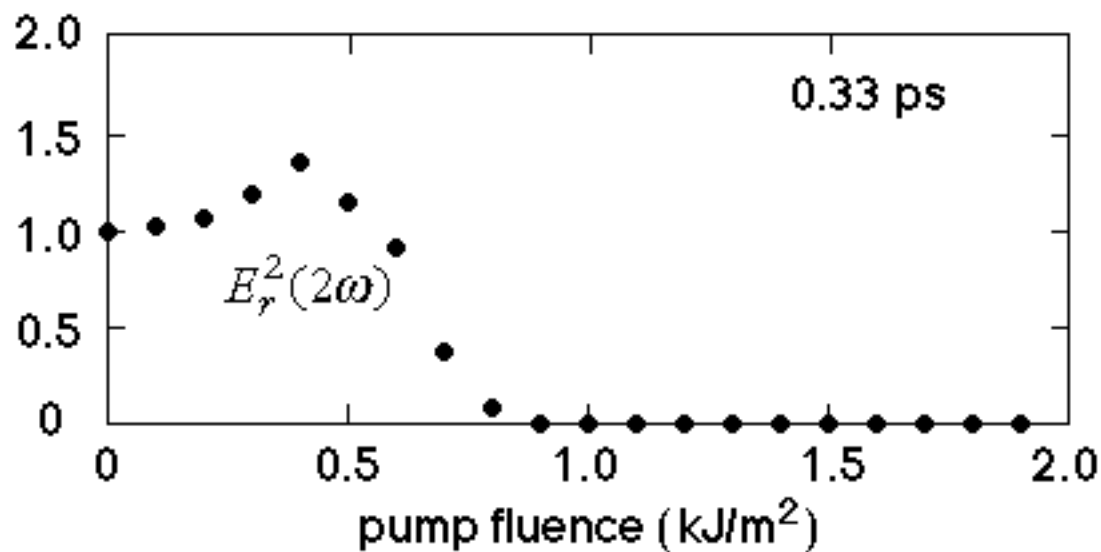
$$P^{(2)}(2\omega) = 2\chi^{(2)} E_i^2(\omega) f[\epsilon(\omega), \theta_i]$$

so:

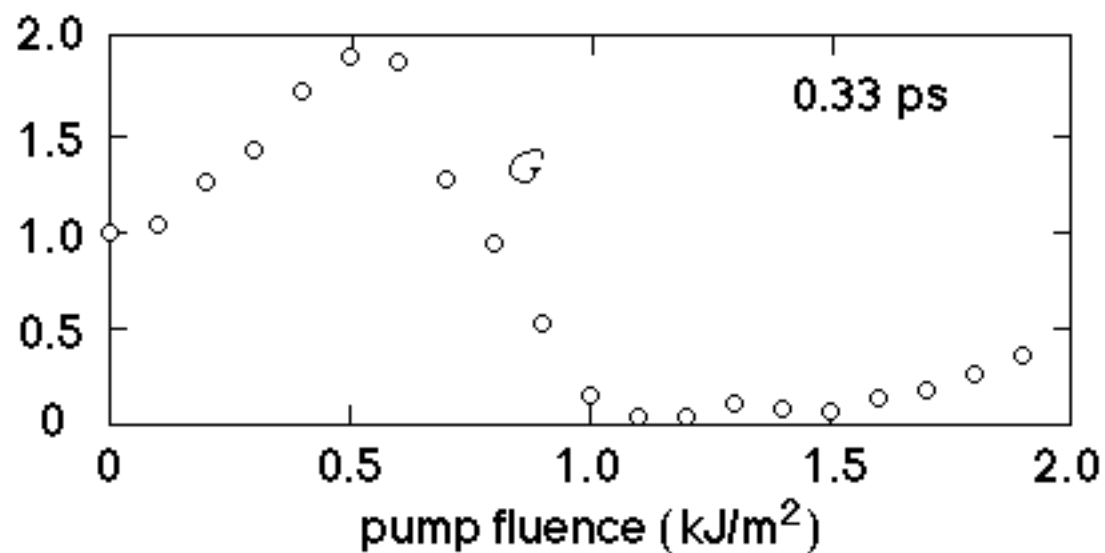
$$|\chi^{(2)}|^2 = \frac{E_r^2(2\omega)}{E_i^2(\omega) G[\epsilon(\omega), \epsilon(2\omega), \theta_i]}$$



