



PHASE TRANSITIONS INDUCED BY FEMTOSECOND LASER PULSES

Eric Mazur

Eli Glezer

Li Huang

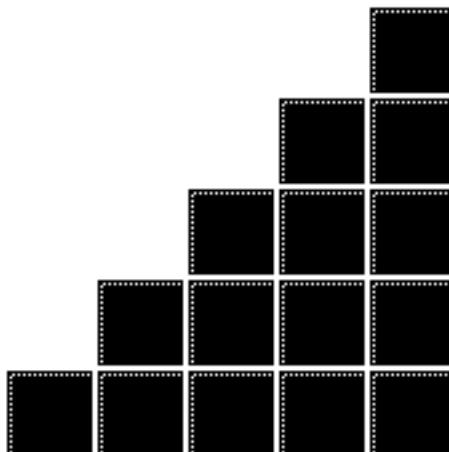
Paul Callan

Harvard University

FST '96 Workshop

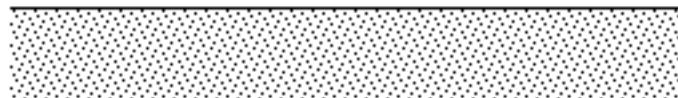
Tsukuba Research Center

22 February 1996



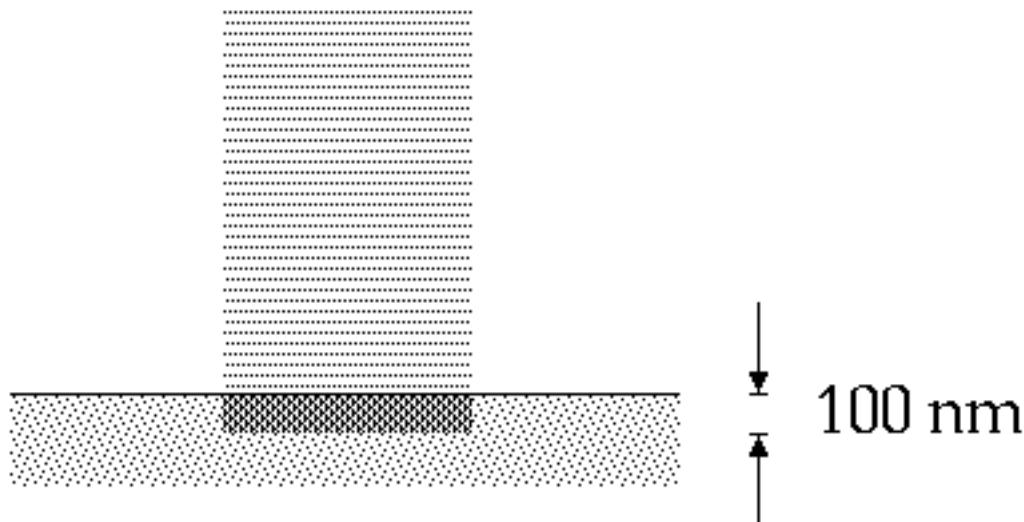
INTRODUCTION

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



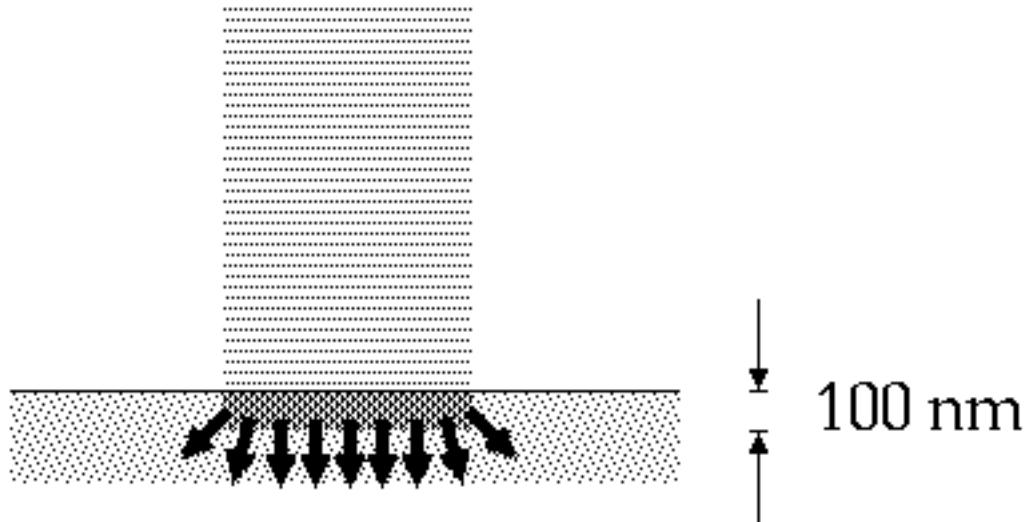
INTRODUCTION

laser deposits energy near surface...



INTRODUCTION

energy rapidly diffuses into bulk...

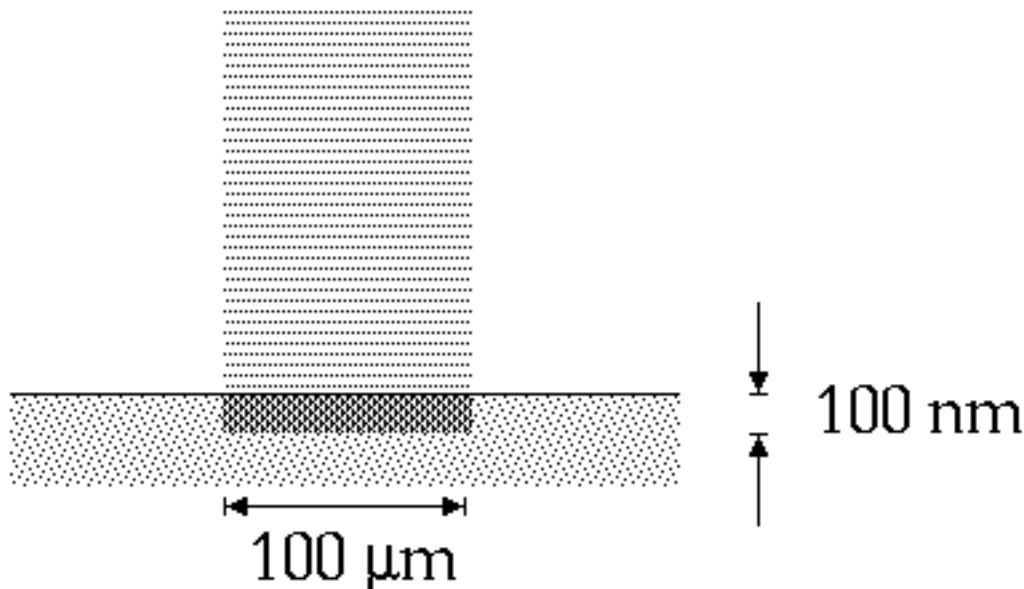


in 1 ns, diffusion length \approx 100 nm



INTRODUCTION

For $t < 1 \text{ ns}$ can ignore diffusion



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

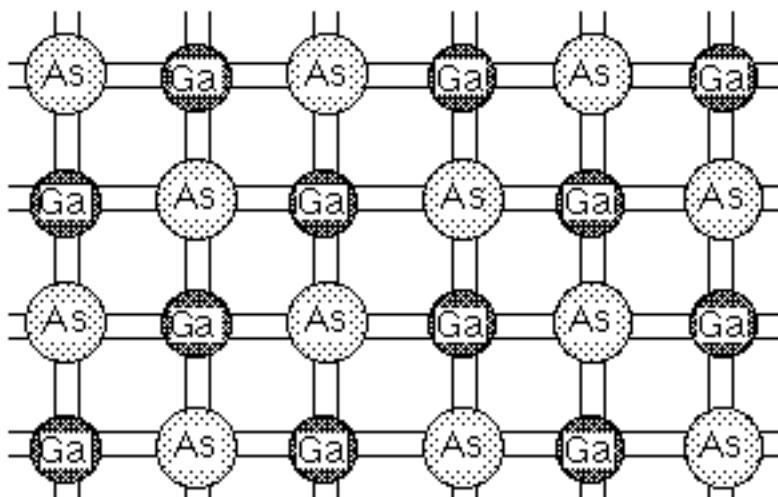


OUTLINE

- ① Background
- ② Femtosecond work
- ③ Future



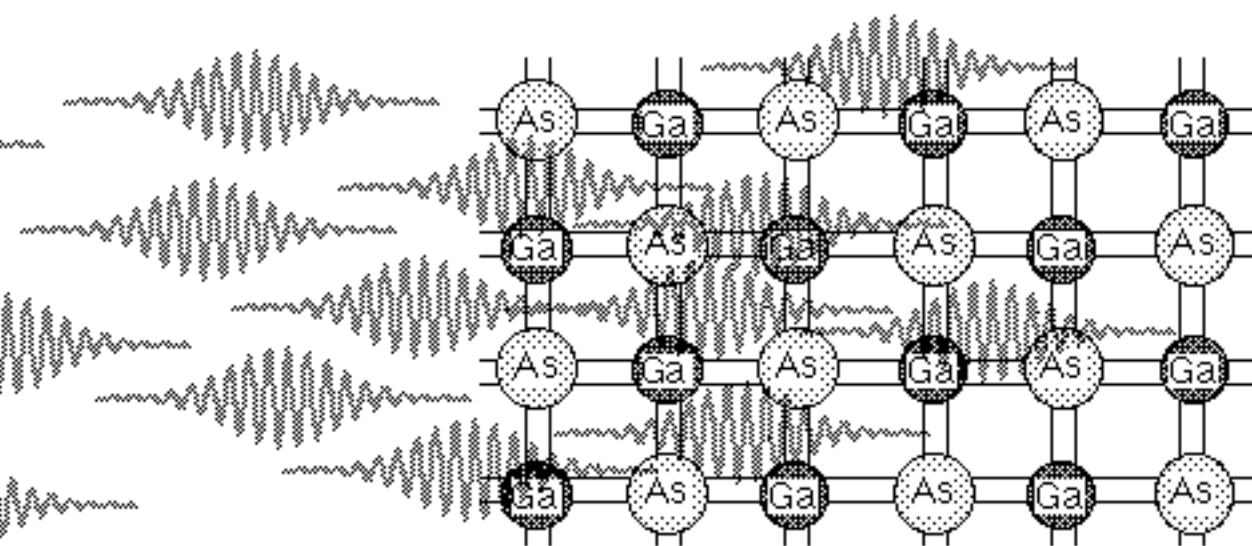
EARLY MODELS



how does light melt a solid?



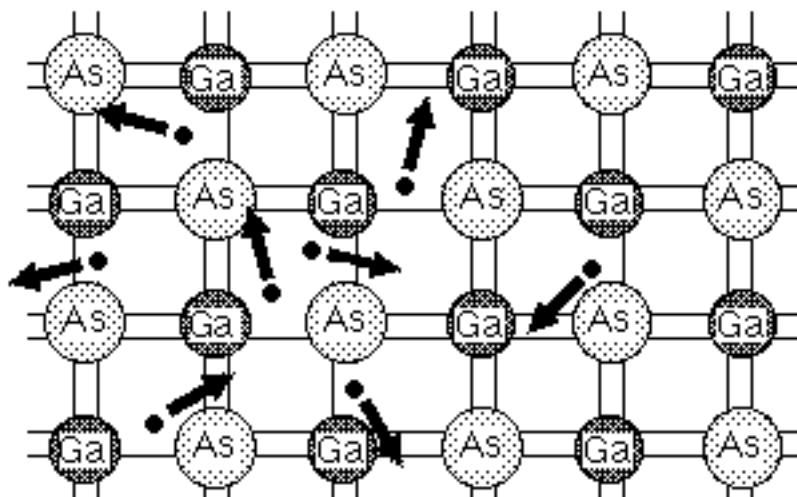
THERMAL MODEL



photons excite valence electrons...



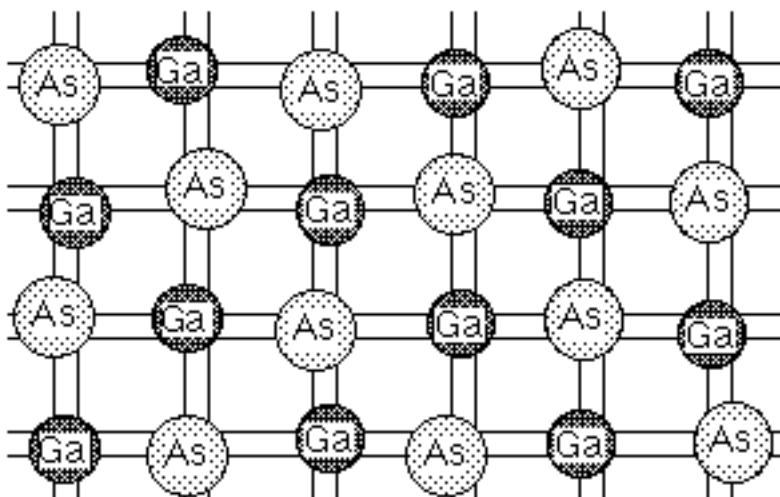
THERMAL MODEL



...and create hot electrons...



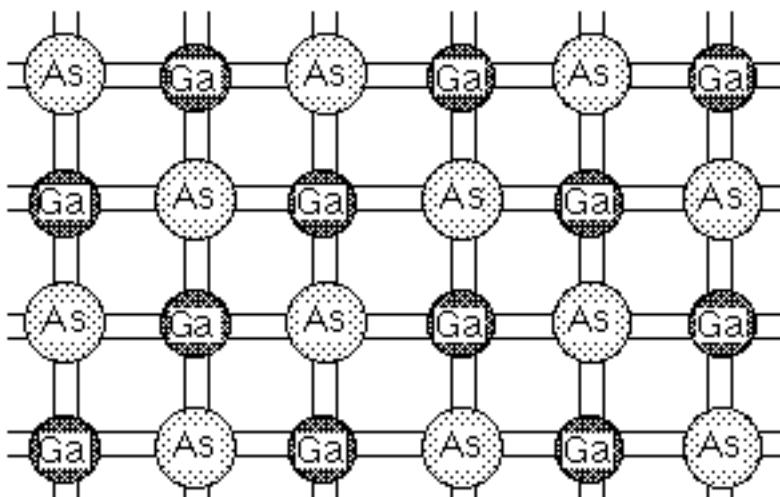
THERMAL MODEL



...which heat lattice



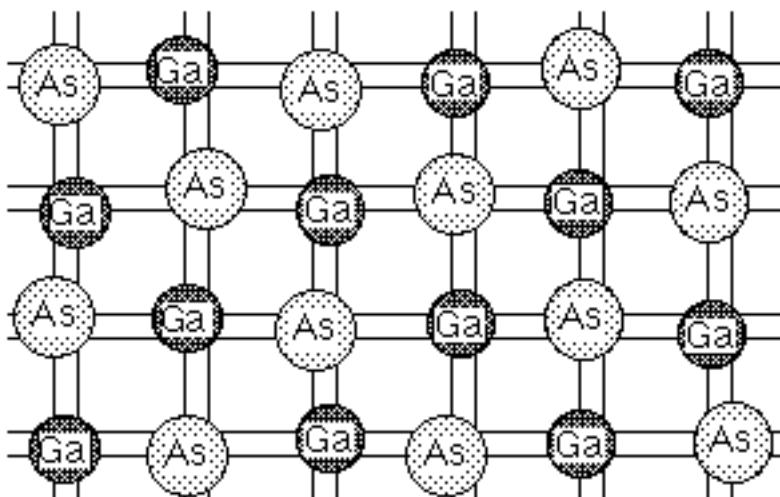
THERMAL MODEL



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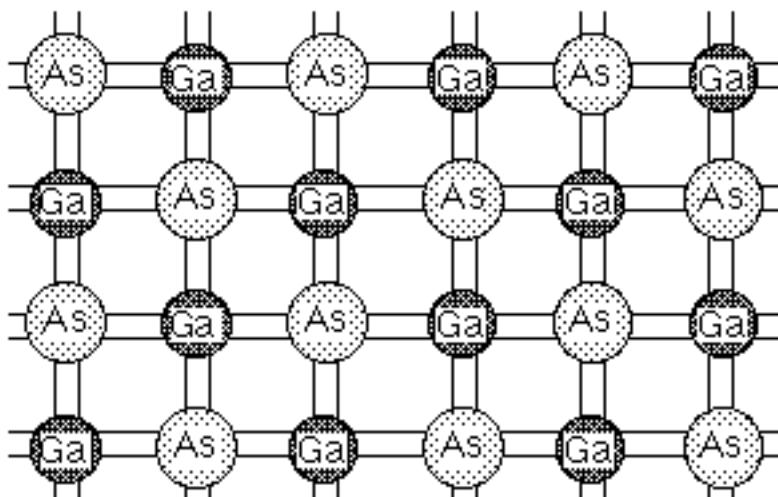
THERMAL MODEL



...which heat lattice



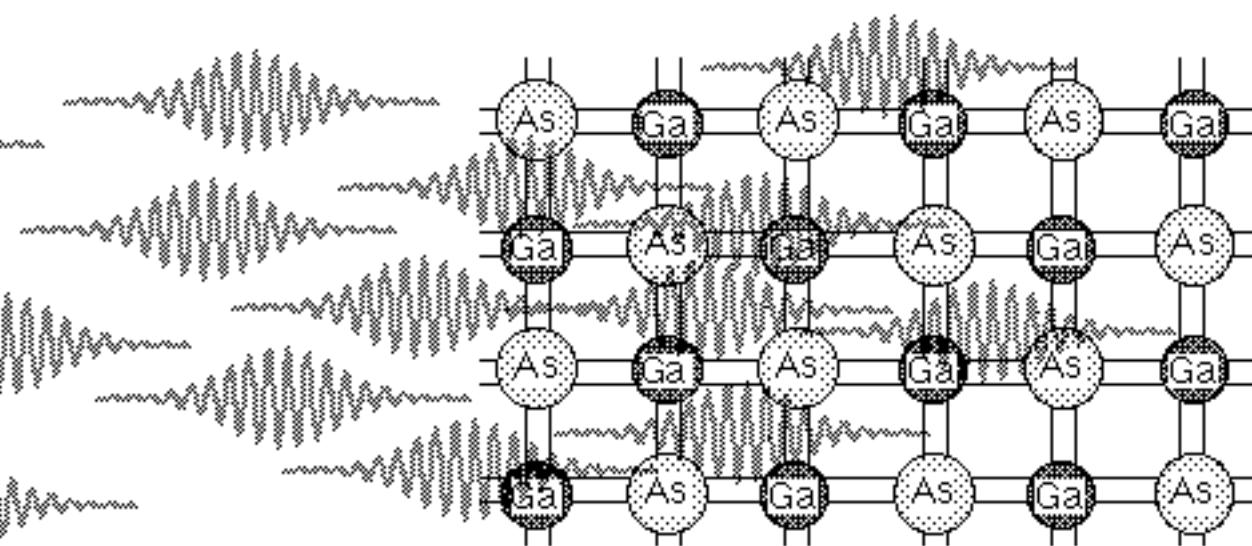
EARLY MODELS



...or...