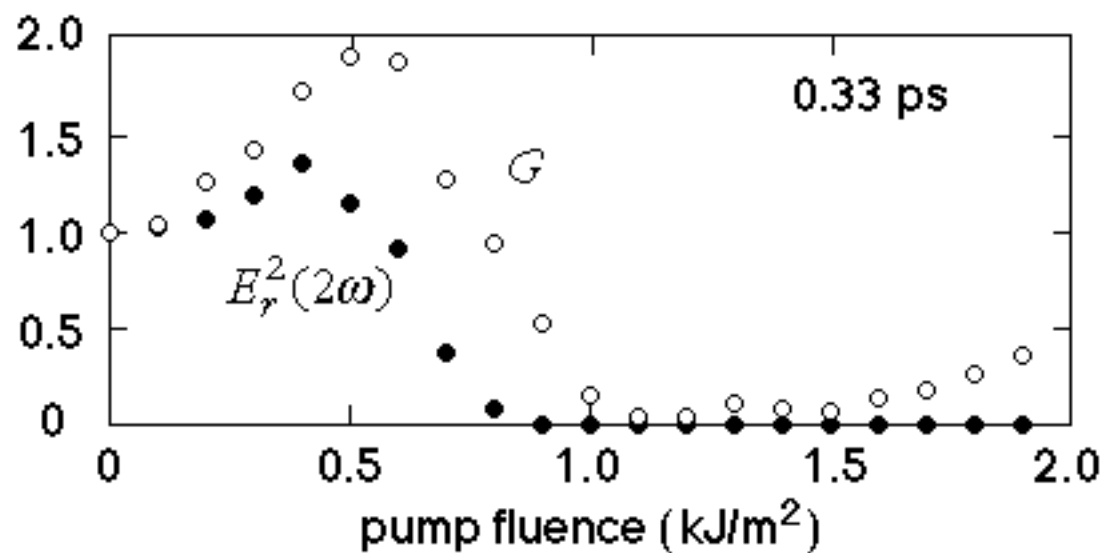
SECOND HARMONIC DATA



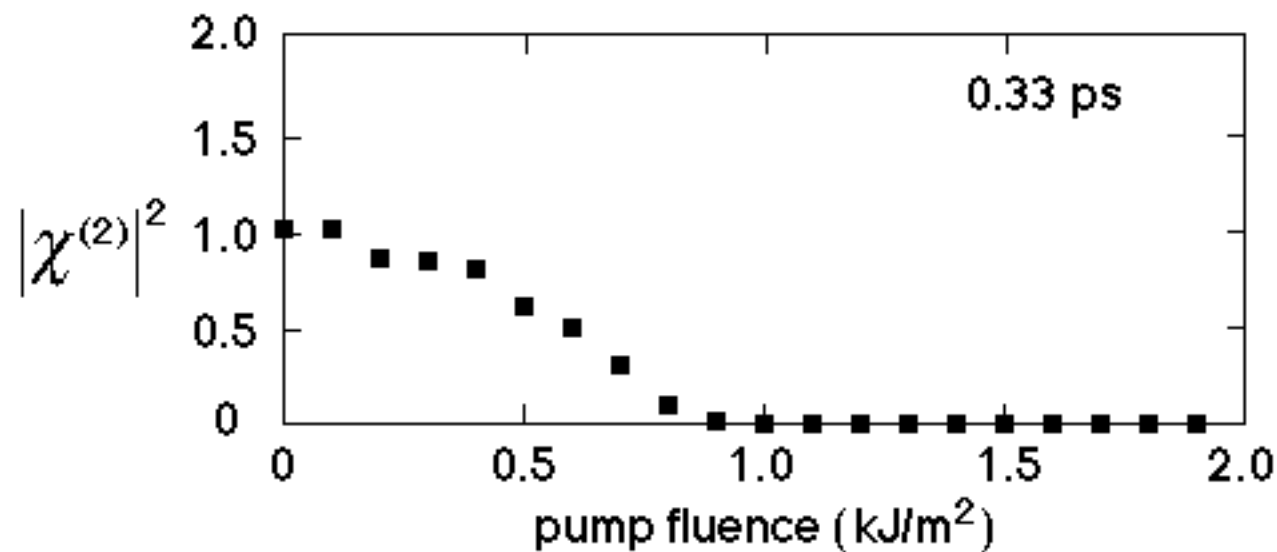
SECOND HARMONIC DATA



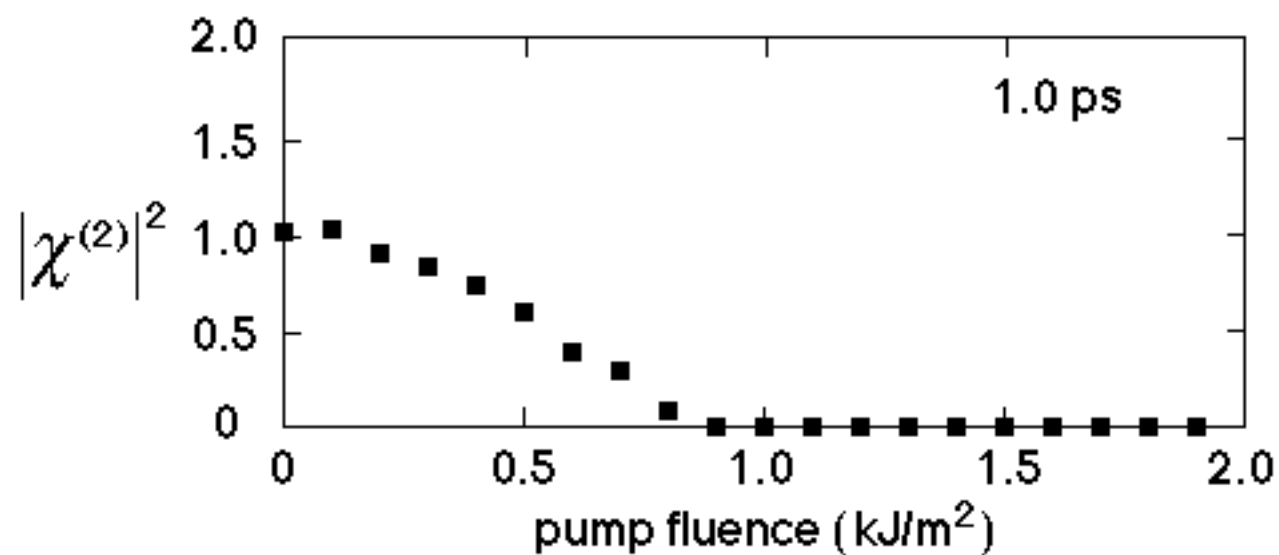
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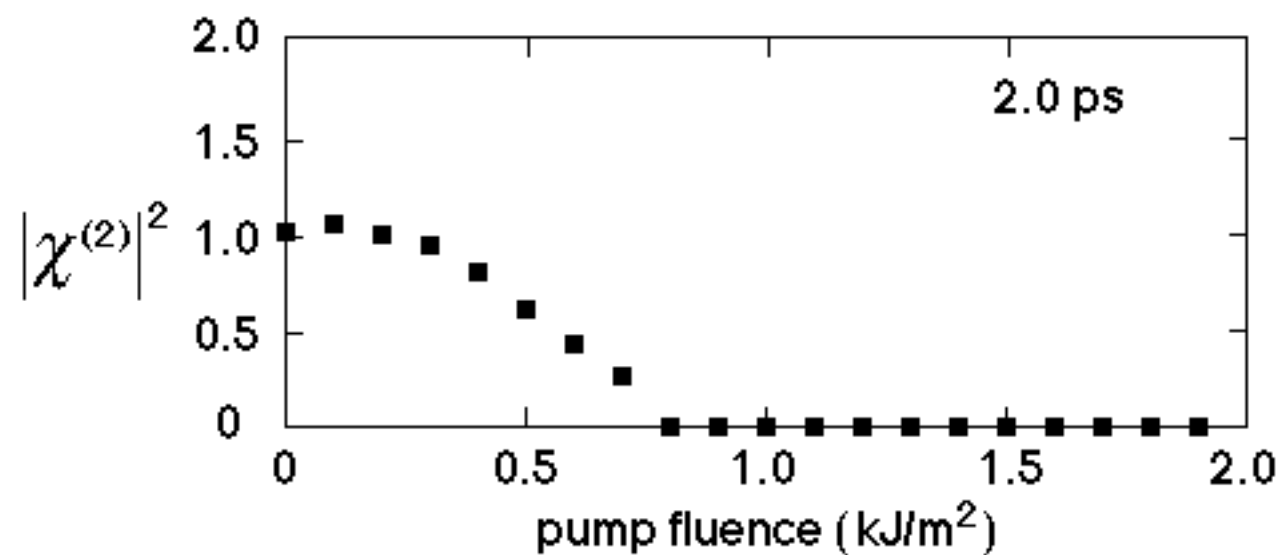
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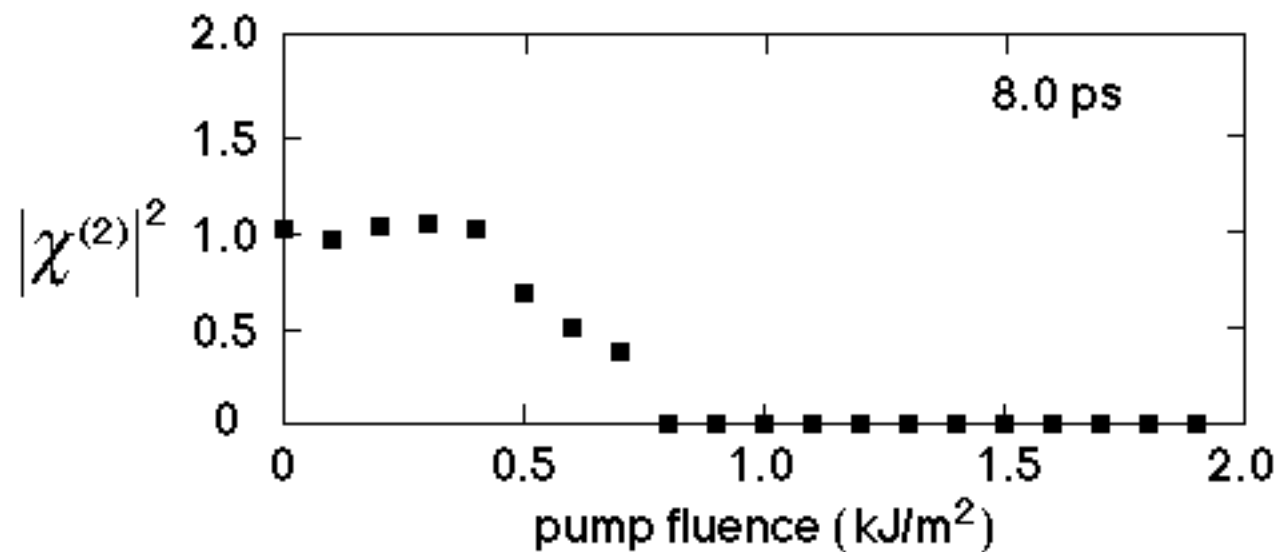
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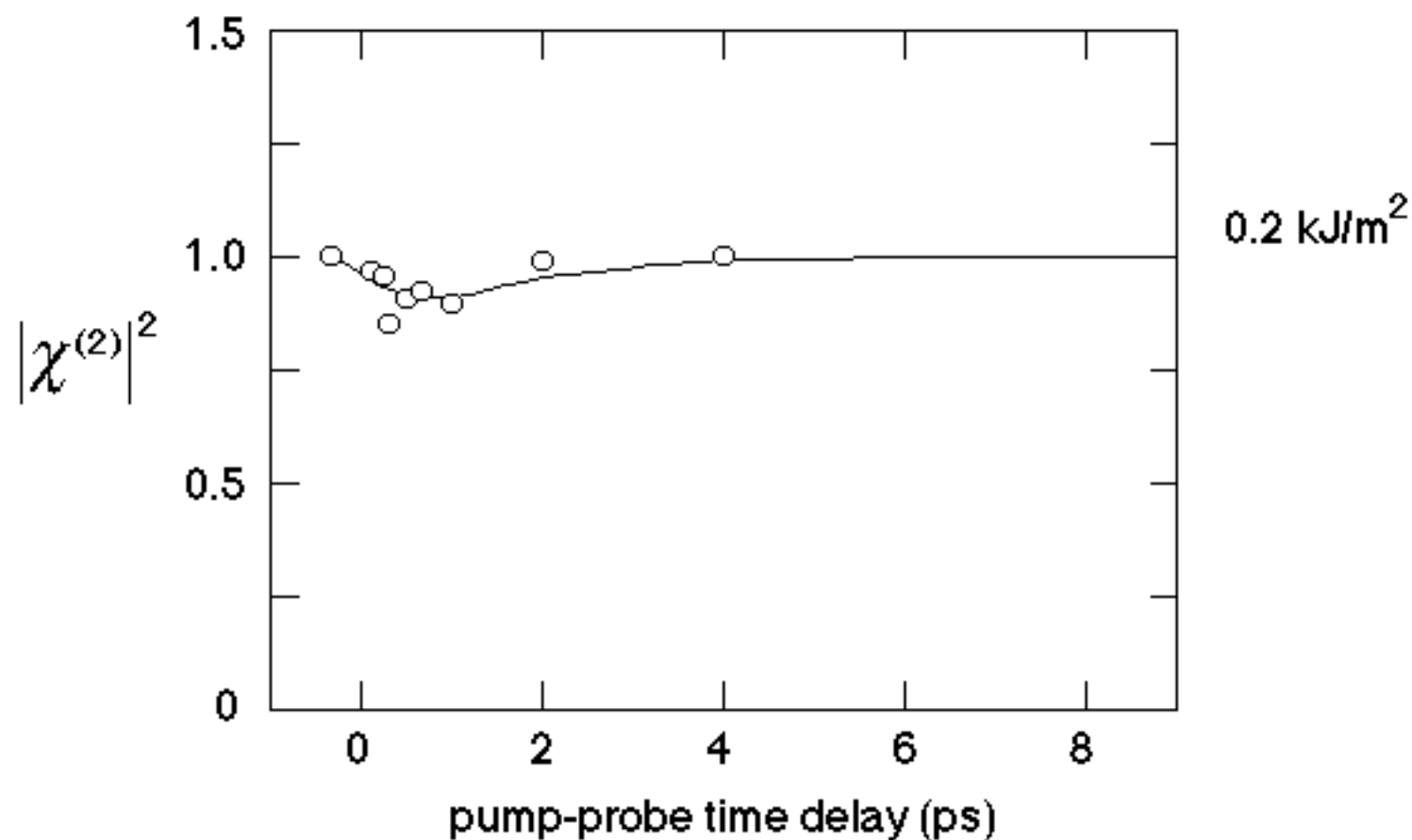
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