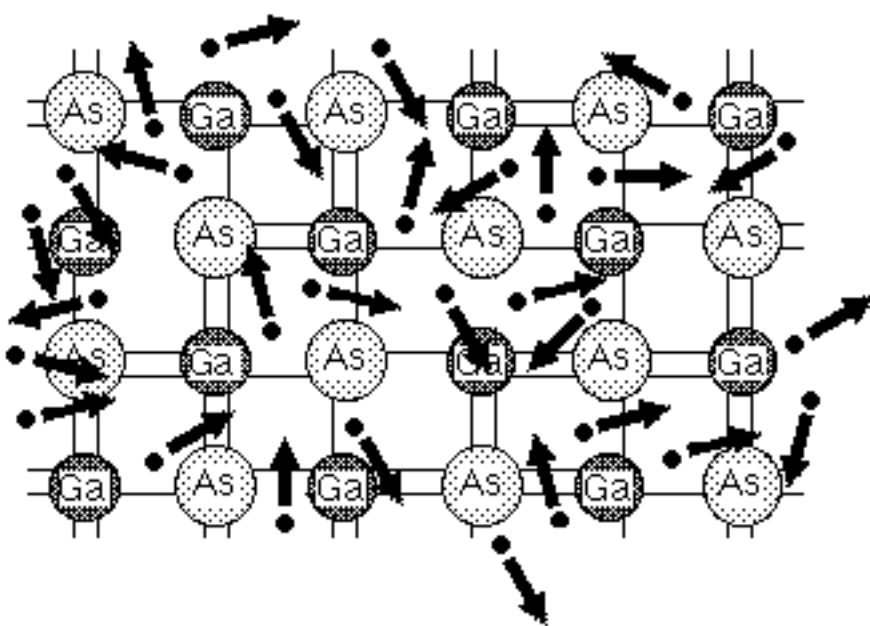
PLASMA ANNEALING



photons excite valence electrons...



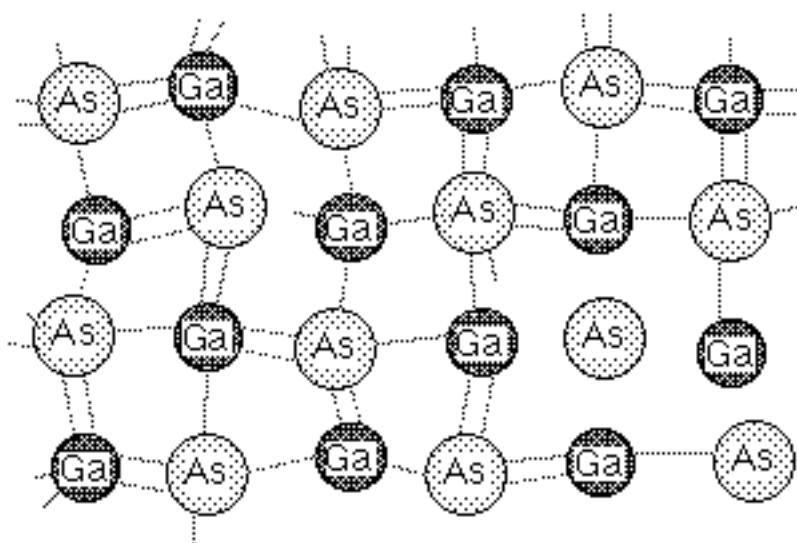
PLASMA ANNEALING



...creating a dense plasma...