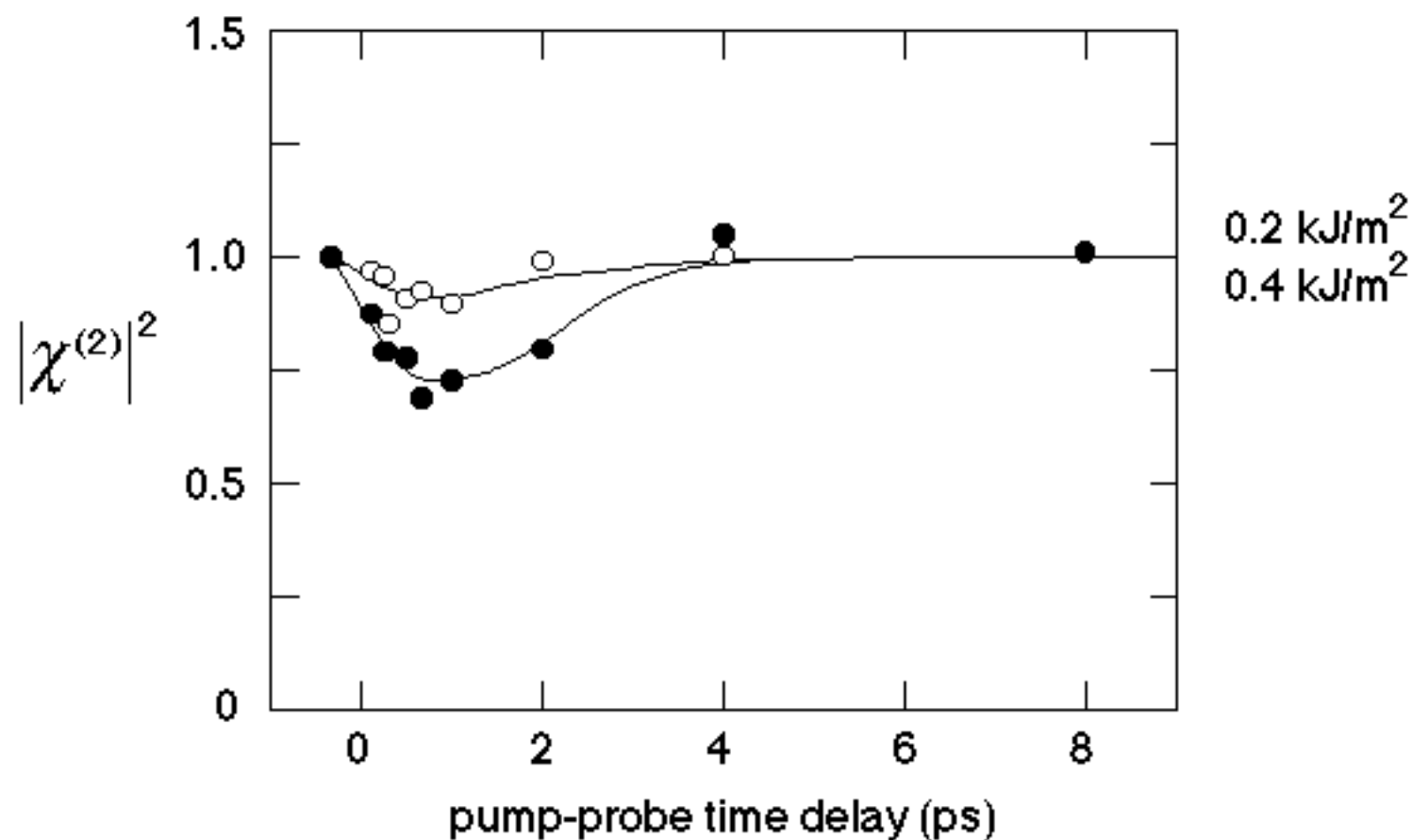
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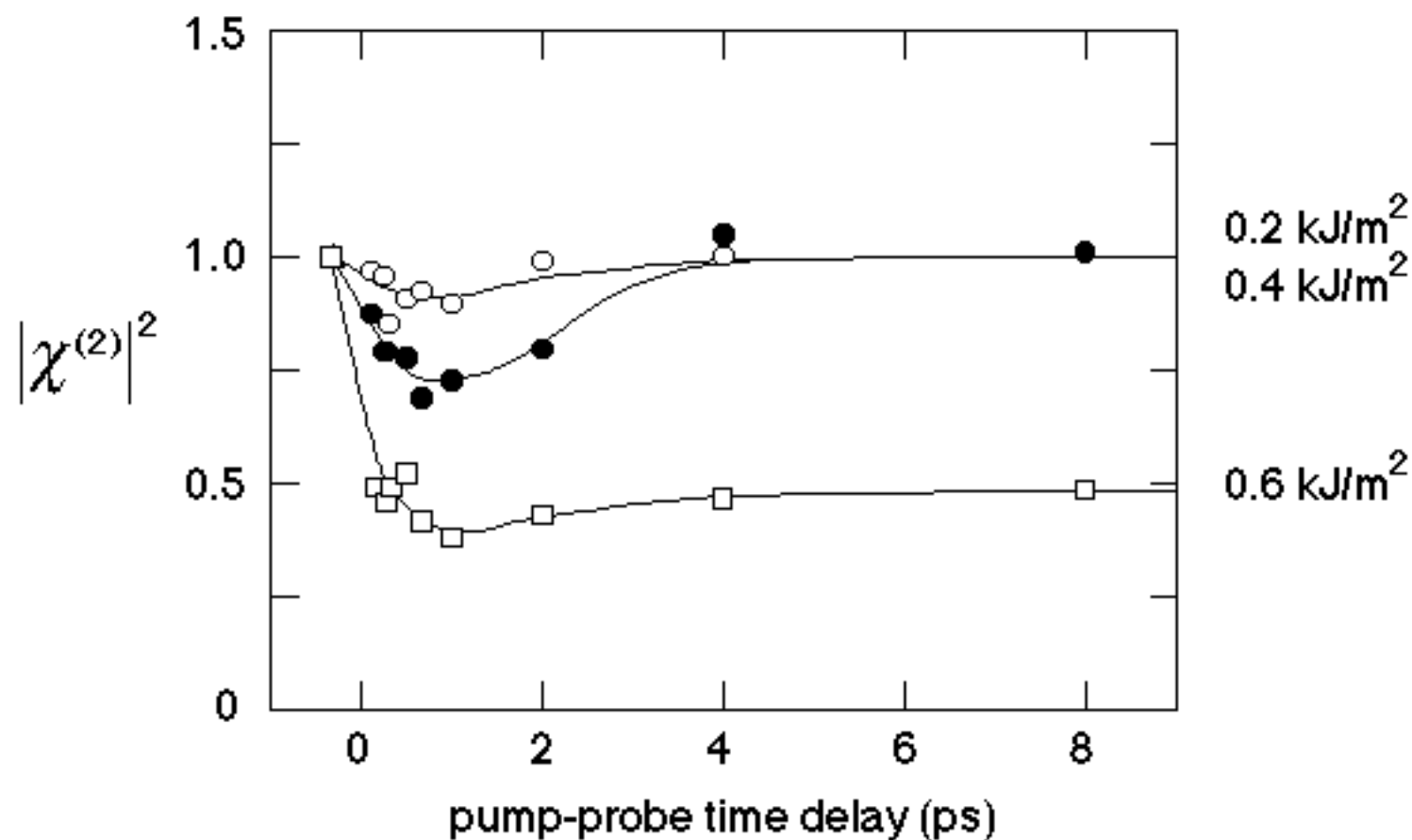
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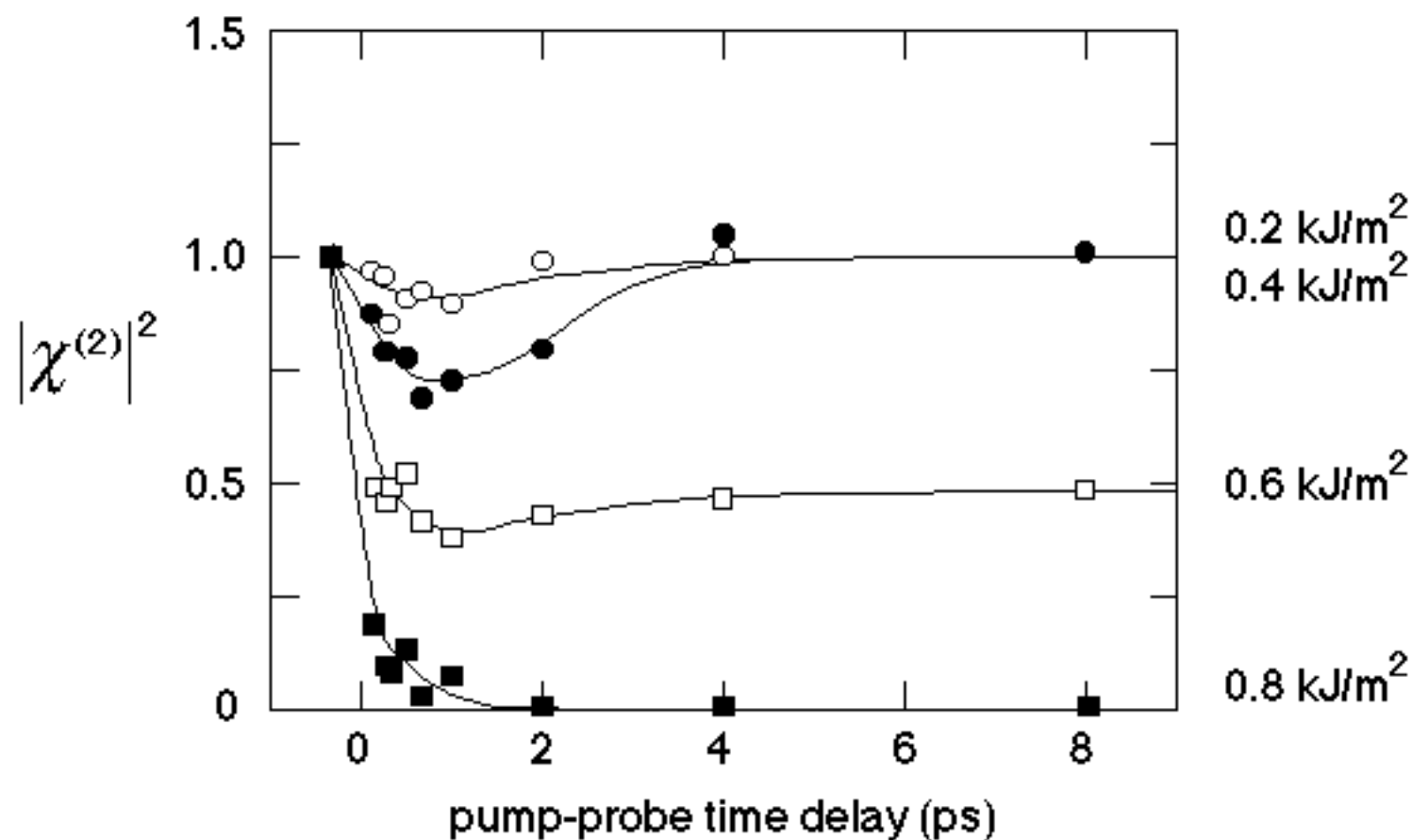
SECOND HARMONIC DATA



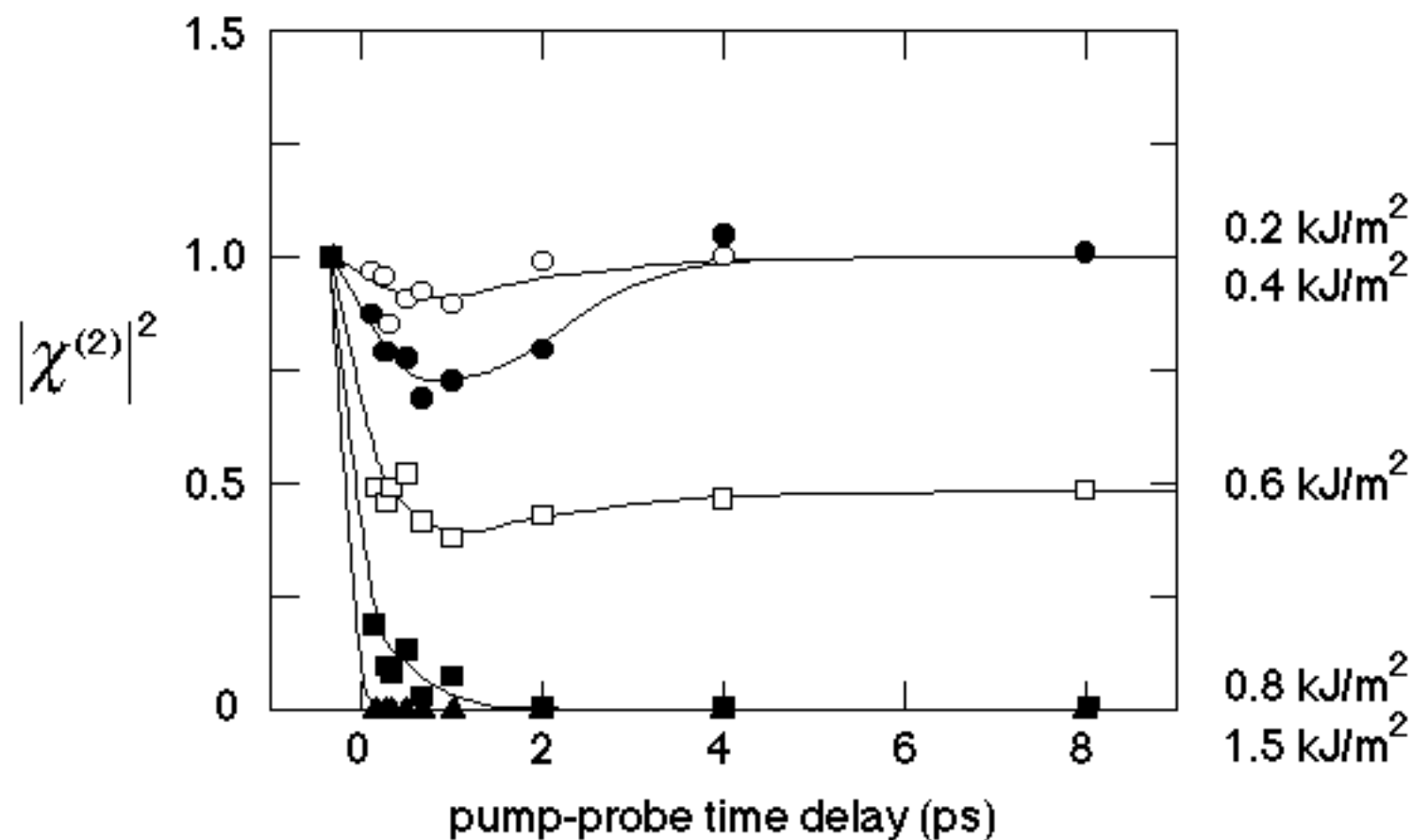
SECOND HARMONIC DATA



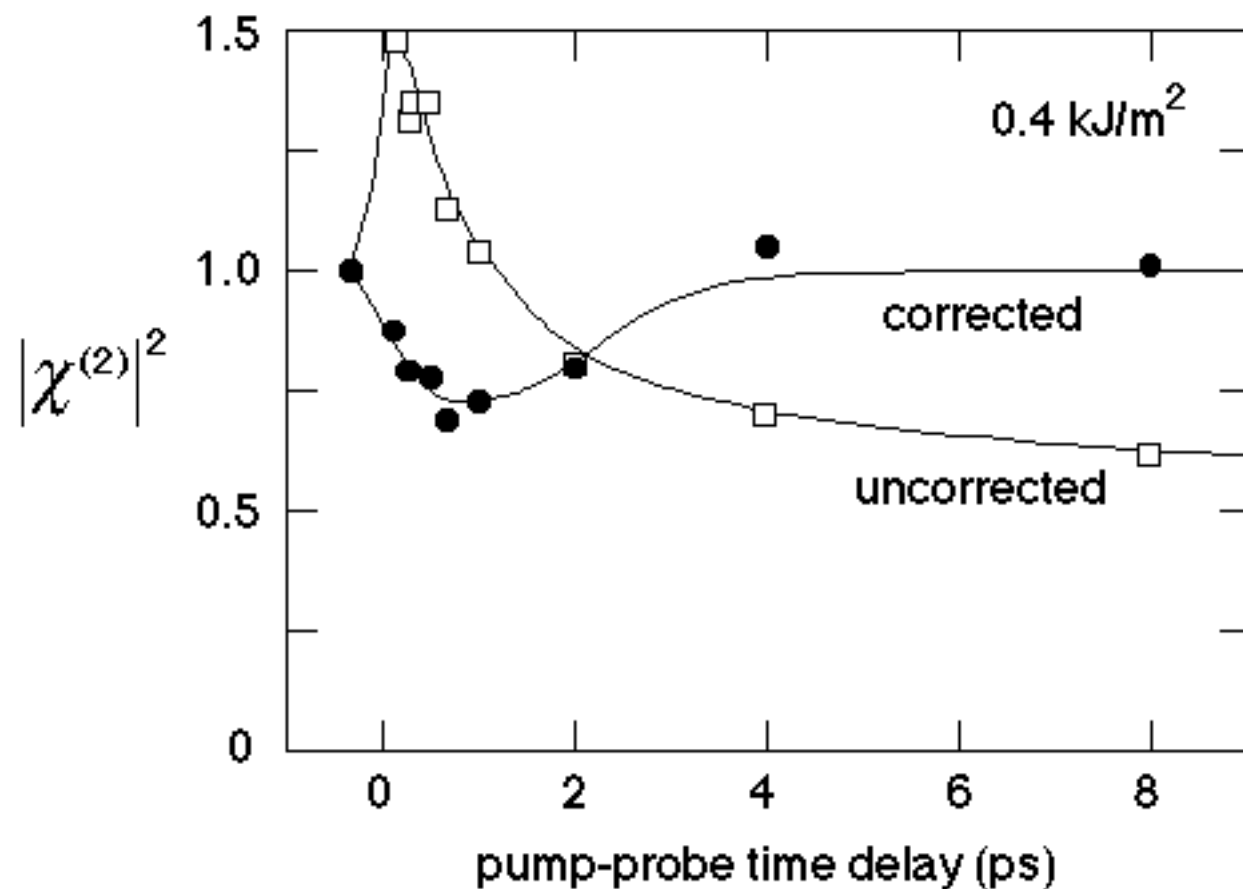
SECOND HARMONIC DATA



SECOND HARMONIC DATA



EFFECT OF CHANGES IN ϵ



FEMTOSECOND WORK

$\chi^{(2)}$ sensitive to loss of long-range order

long-range order disappears even below threshold

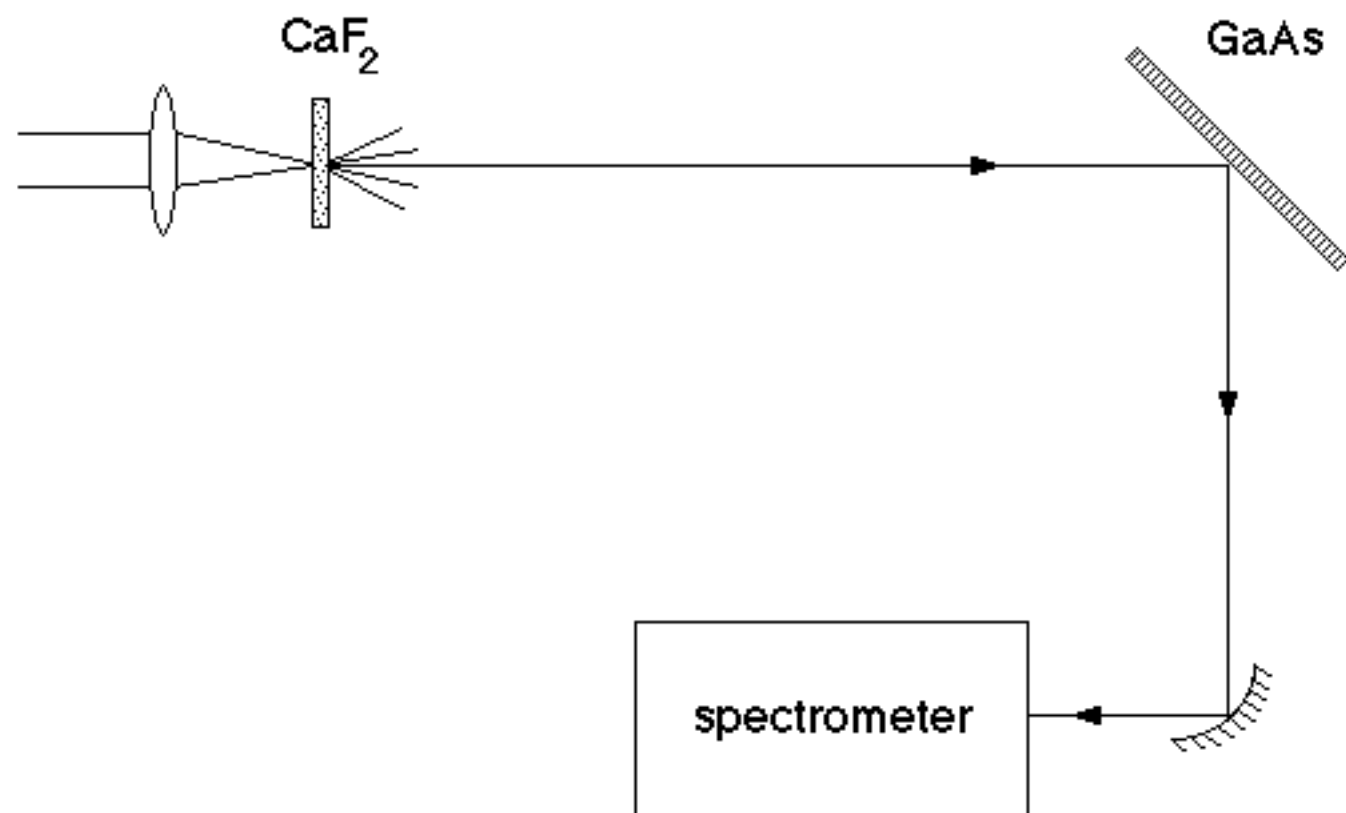
structural change due to destabilization of covalent bonds



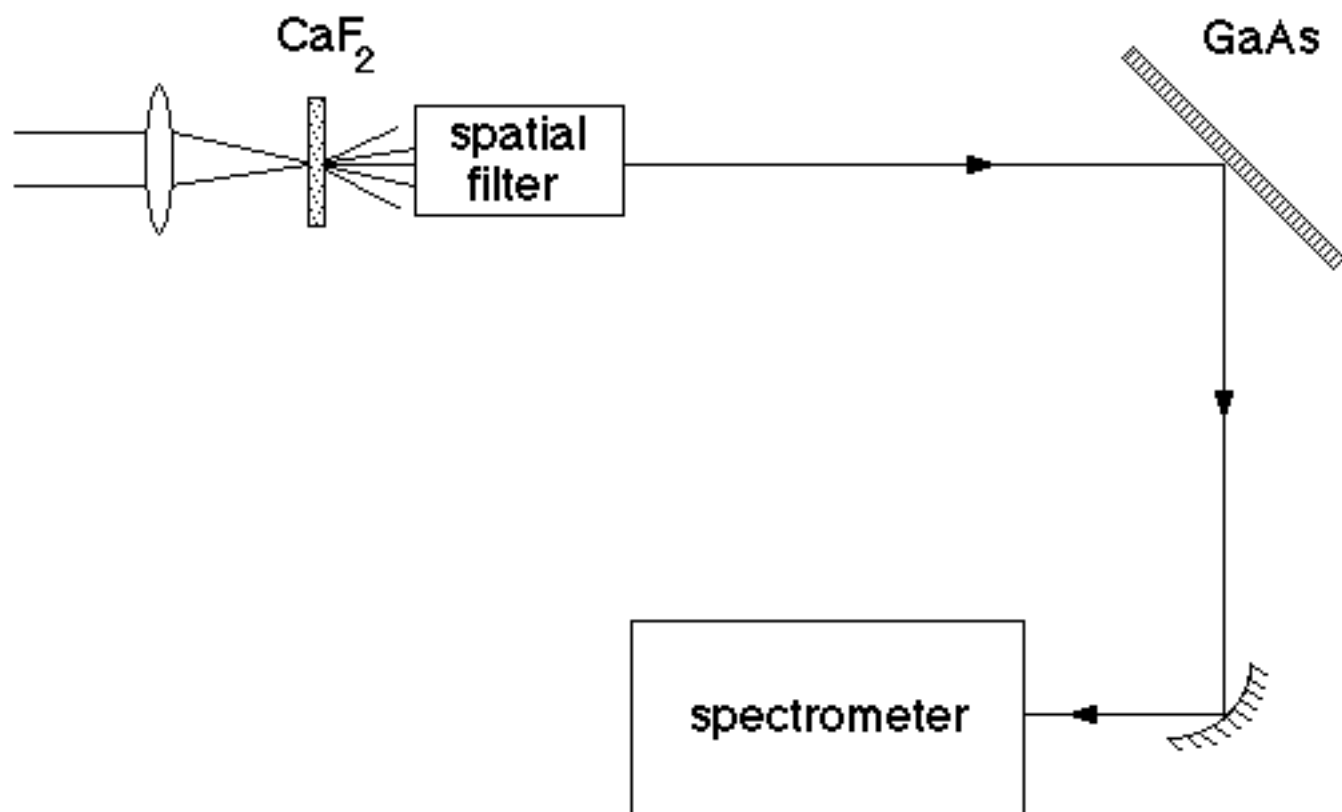
- ① Background
- ② Femtosecond work
- ③ Future