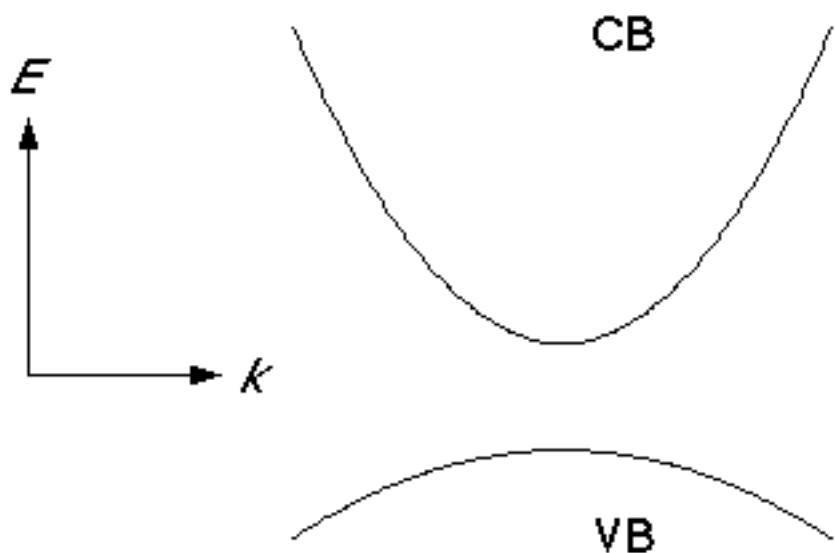
PLASMA ANNEALING



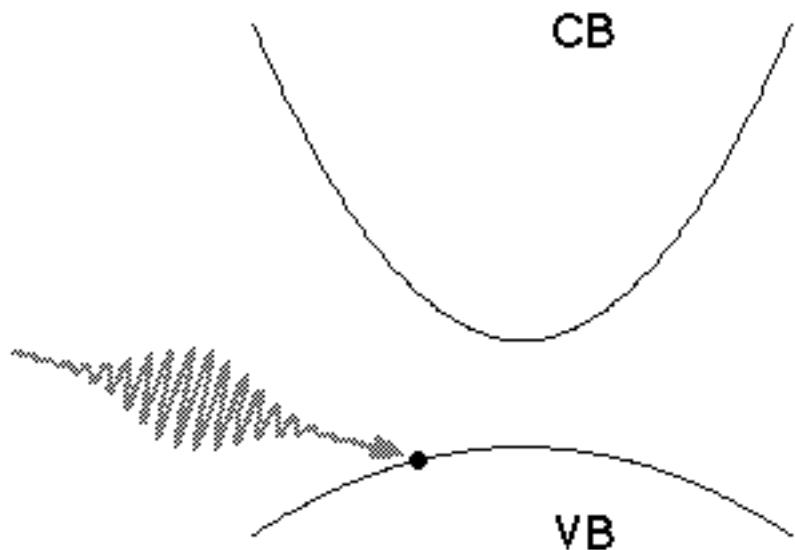
...and breaking bonds



EXCITATION



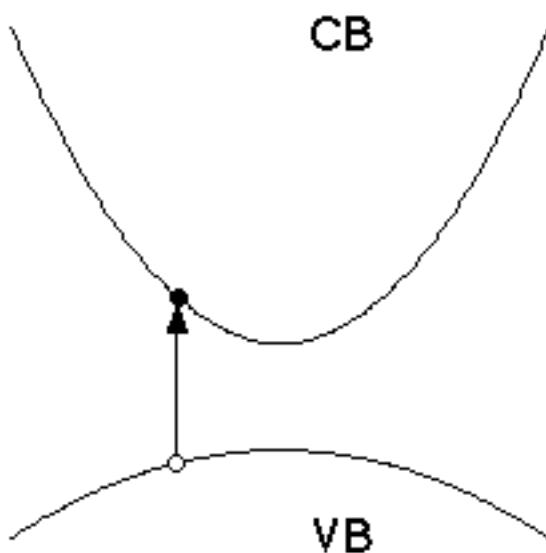
EXCITATION



photon excites valence electron



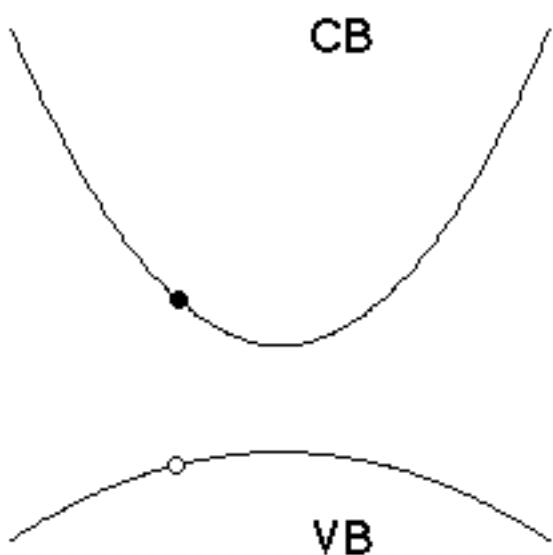
EXCITATION



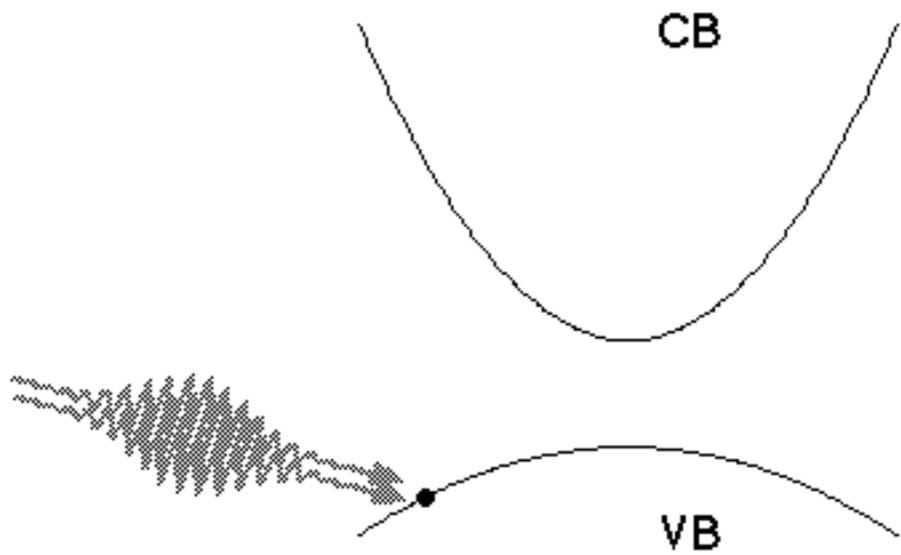
creating an electron-hole pair



EXCITATION



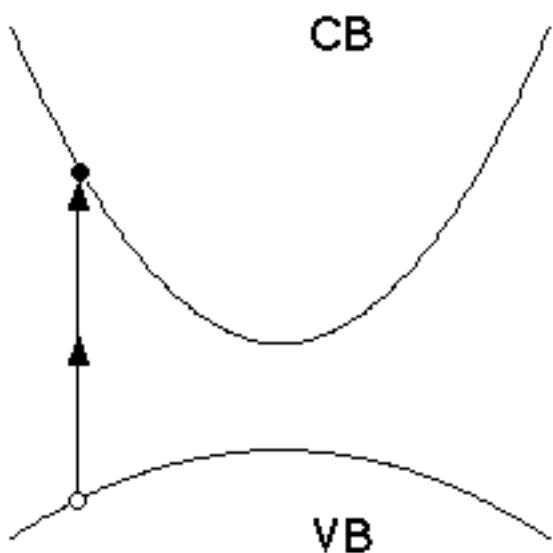
EXCITATION



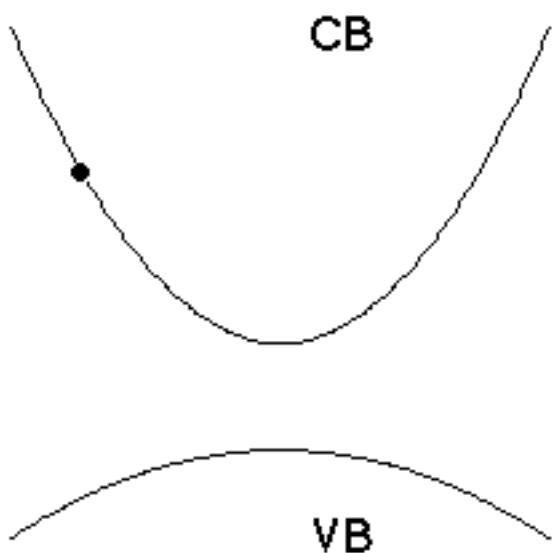
multiphoton excitation



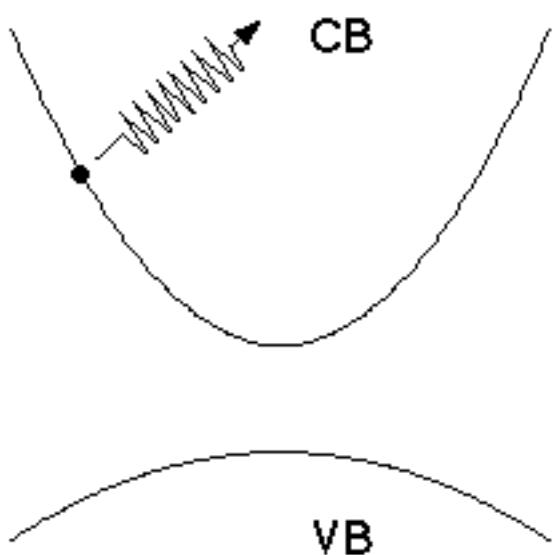
EXCITATION



EXCITATION



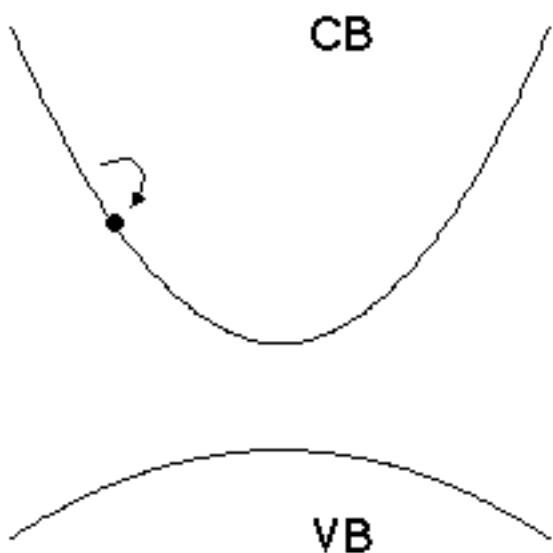
RELAXATION



emission of LO phonons



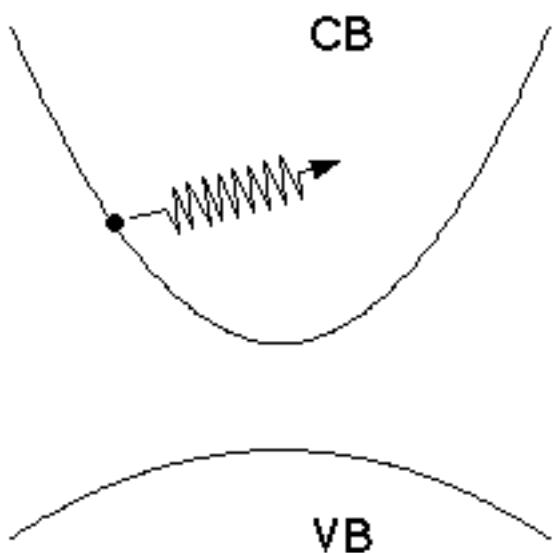
RELAXATION



emission of LO phonons



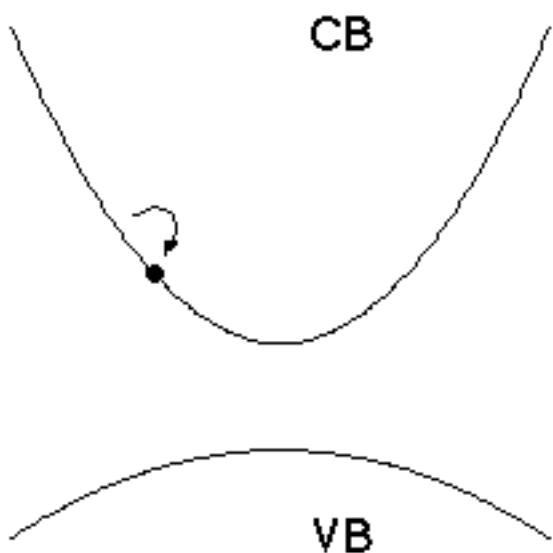
RELAXATION



emission of LO phonons



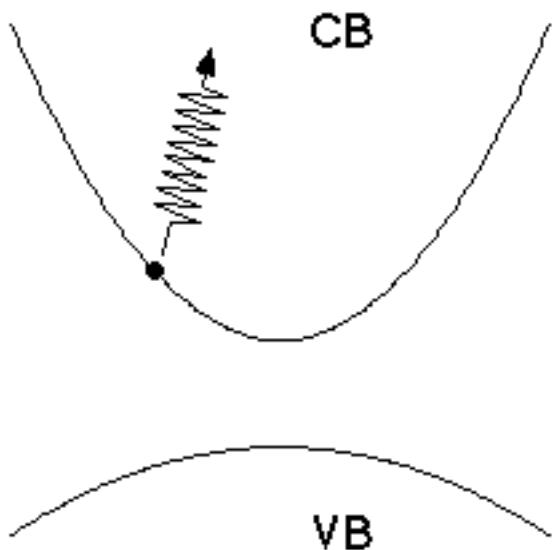
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emission of LO phonons



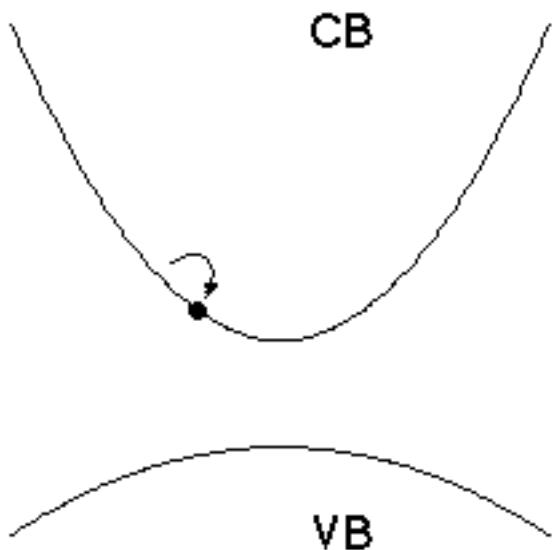
RELAXATION



emission of LO phonons



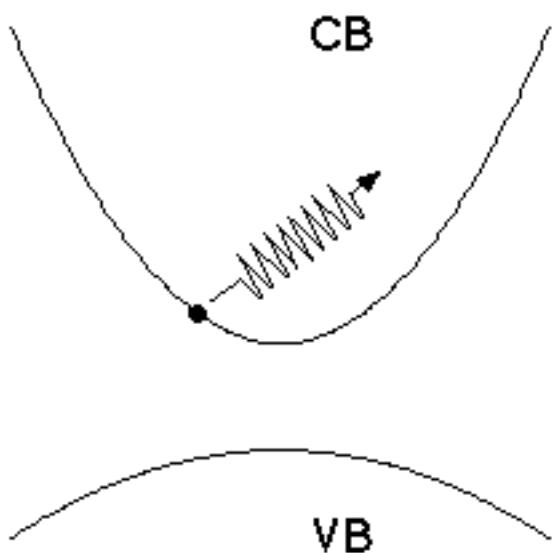
RELAXATION



emission of LO phonons



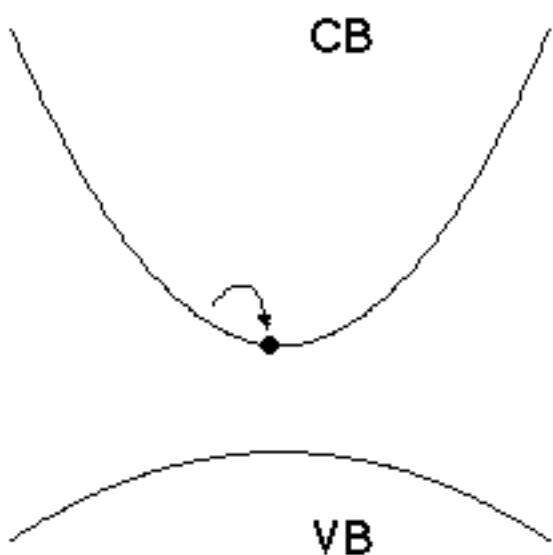
RELAXATION



emission of LO phonons



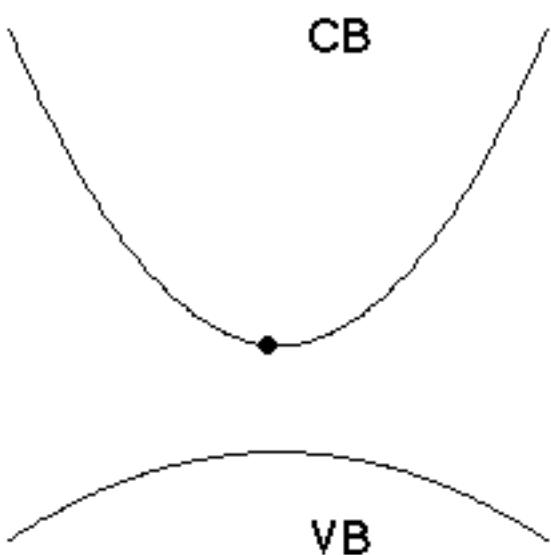
RELAXATION



emission of LO phonons

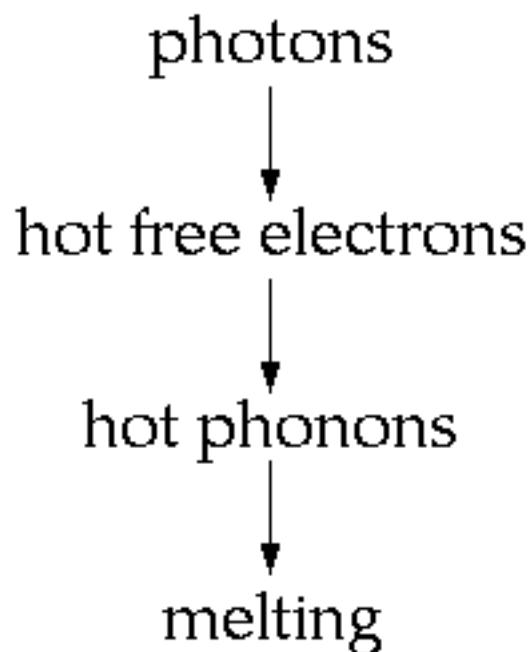


RELAXATION



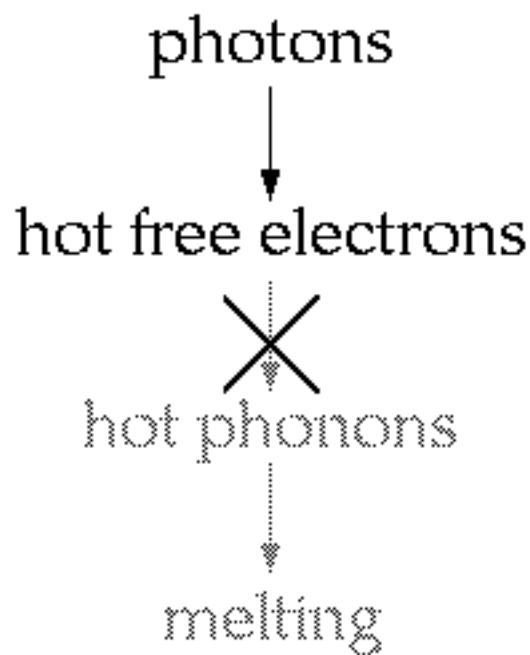
TIME SCALE

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



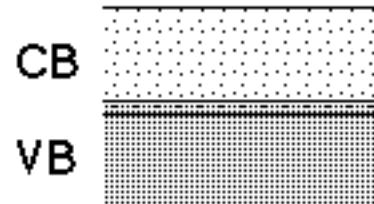
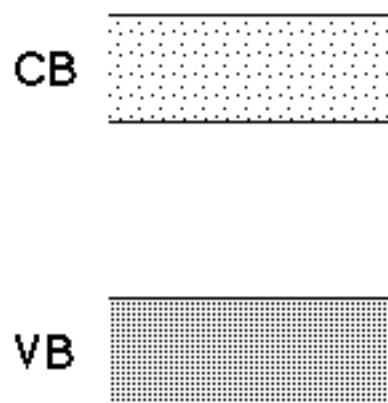
TIME SCALE

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



BEYOND PHONONS

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



BEYOND PHONONS

intrinsic properties

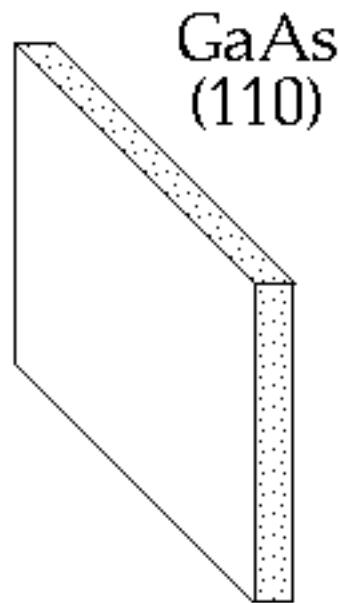
measured properties

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

$$\begin{array}{ccc} \varepsilon', \varepsilon''(\omega) & \longrightarrow & \text{SHG} \\ \varepsilon', \varepsilon''(2\omega) \\ \chi^{(2)} \end{array}$$

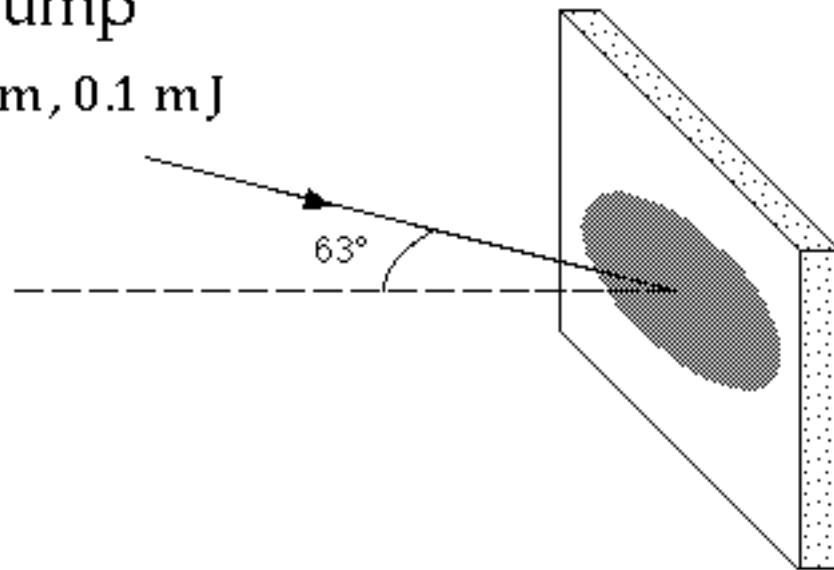


2.2 eV DIELECTRIC CONSTANT



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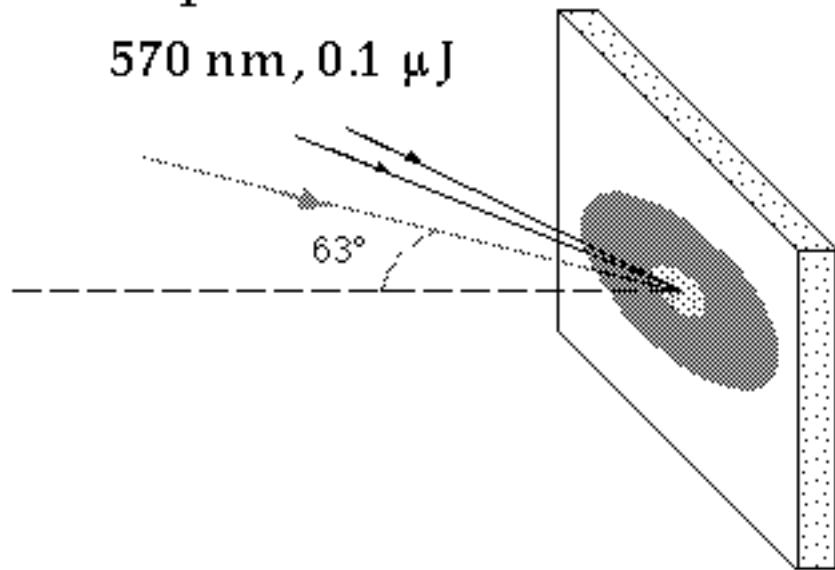
pump
640 nm, 0.1 mJ