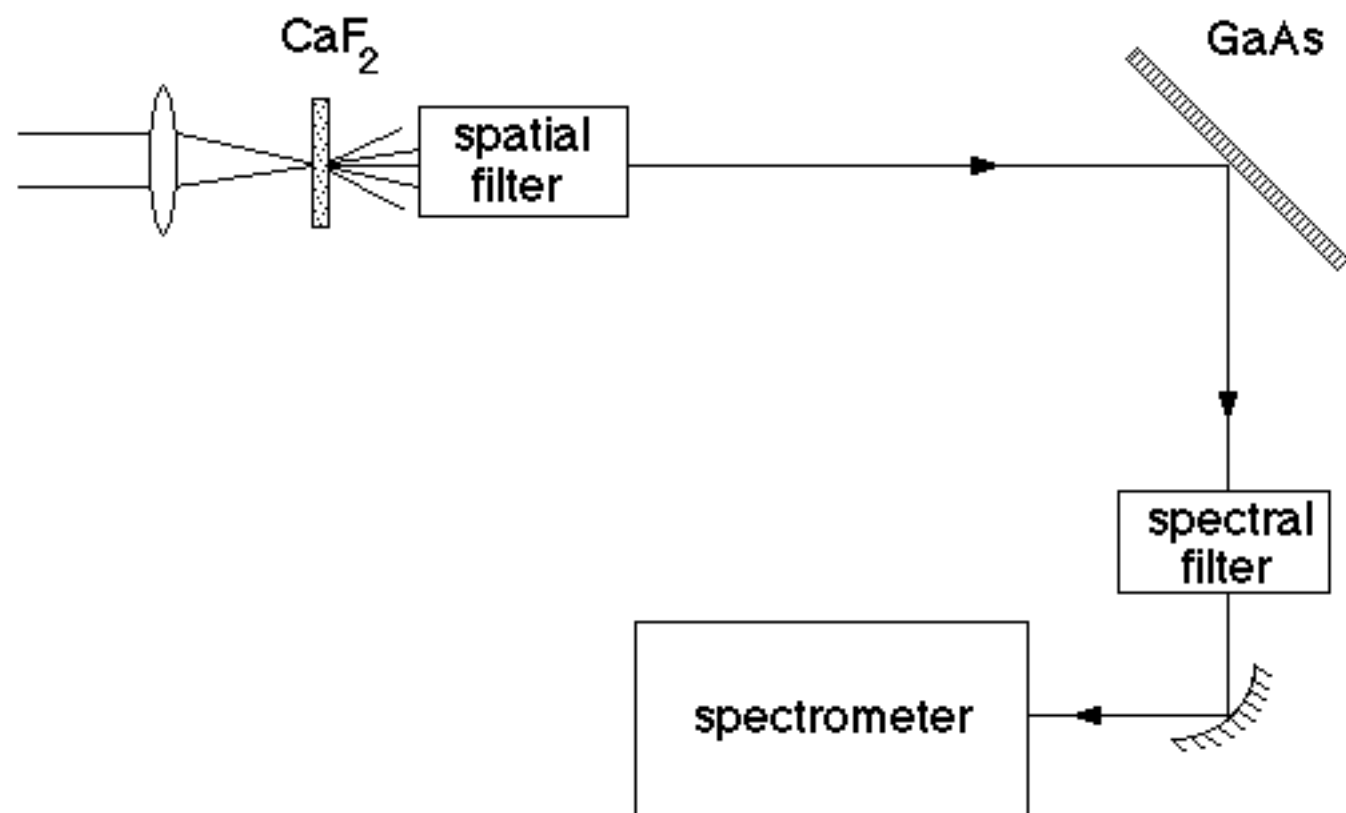
BROADBAND PROBE



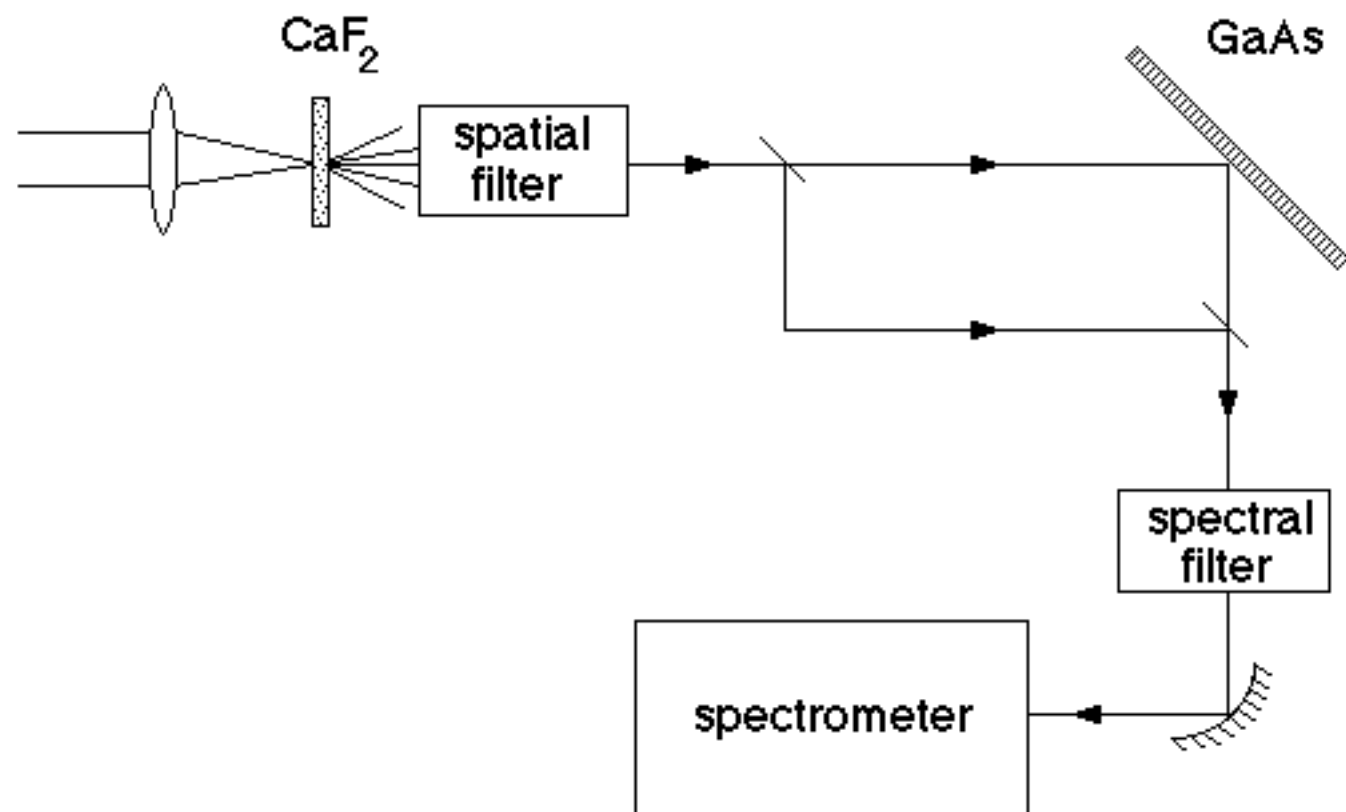
BROADBAND PROBE



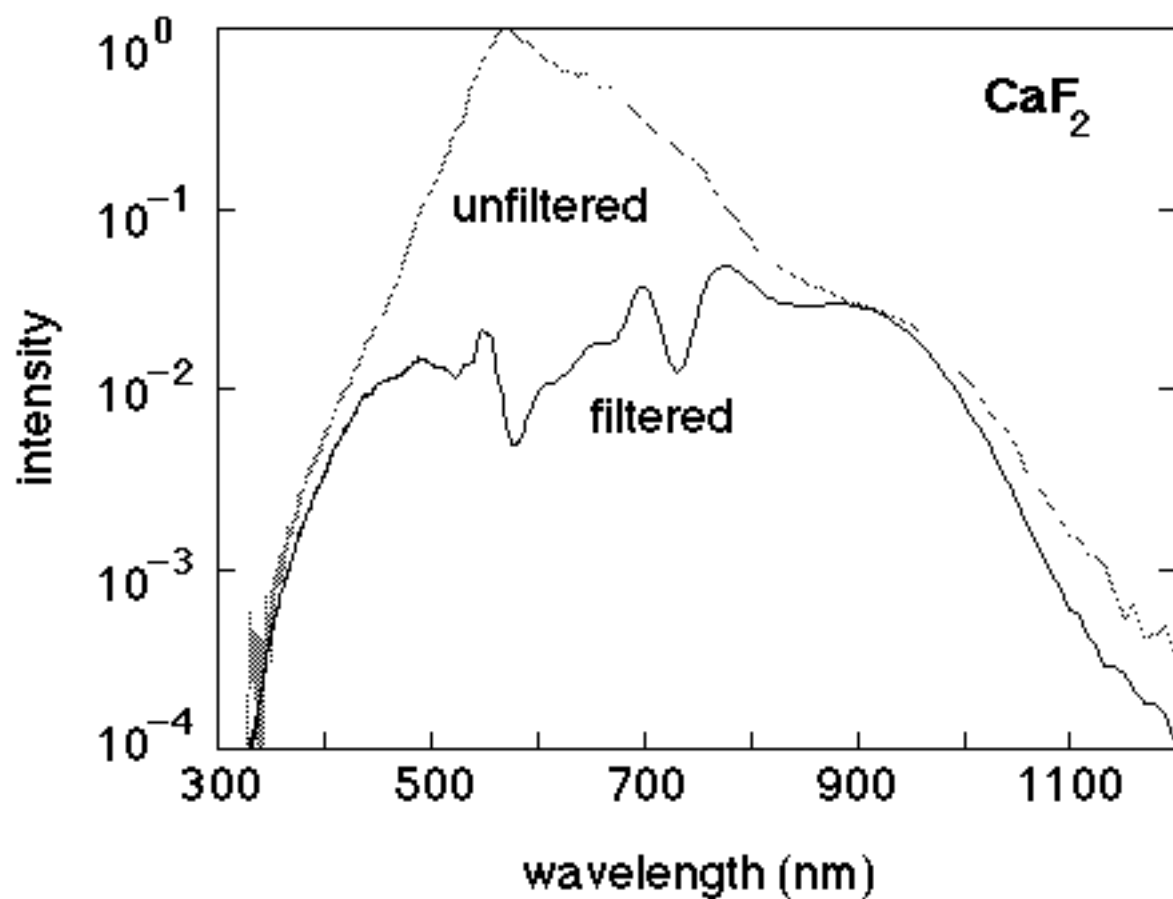
BROADBAND PROBE



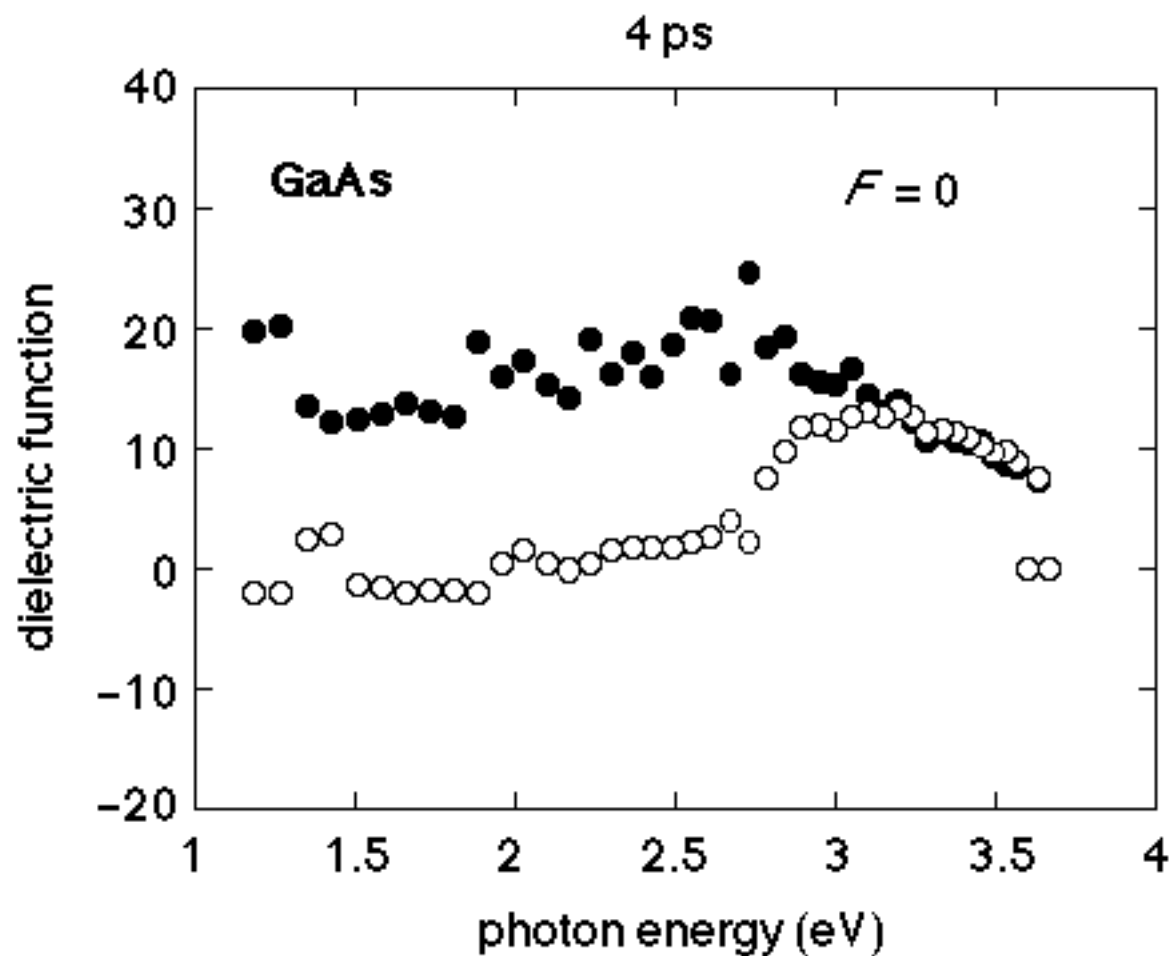
BROADBAND PROBE



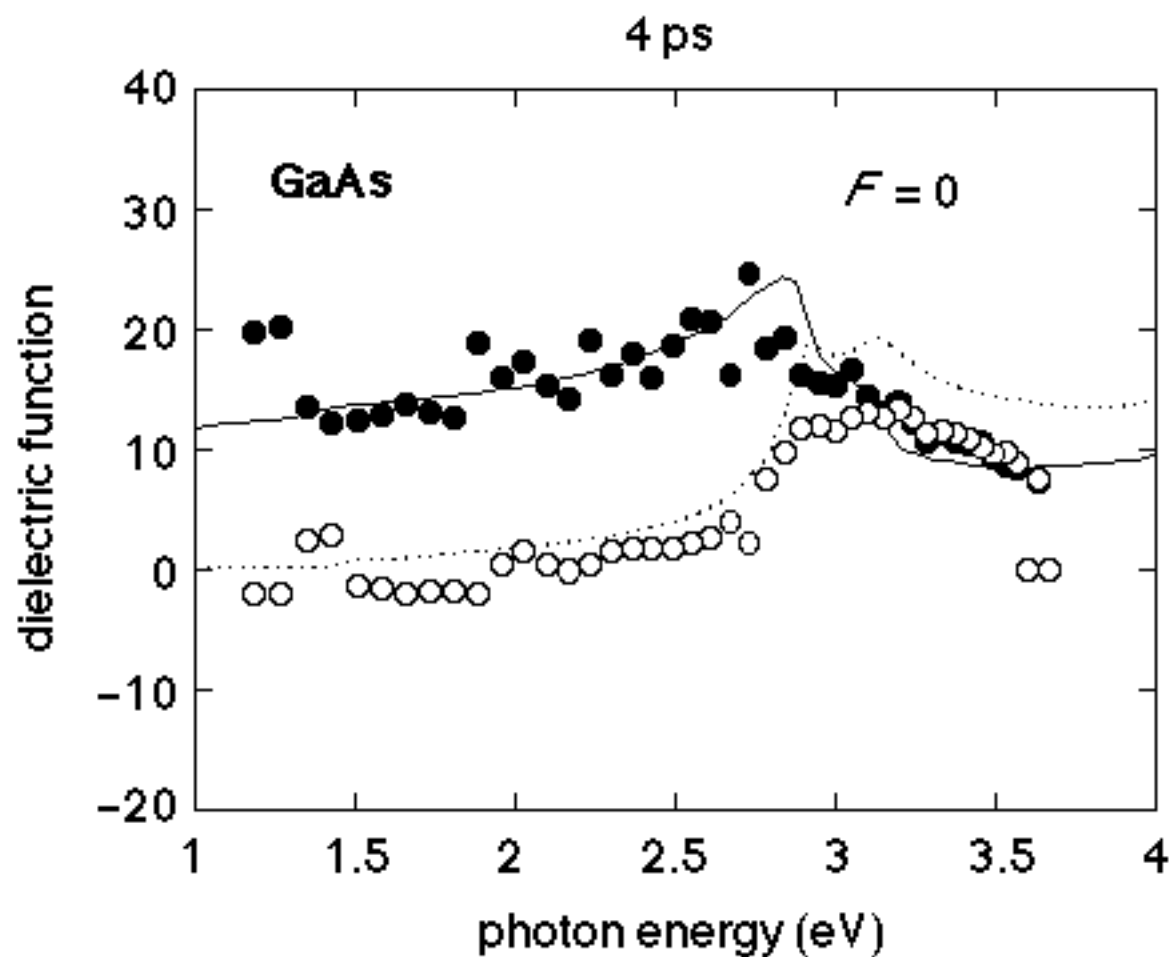
WHITE LIGHT SPECTRUM



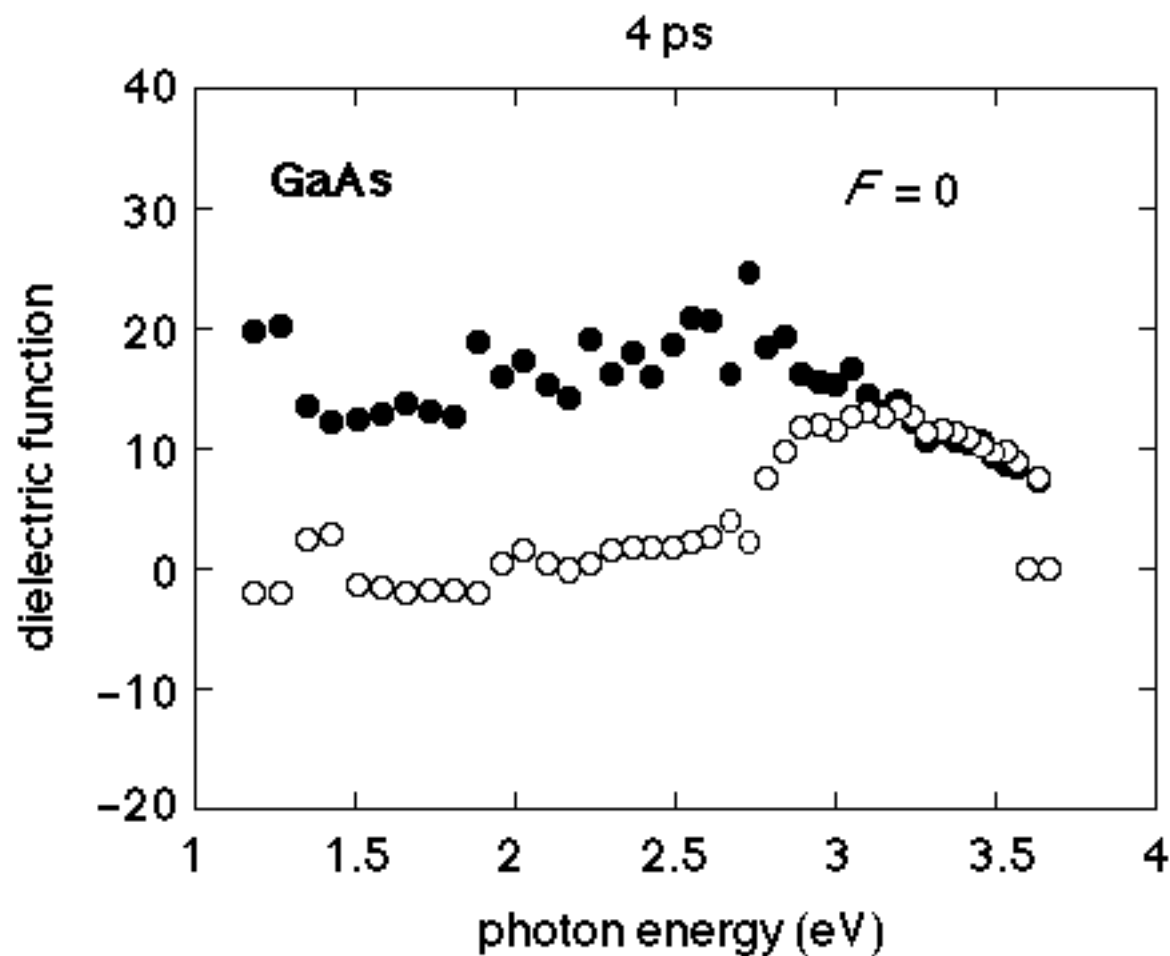
BROADBAND DATA



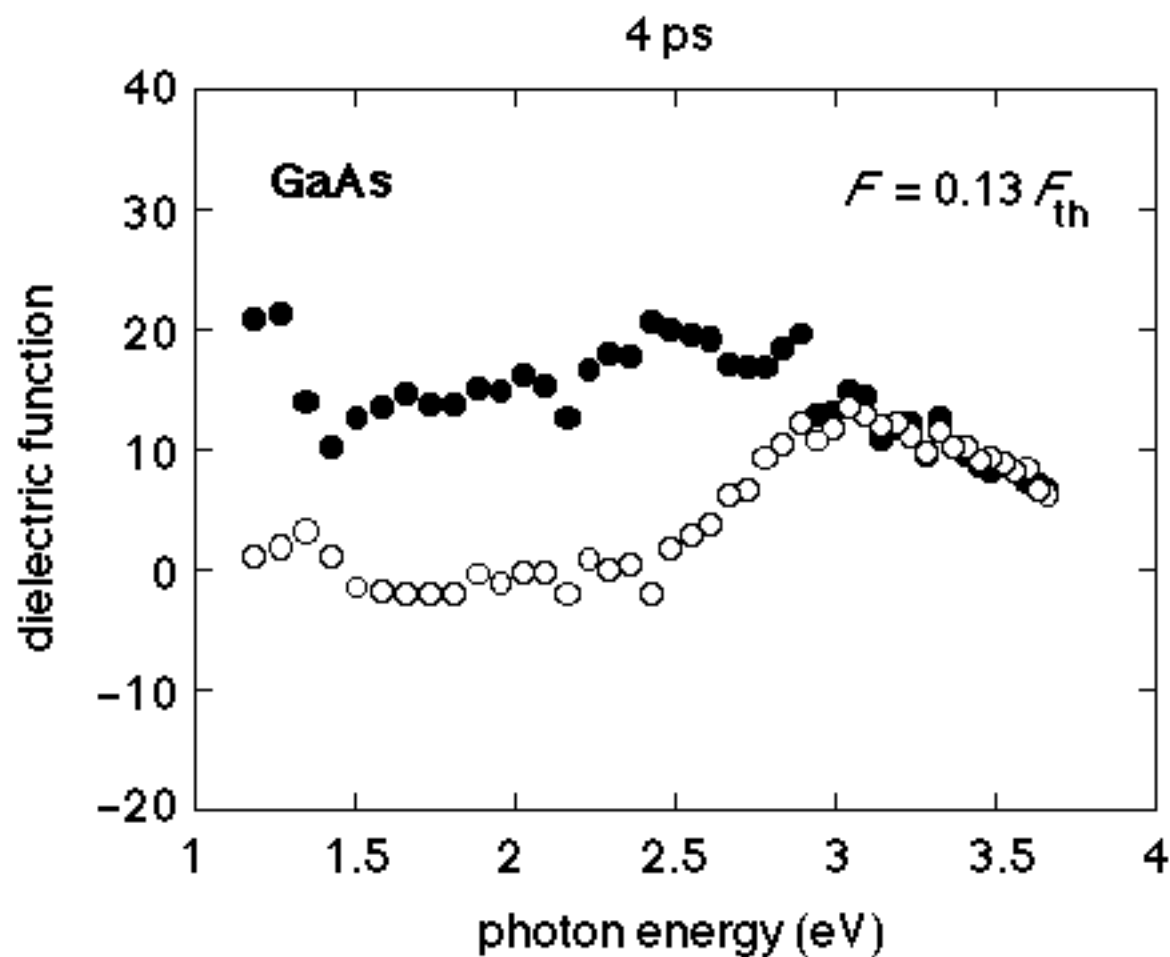
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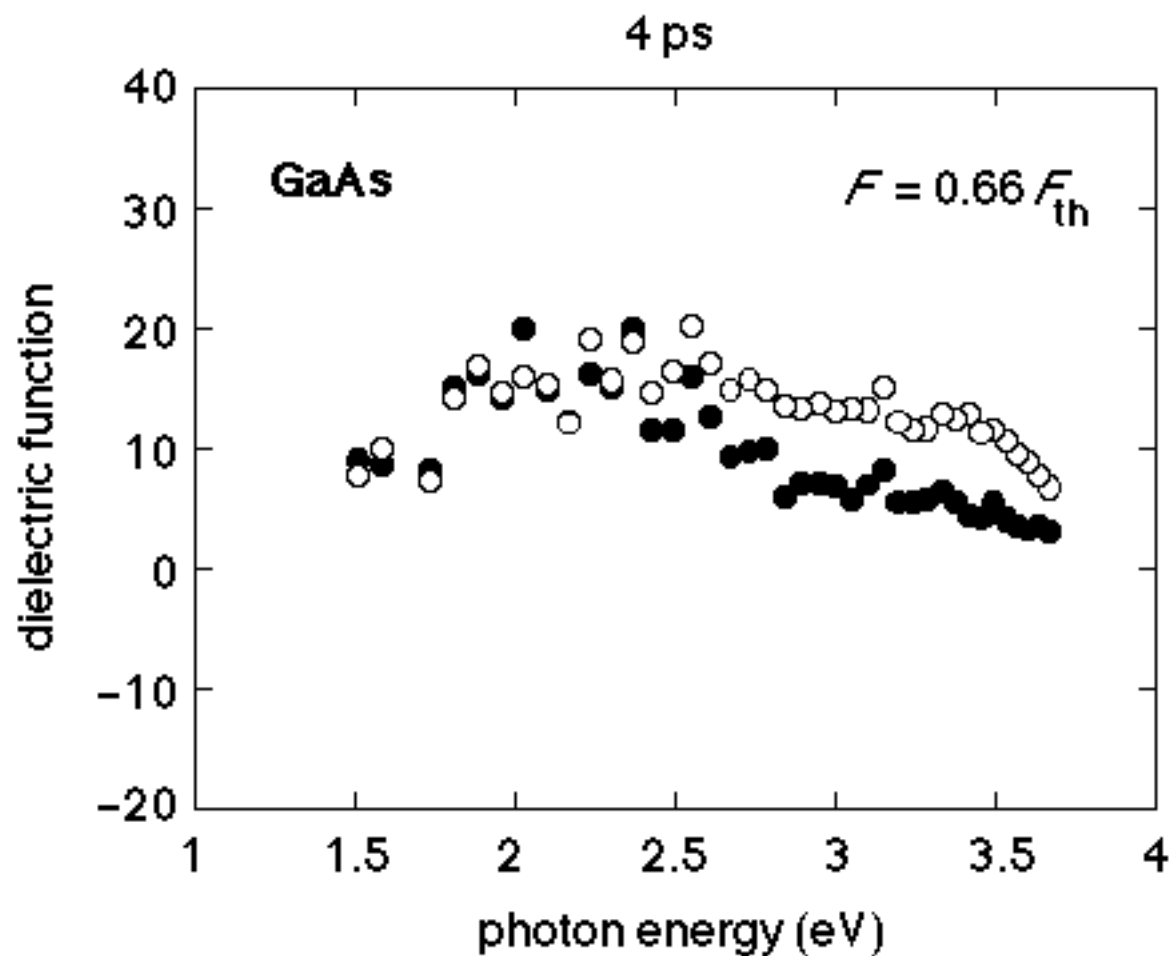