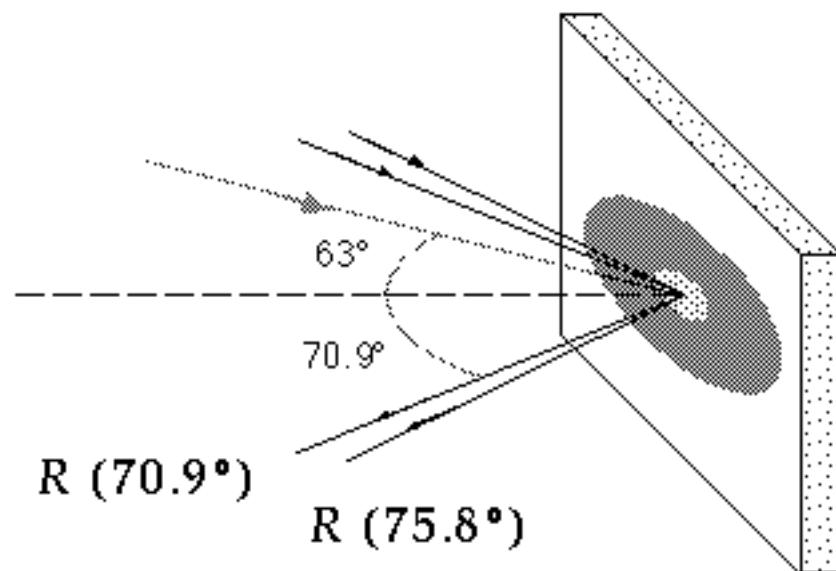
2.2 eV DIELECTRIC CONSTANT

probes
570 nm, 0.1 μ J

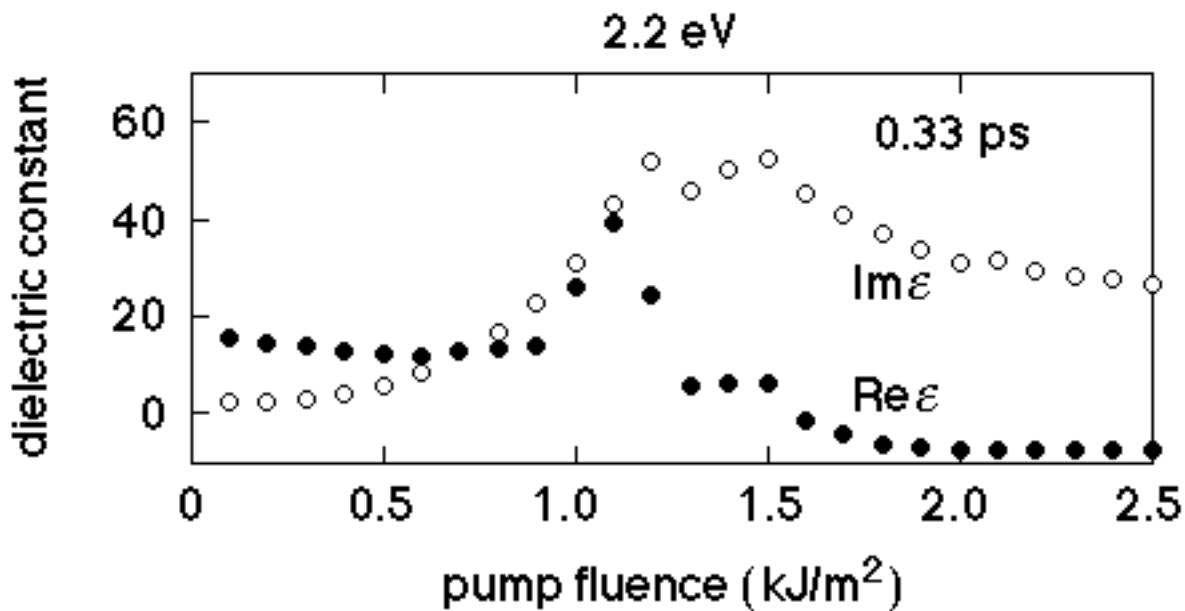
all beams
70 fs, *p*-pol



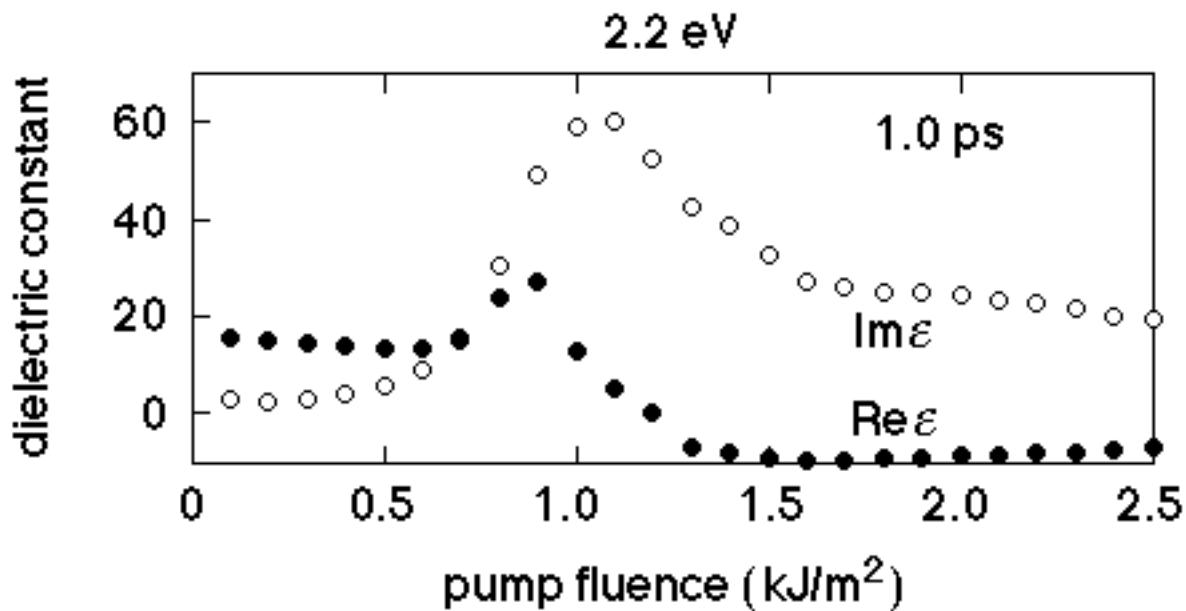
2.2 eV DIELECTRIC CONSTANT



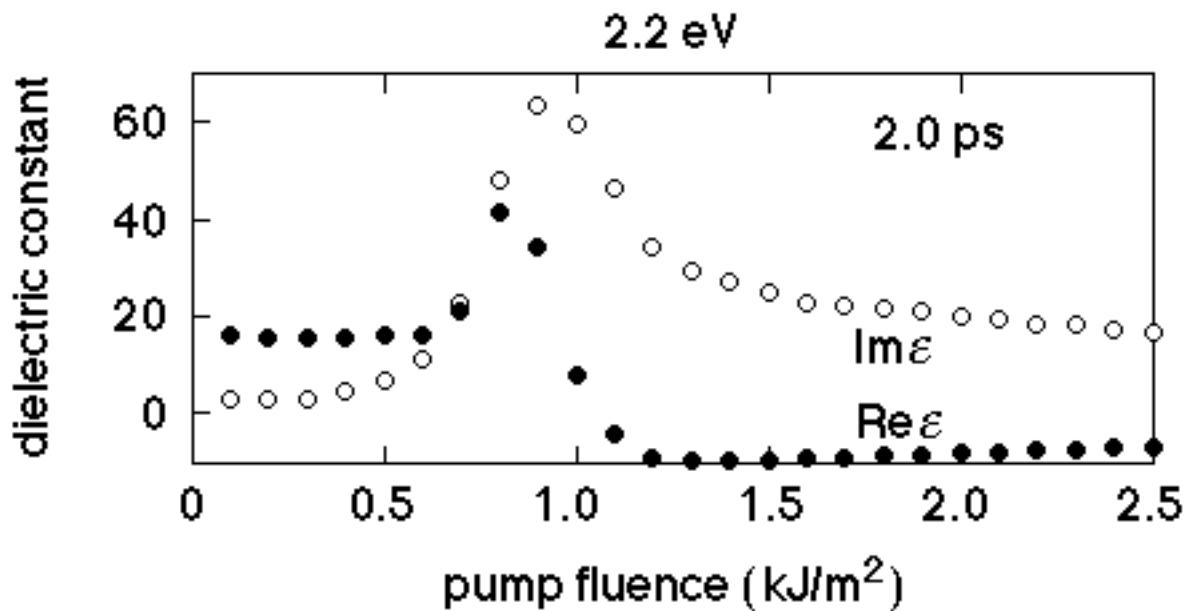
FLUENCE DEPENDENCE



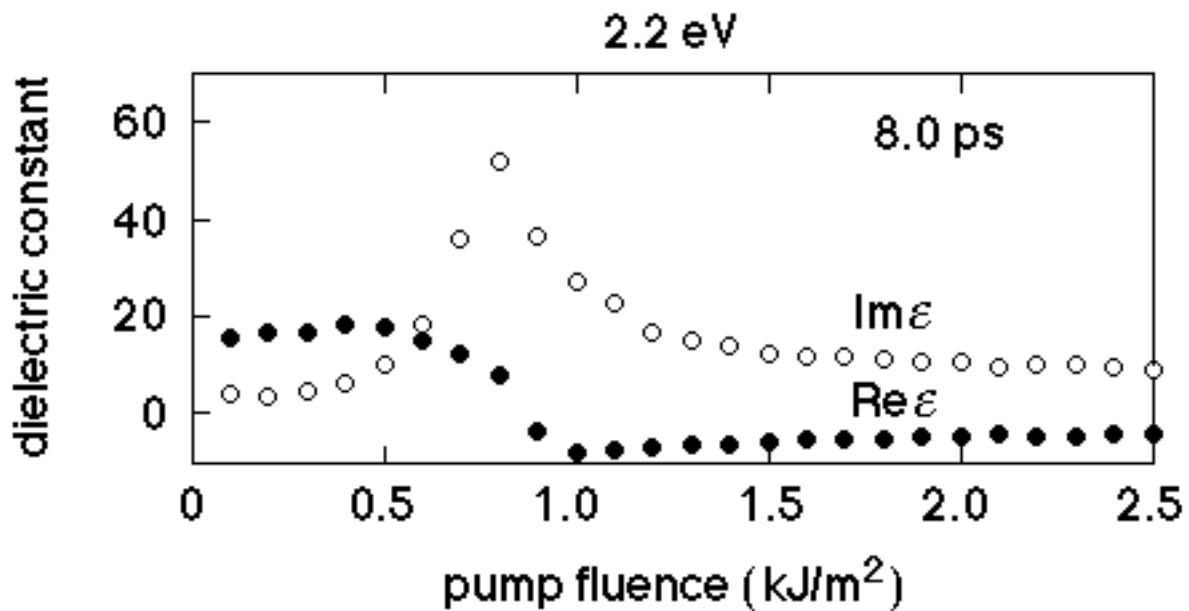
FLUENCE DEPENDENCE



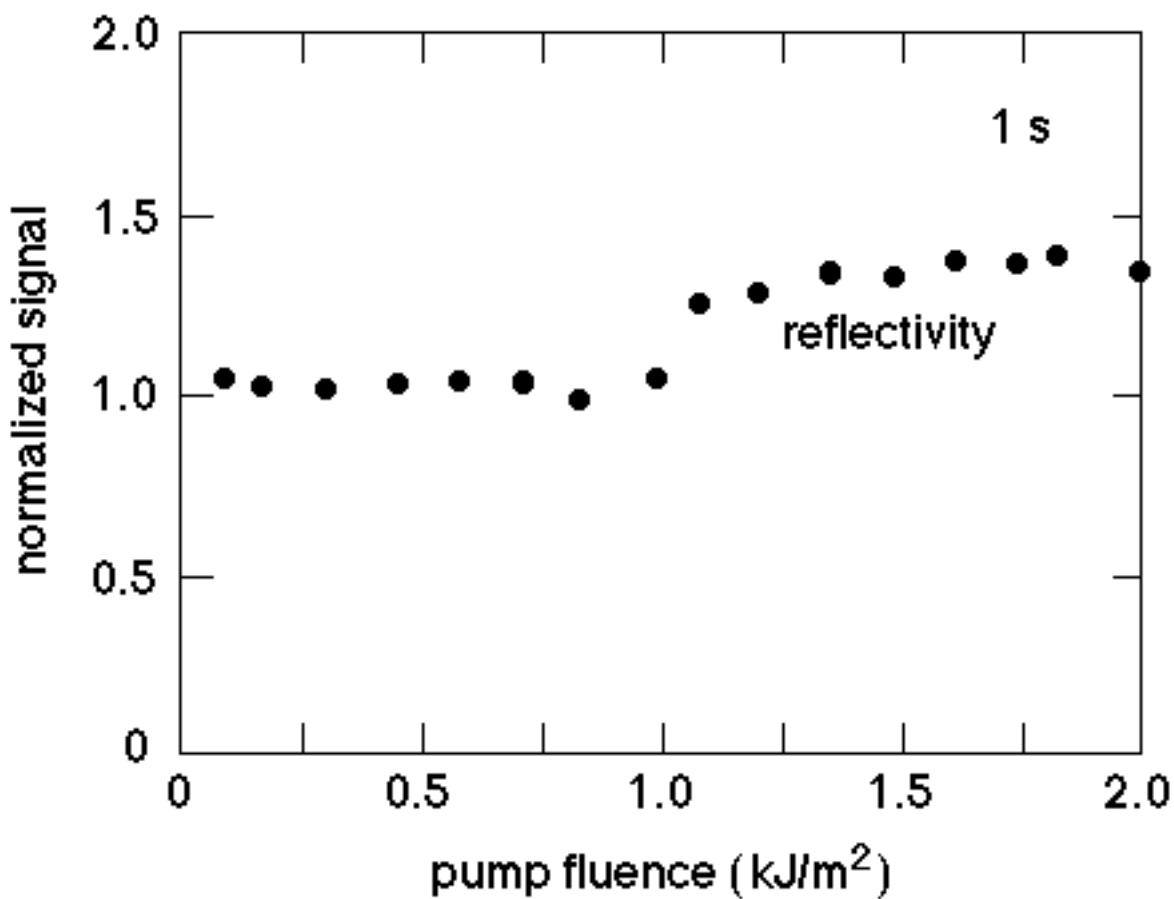
FLUENCE DEPENDENCE



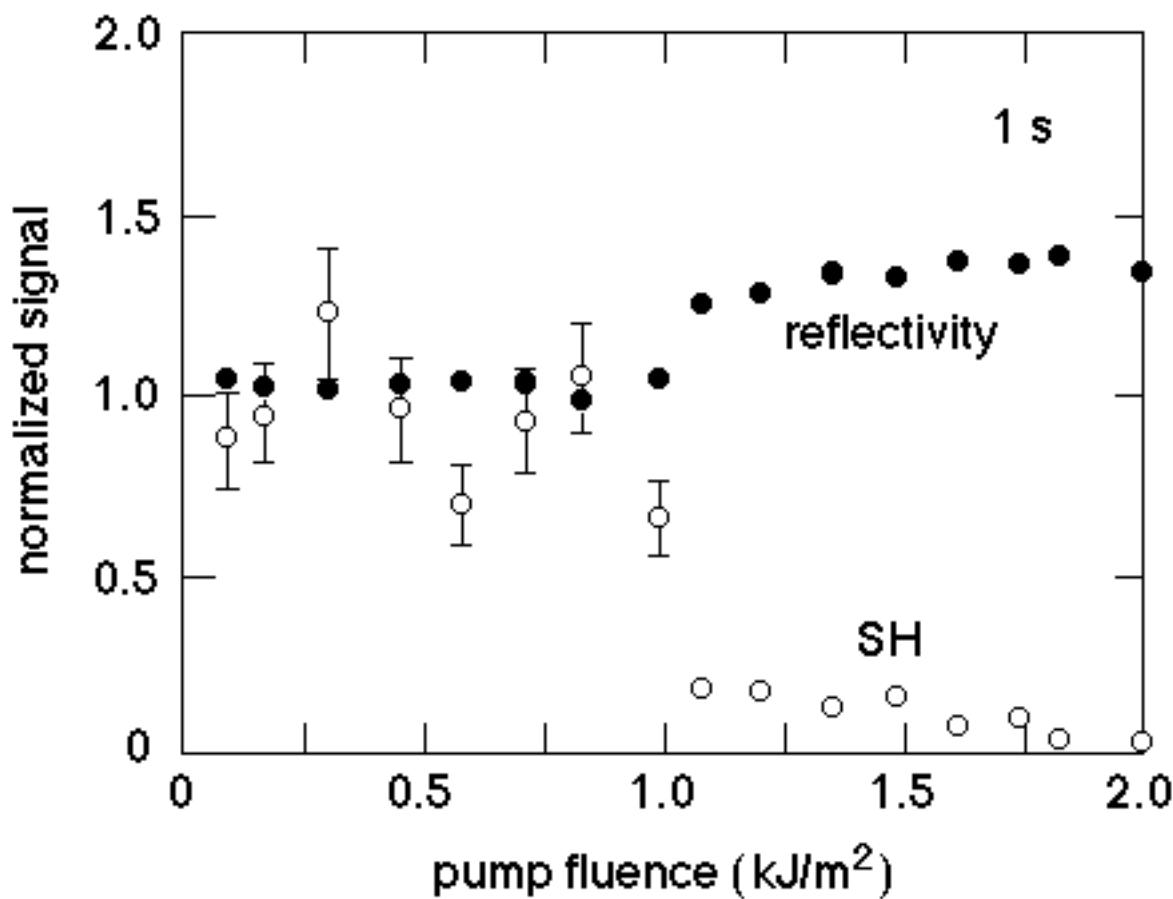
FLUENCE DEPENDENCE



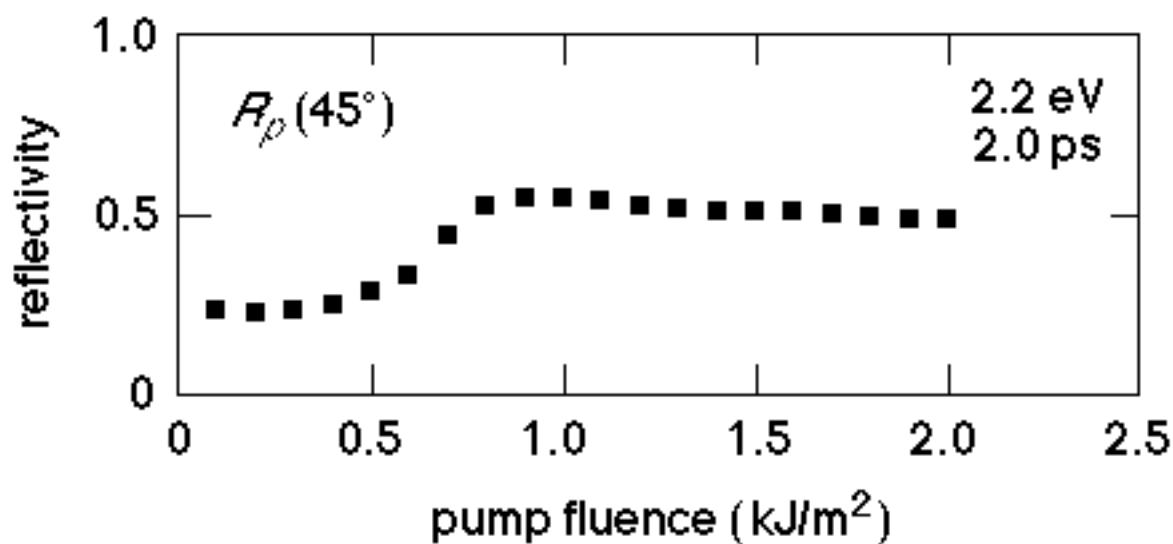
REVERSIBLE vs. IRREVERSIBLE



REVERSIBLE vs. IRREVERSIBLE