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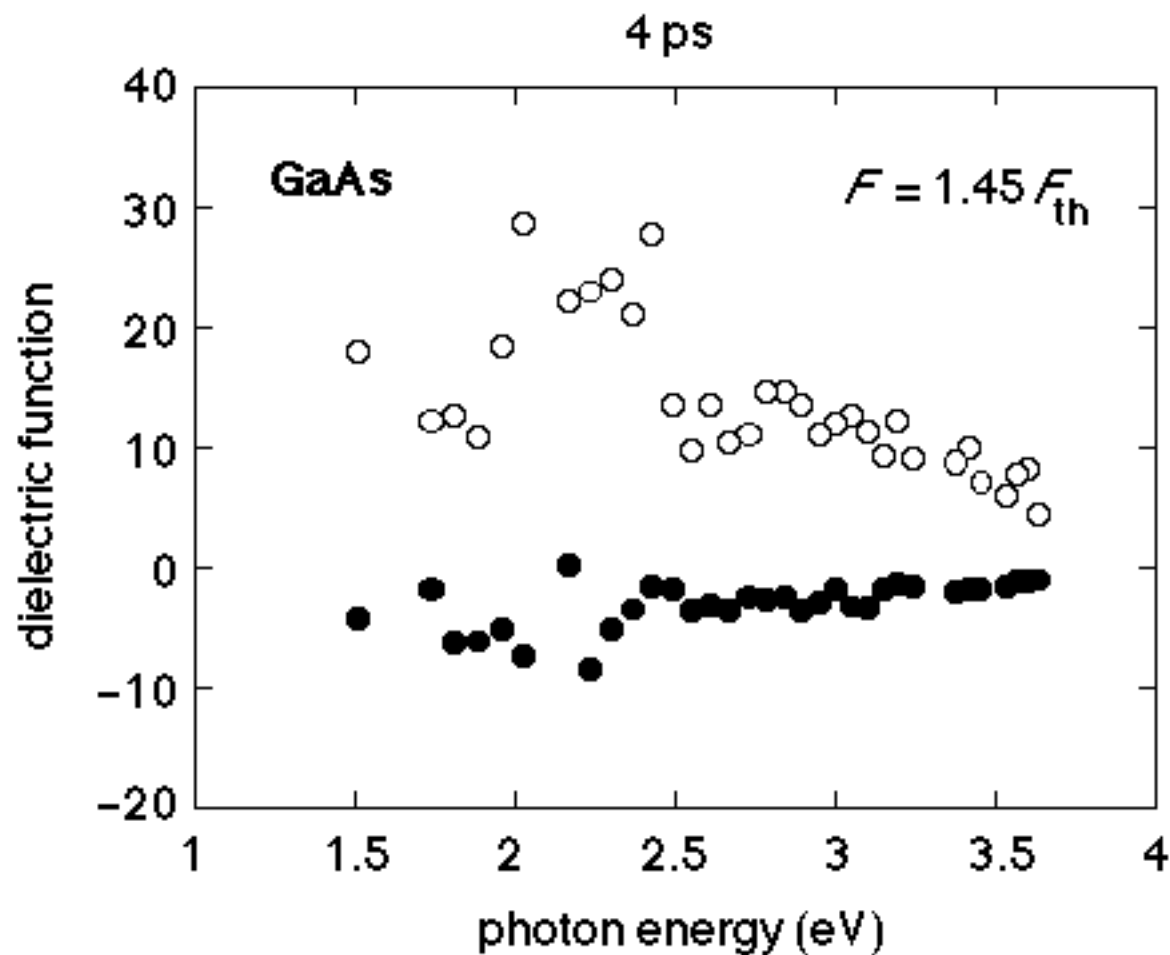
BROADBAND DATA



BROADBAND DATA



BROADBAND DATA



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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