SIMPLE MODEL

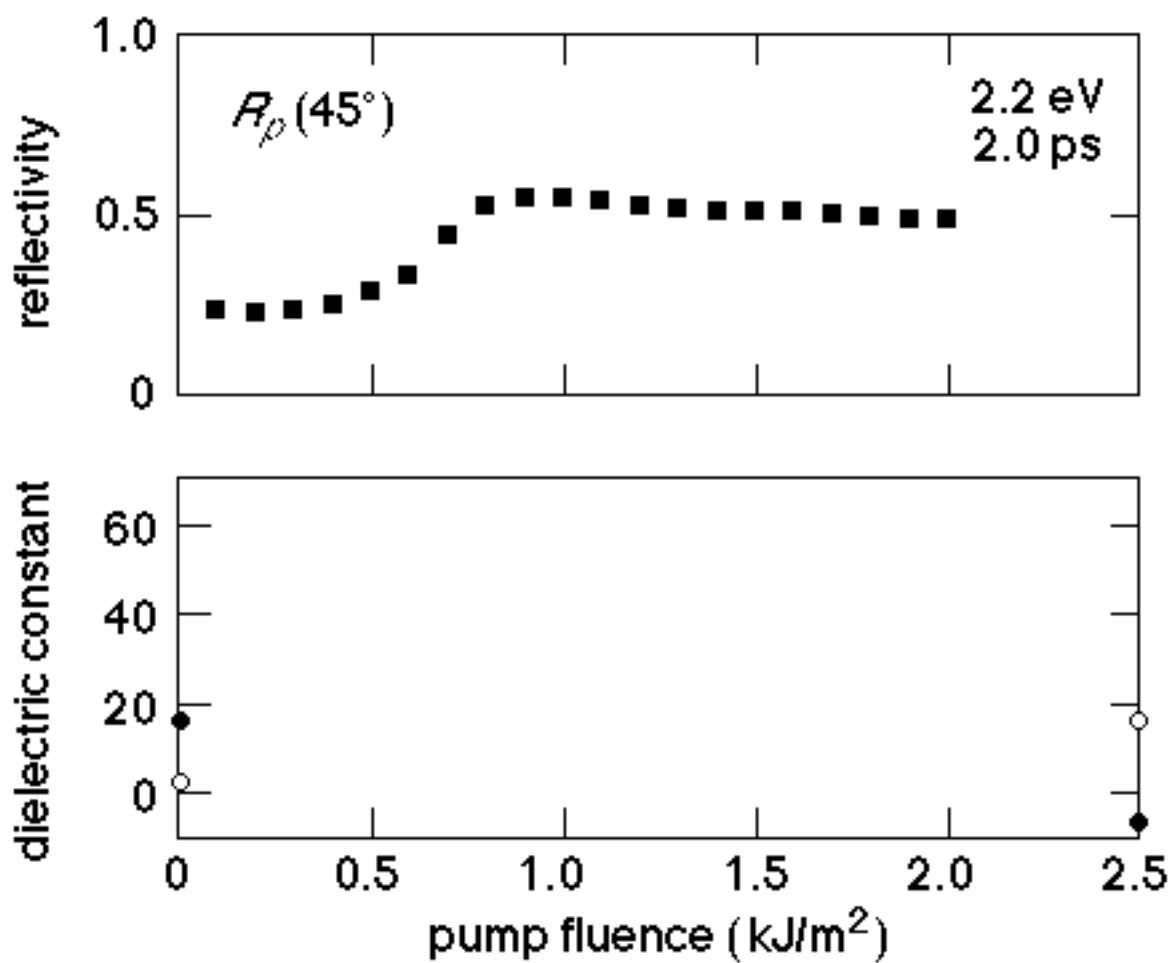


Drude model:

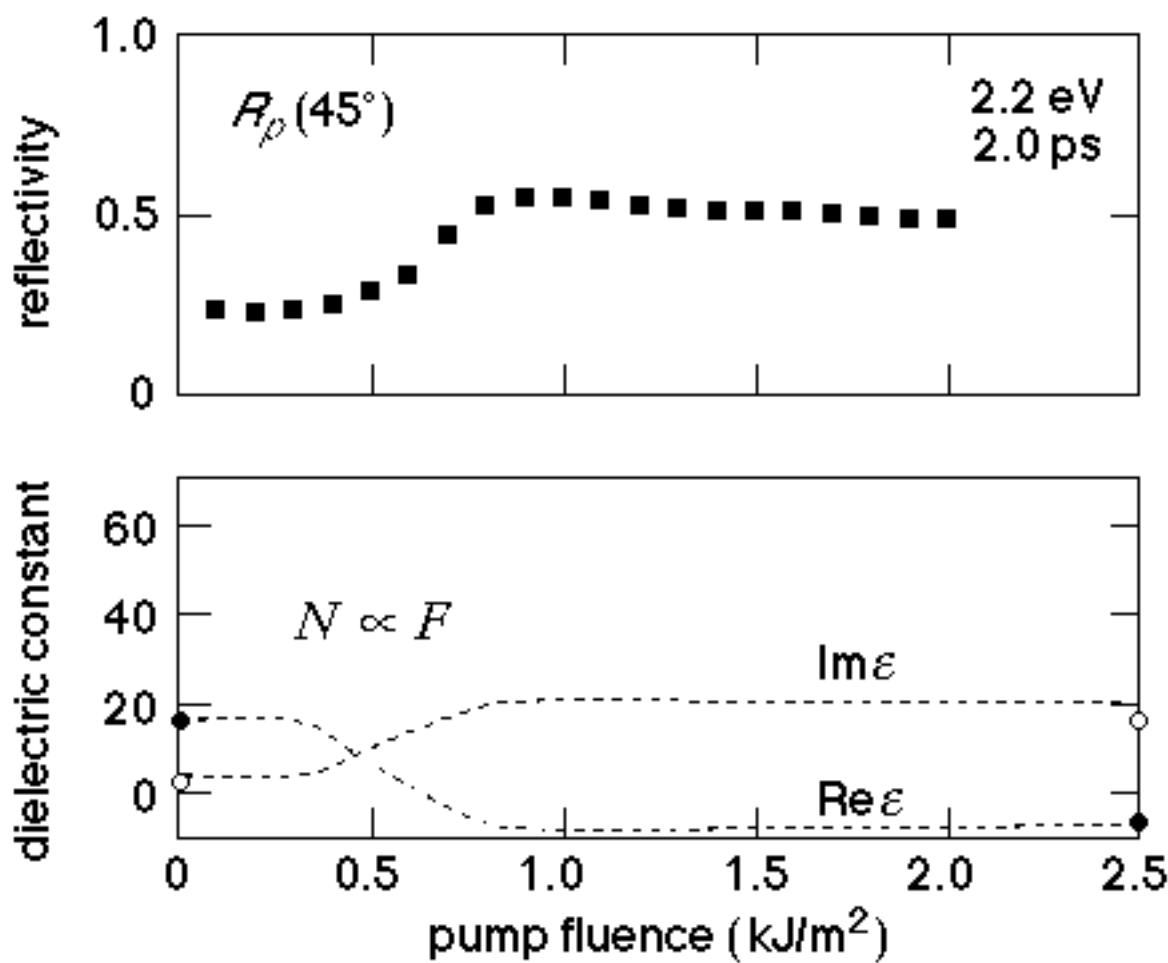
$$\text{Re}(\varepsilon) = 1 - \frac{Ne^2}{m\varepsilon_0} \frac{1}{\omega^2 + \gamma^2} \quad \text{Im}(\varepsilon) = \frac{Ne^2}{m\varepsilon_0} \frac{\gamma\omega}{\omega^4 + \gamma^2\omega^2}$$



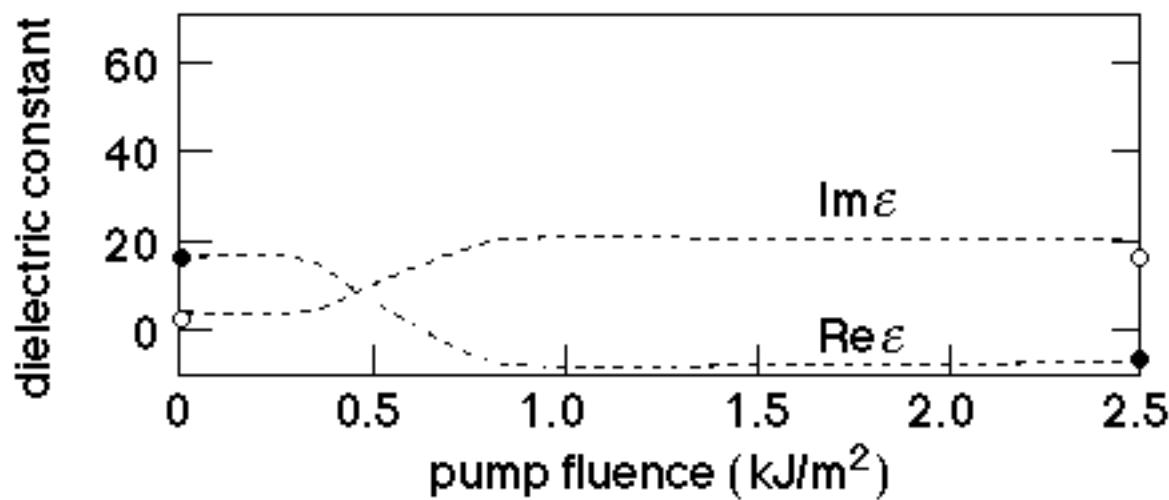
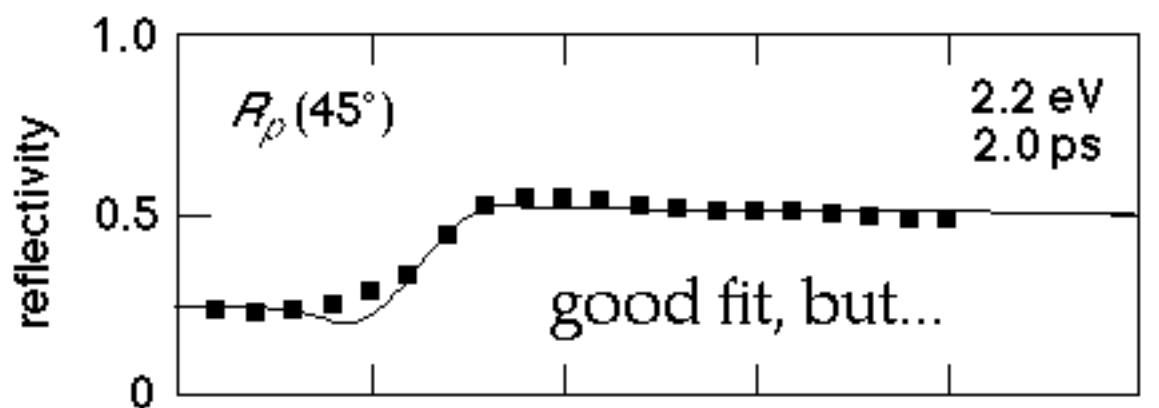
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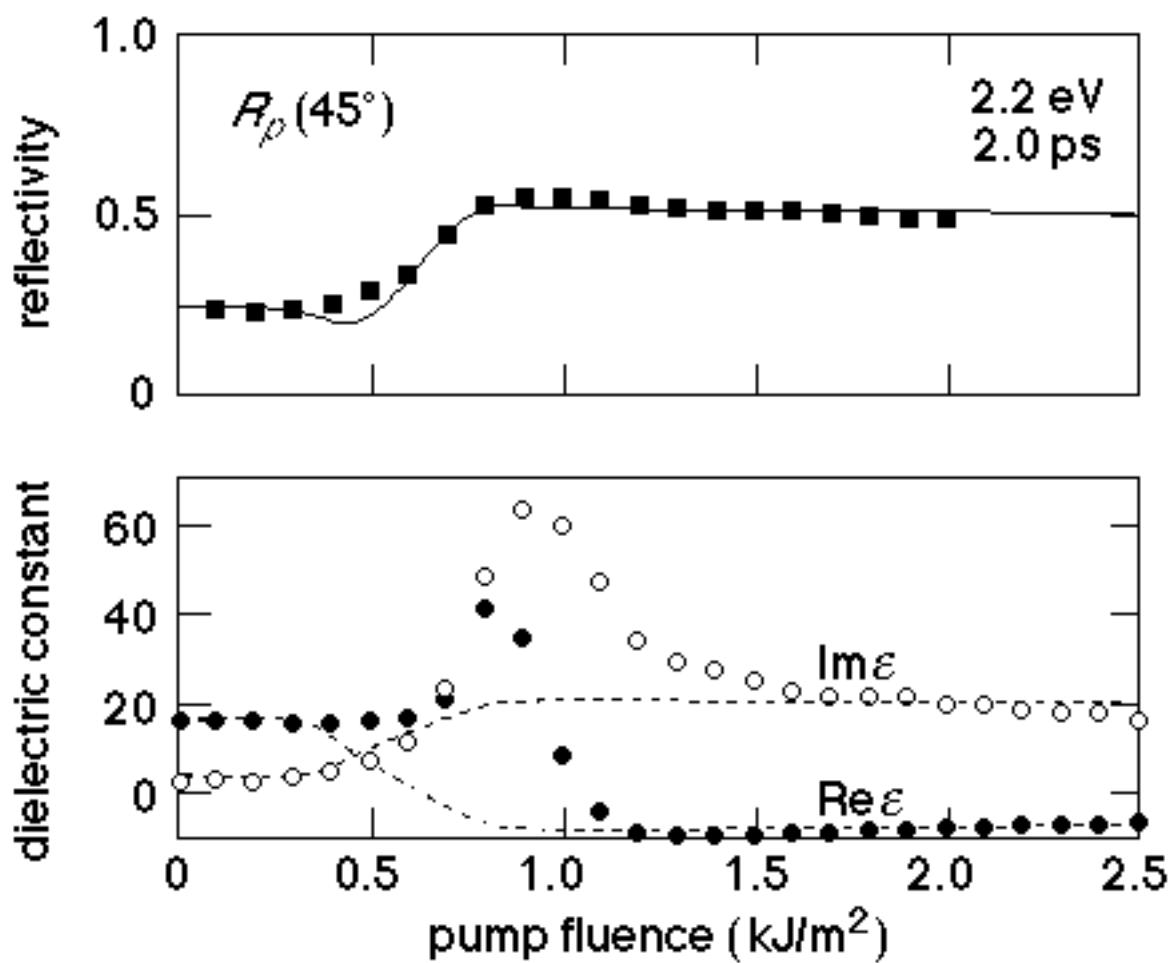
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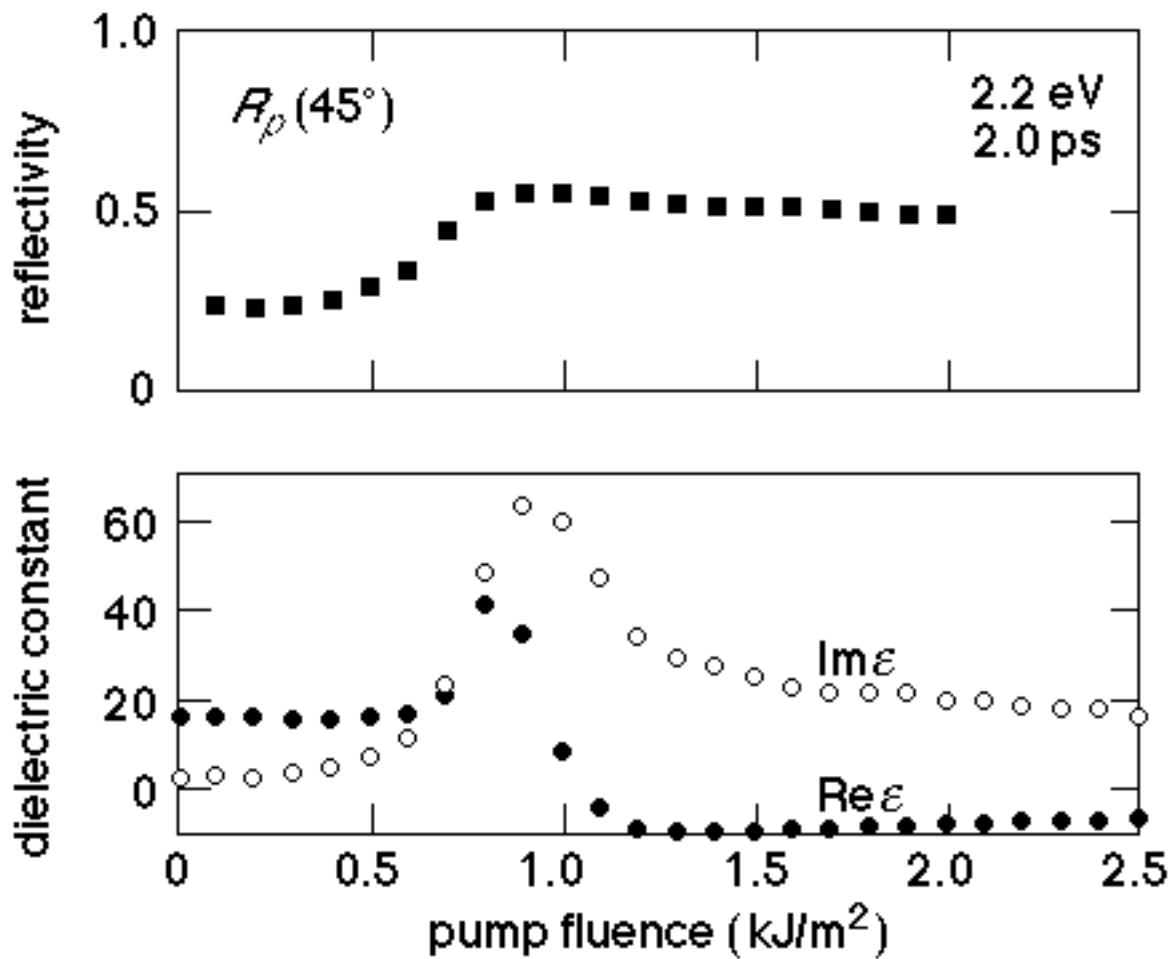
SIMPLE MODEL



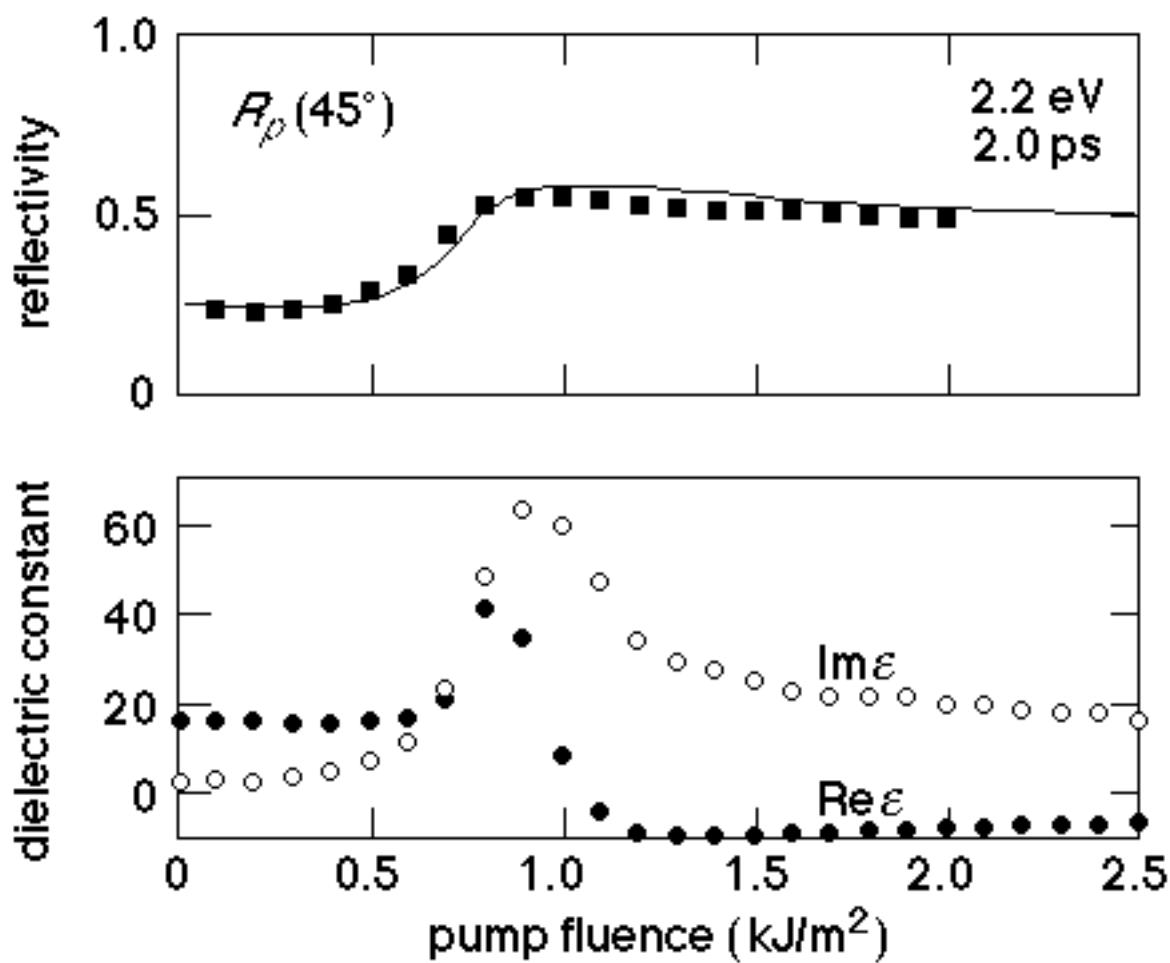
SIMPLE MODEL



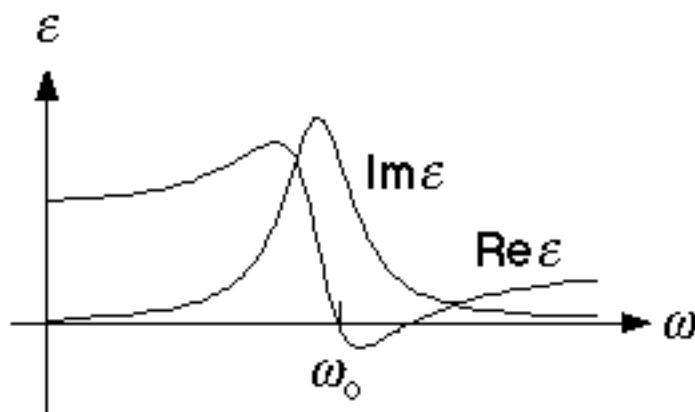
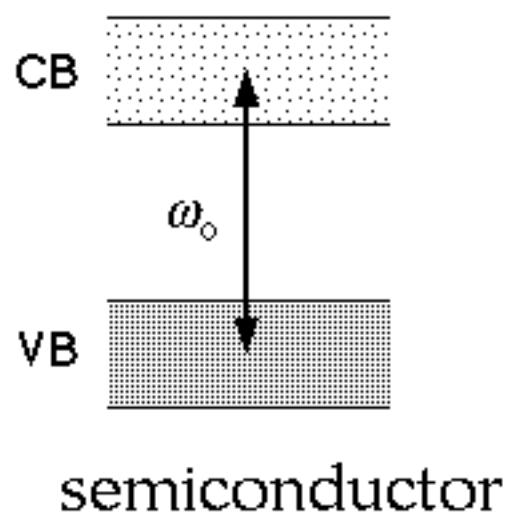
SIMPLE MODEL



SIMPLE MODEL



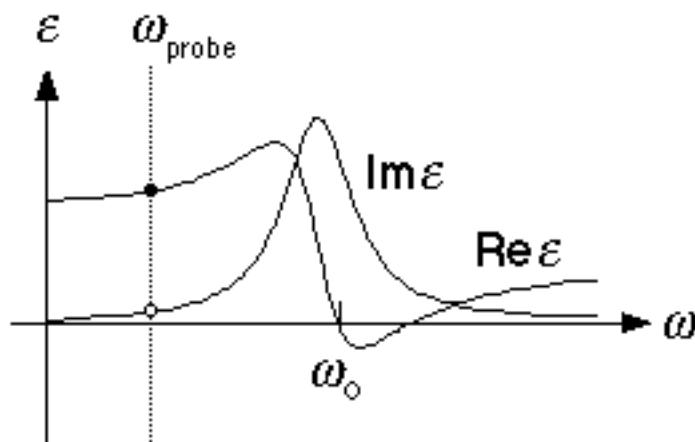
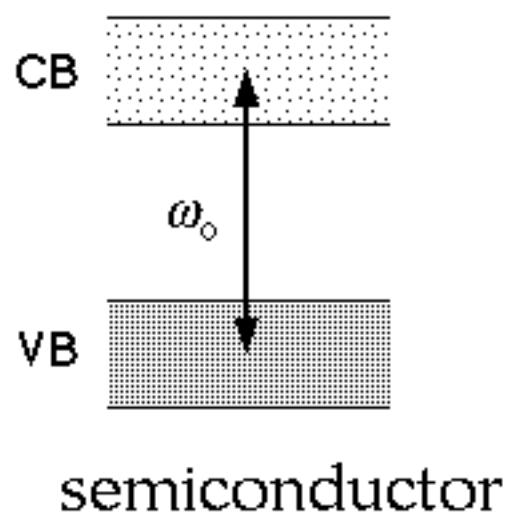
WHAT IS HAPPENING?



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

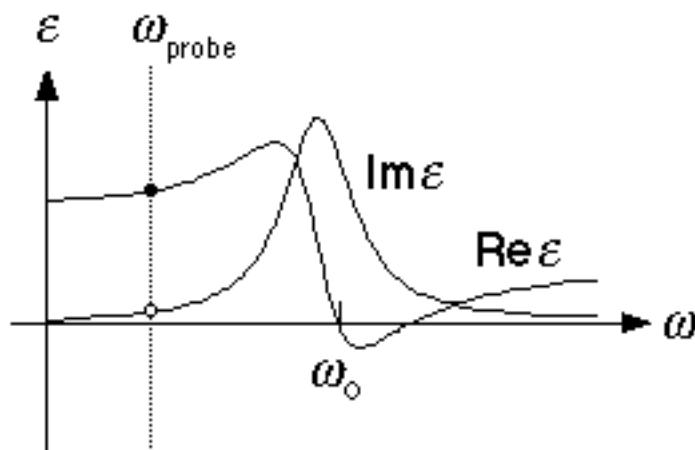
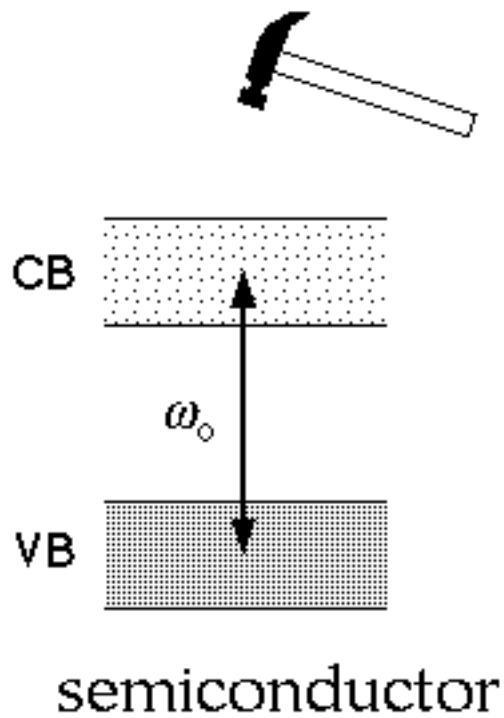


WHAT IS HAPPENING?



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

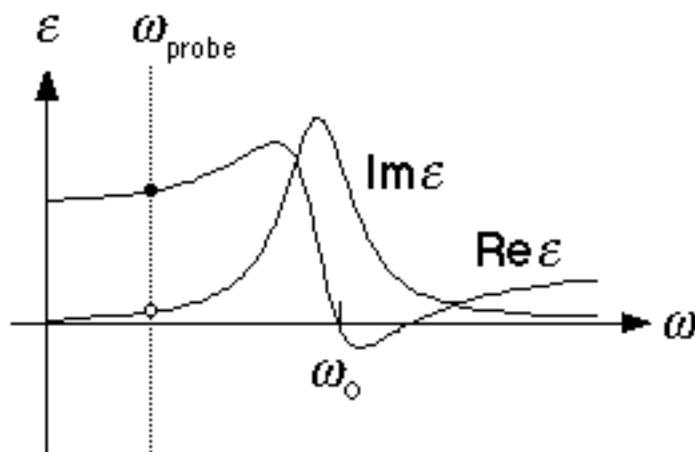
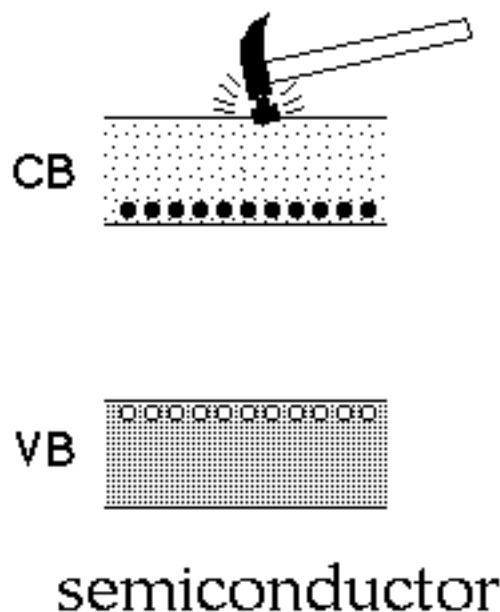
WHAT IS HAPPENING?



pump pulse excites
valence electrons...



WHAT IS HAPPENING?

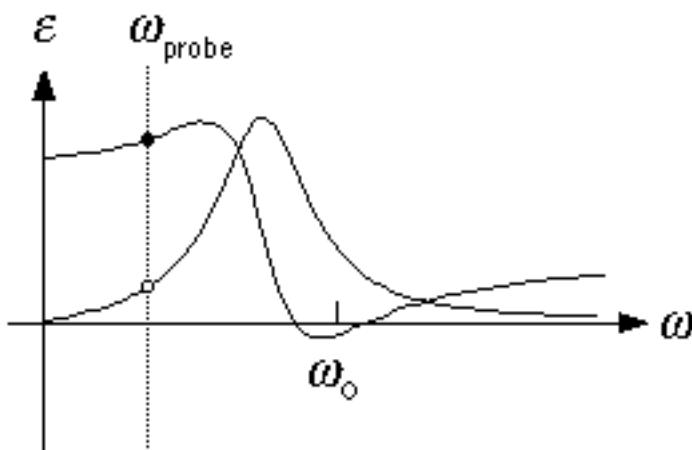
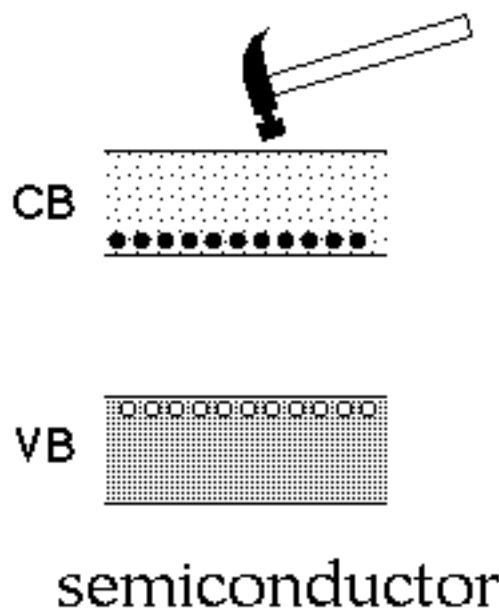


semiconductor

pump pulse excites
valence electrons...



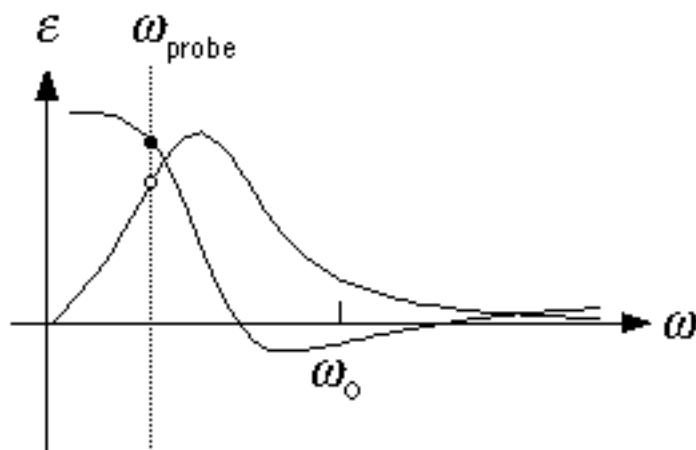
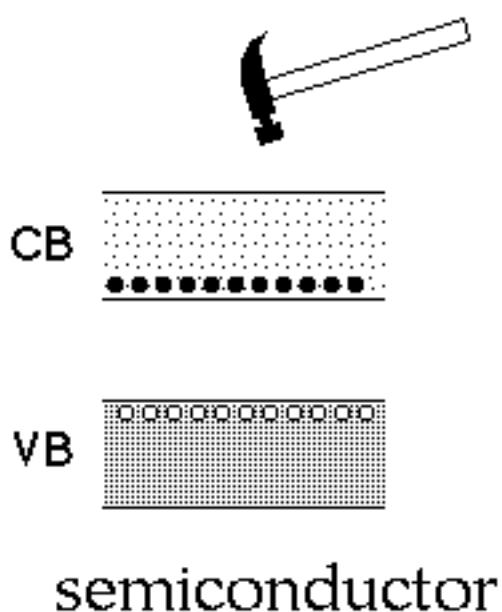
WHAT IS HAPPENING?



...and induces change
in bandstructure



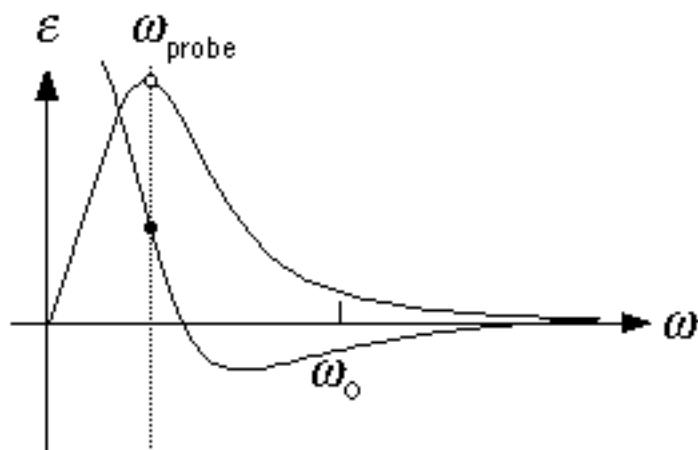
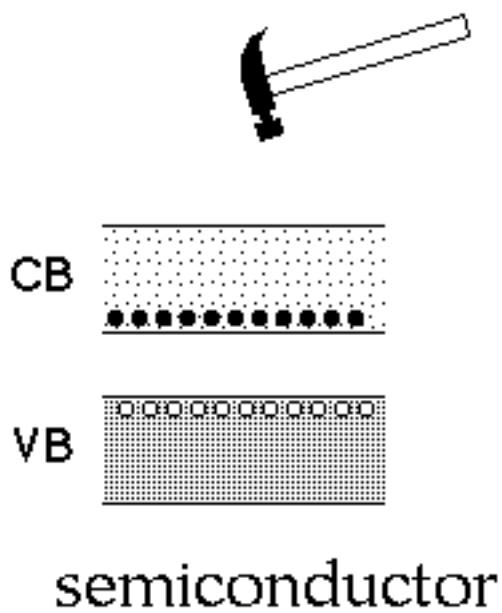
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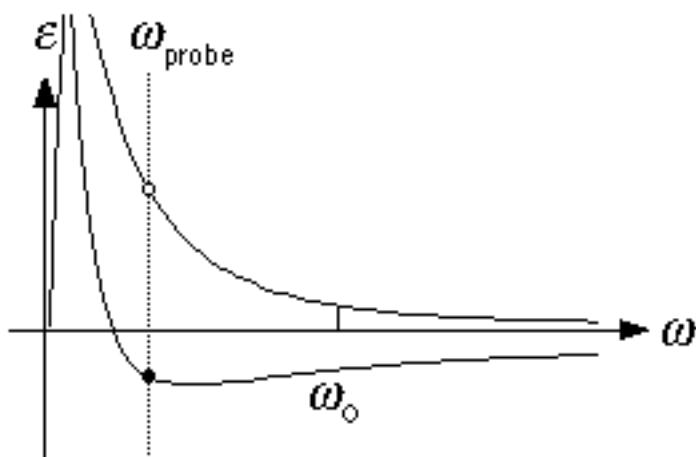
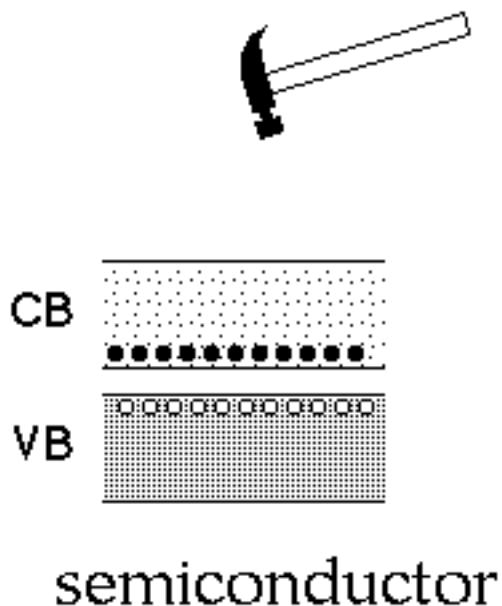
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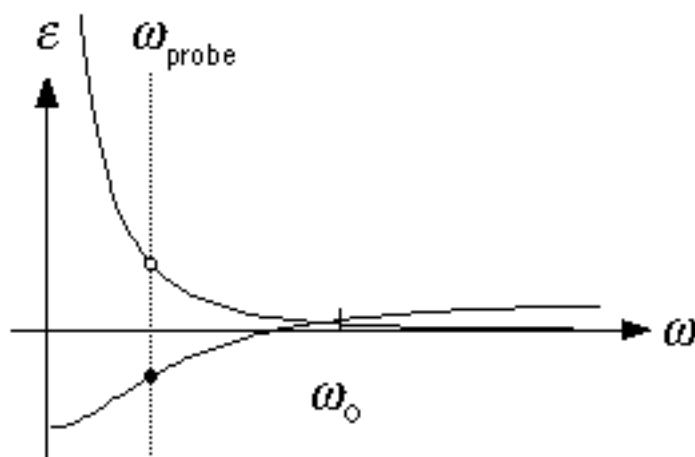
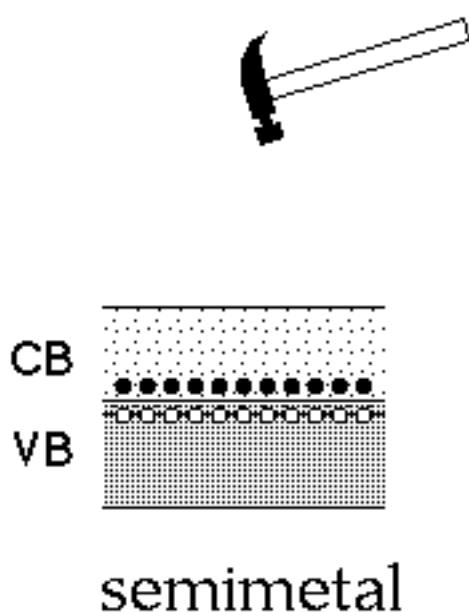
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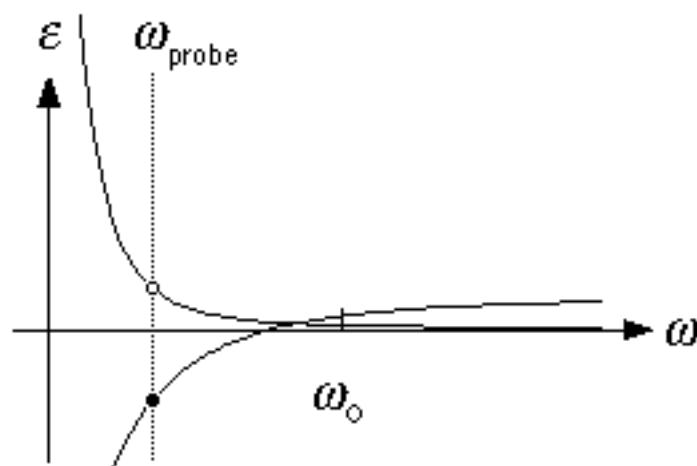
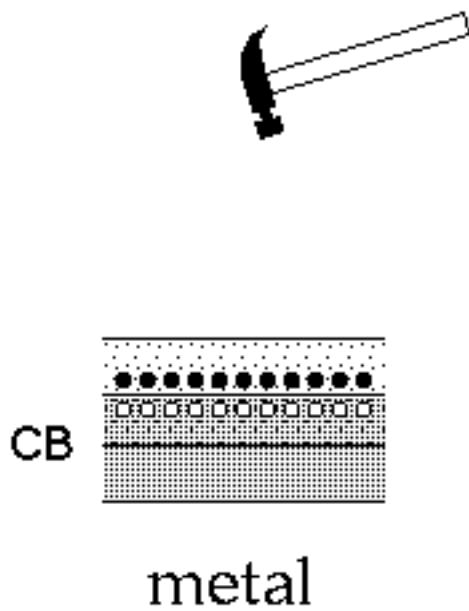
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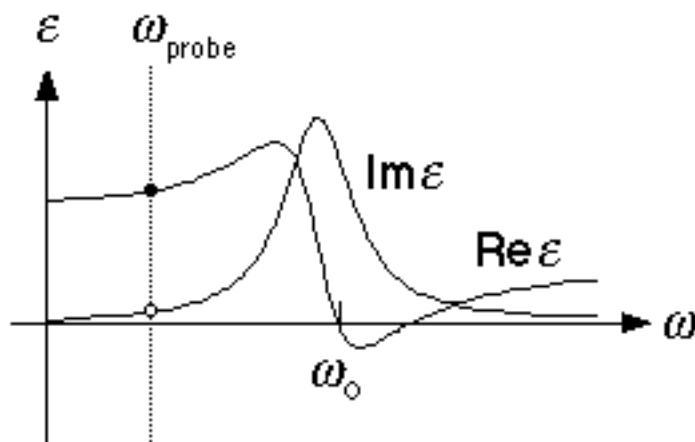
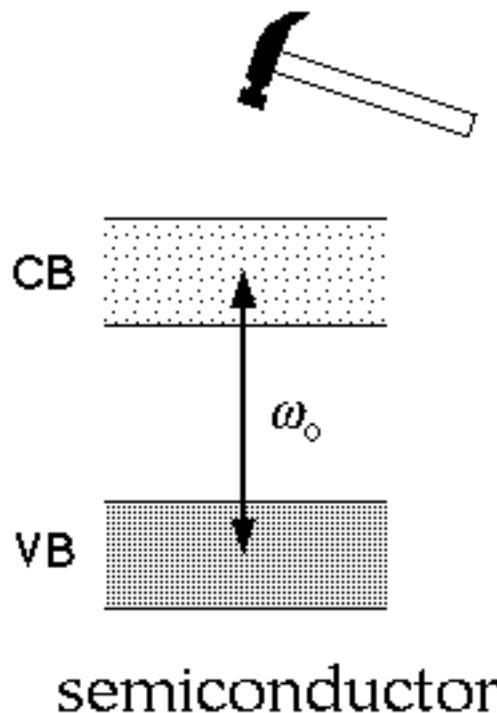
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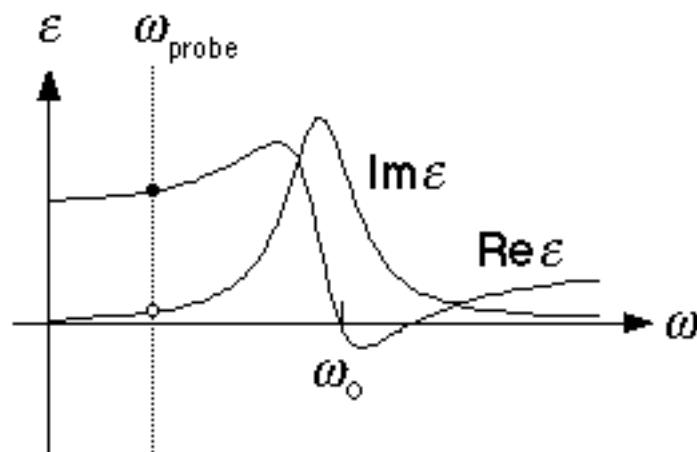
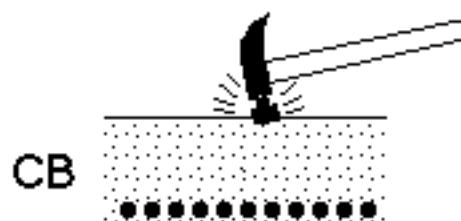
...and induces change
in bandstructure



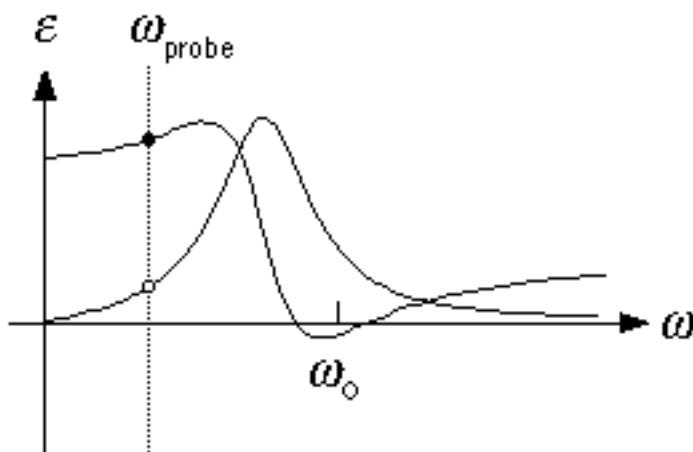
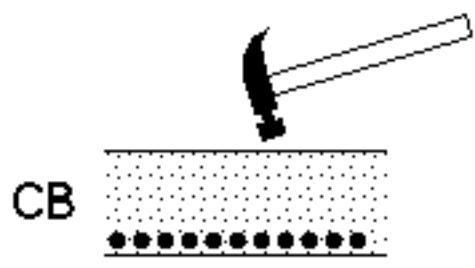
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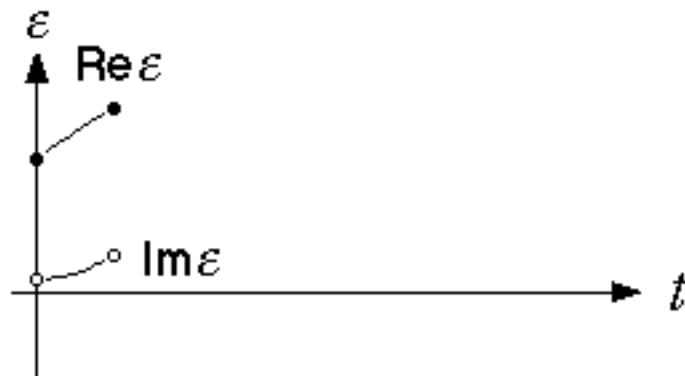
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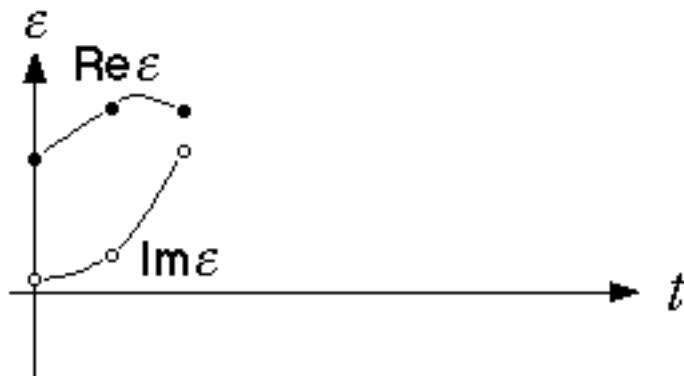
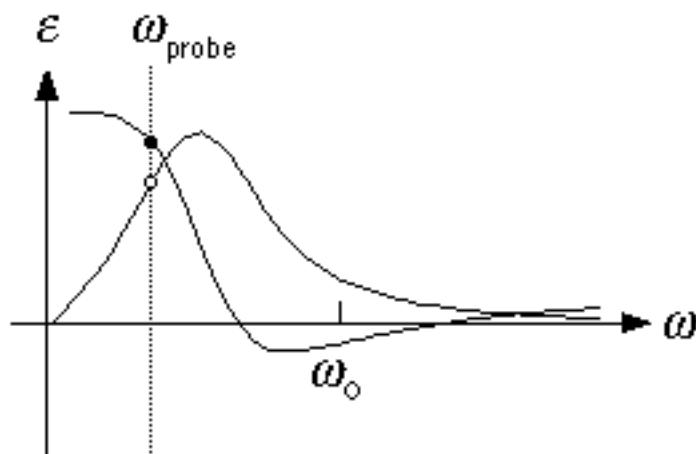
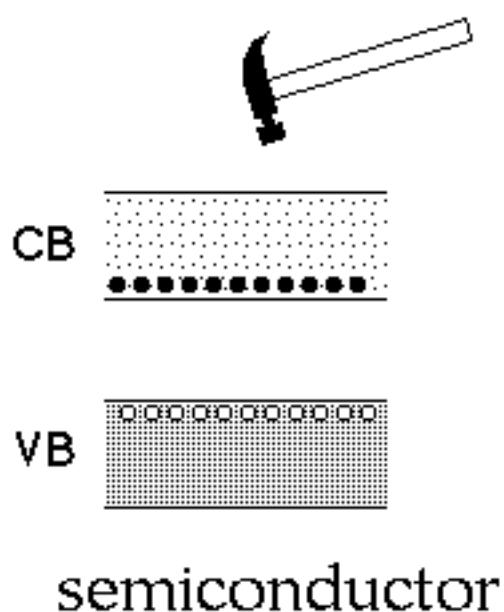
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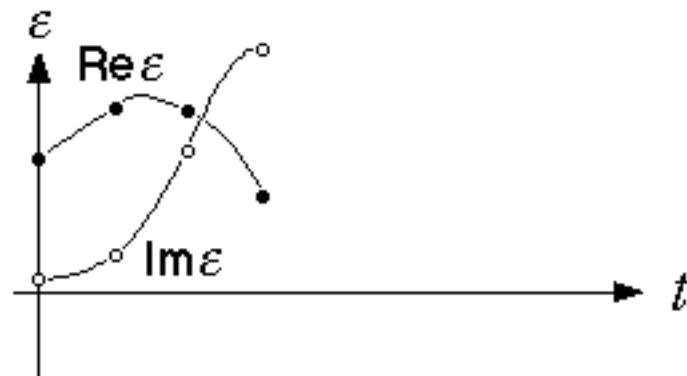
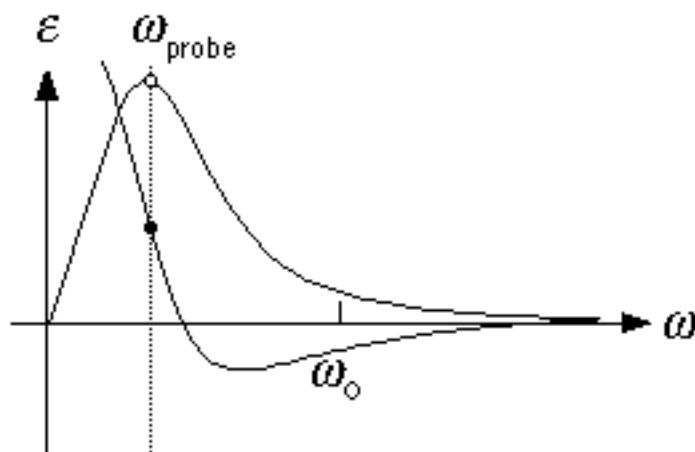
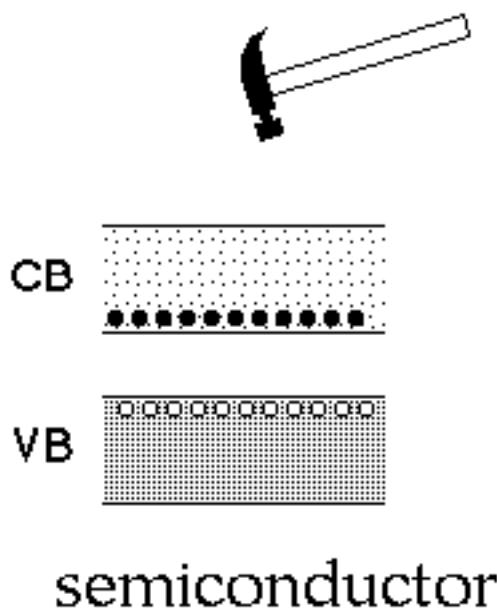
VB
semiconductor



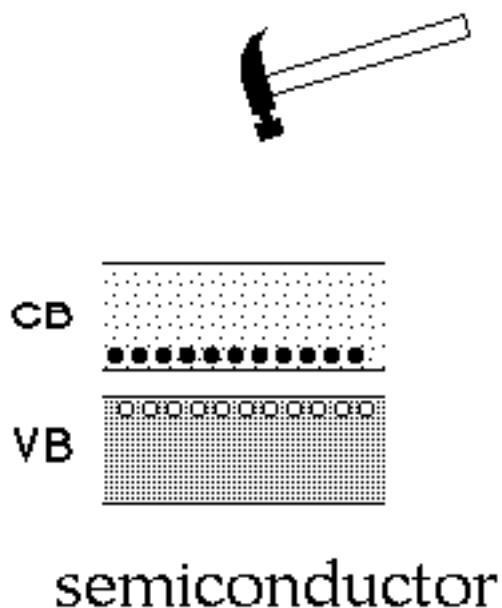
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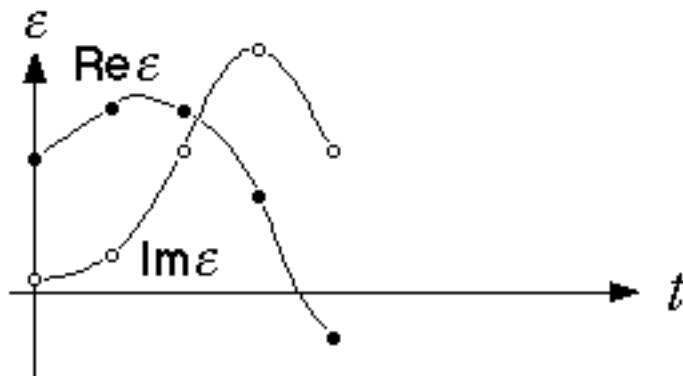
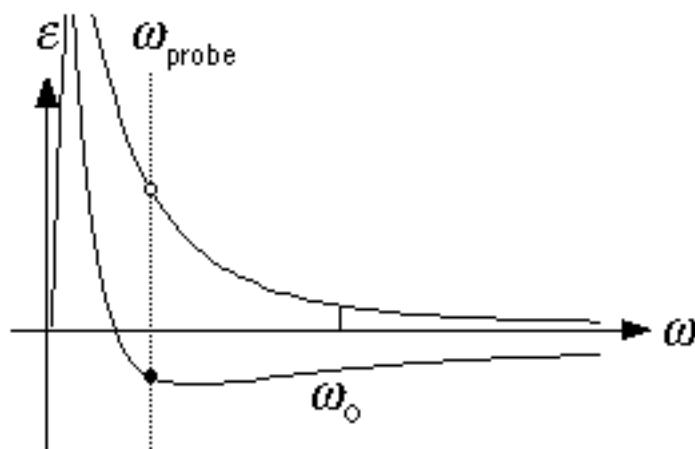
WHAT IS HAPPENING?



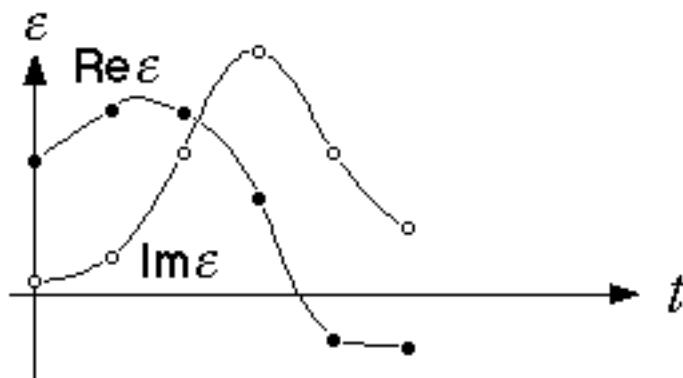
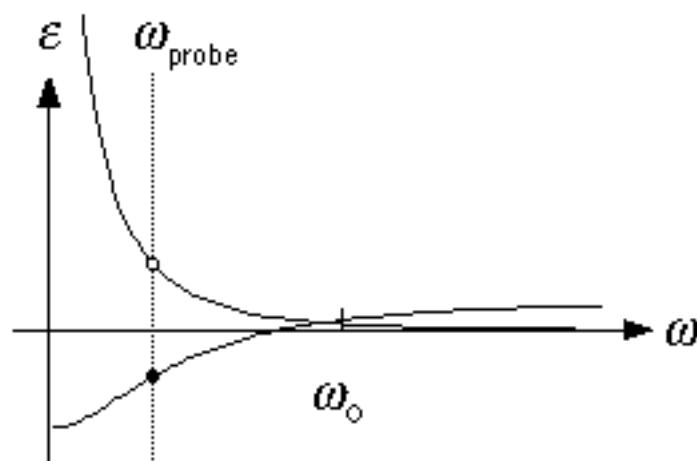
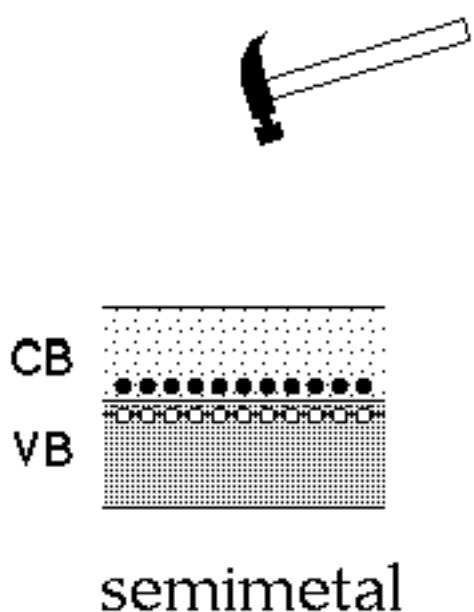
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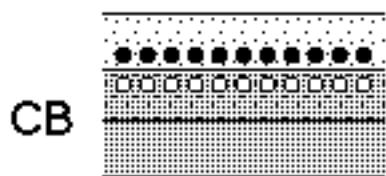
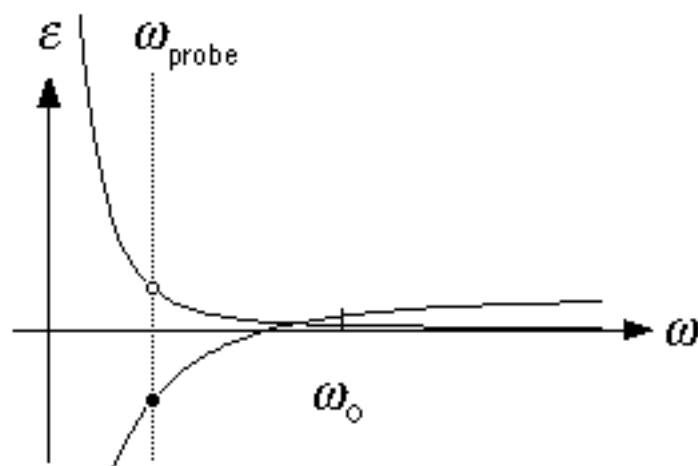
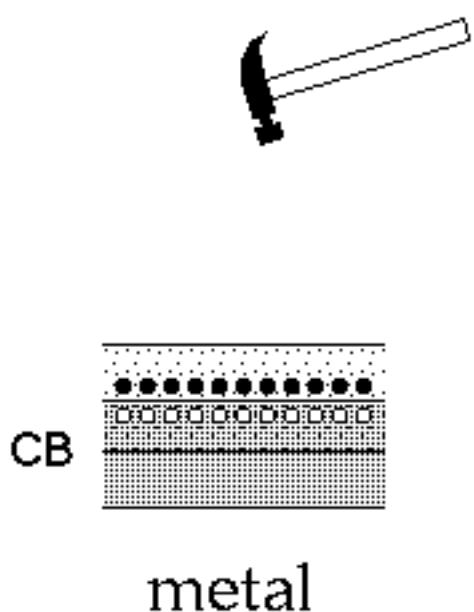
semiconductor



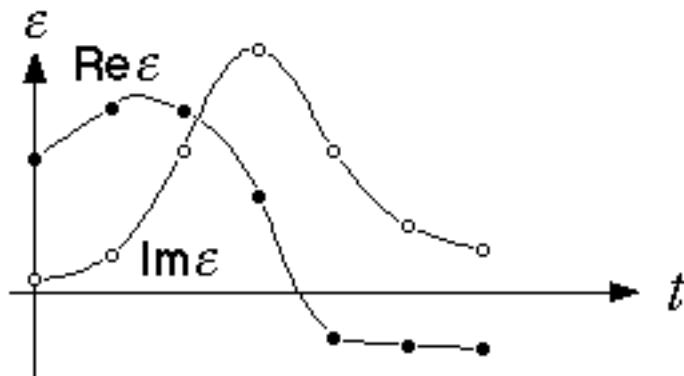
WHAT IS HAPPENING?



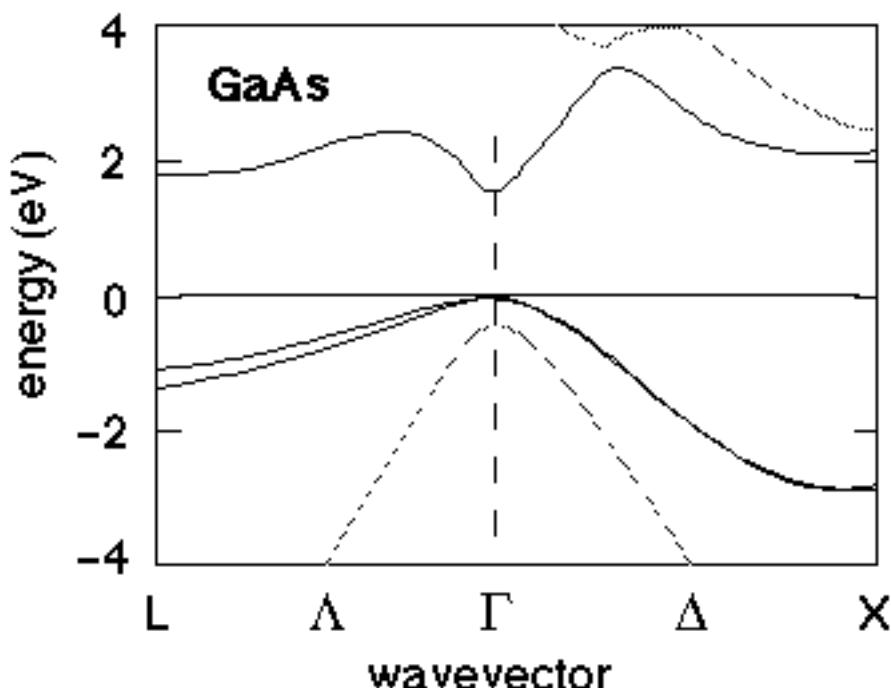
WHAT IS HAPPENING?



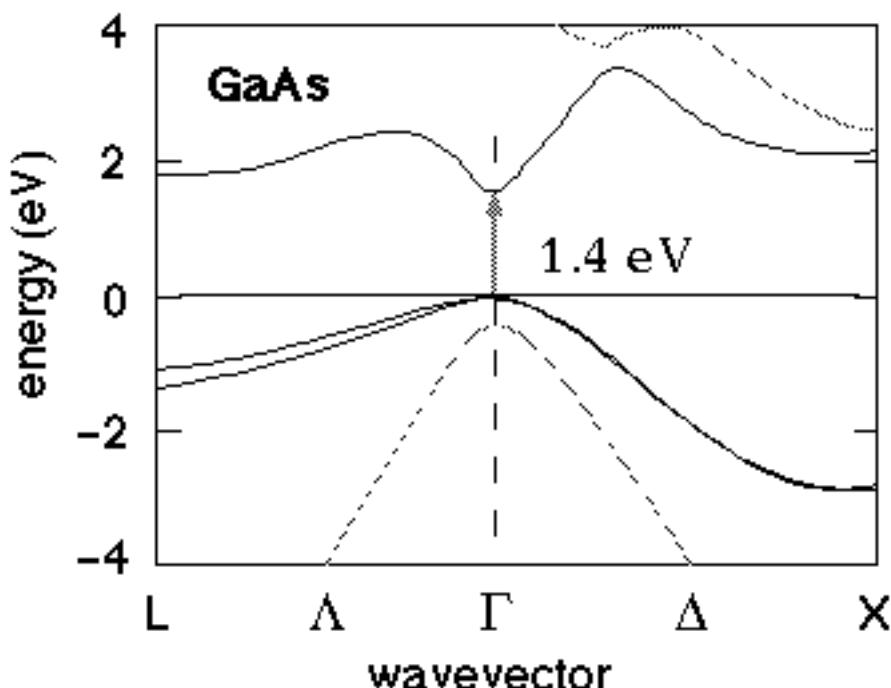
metal



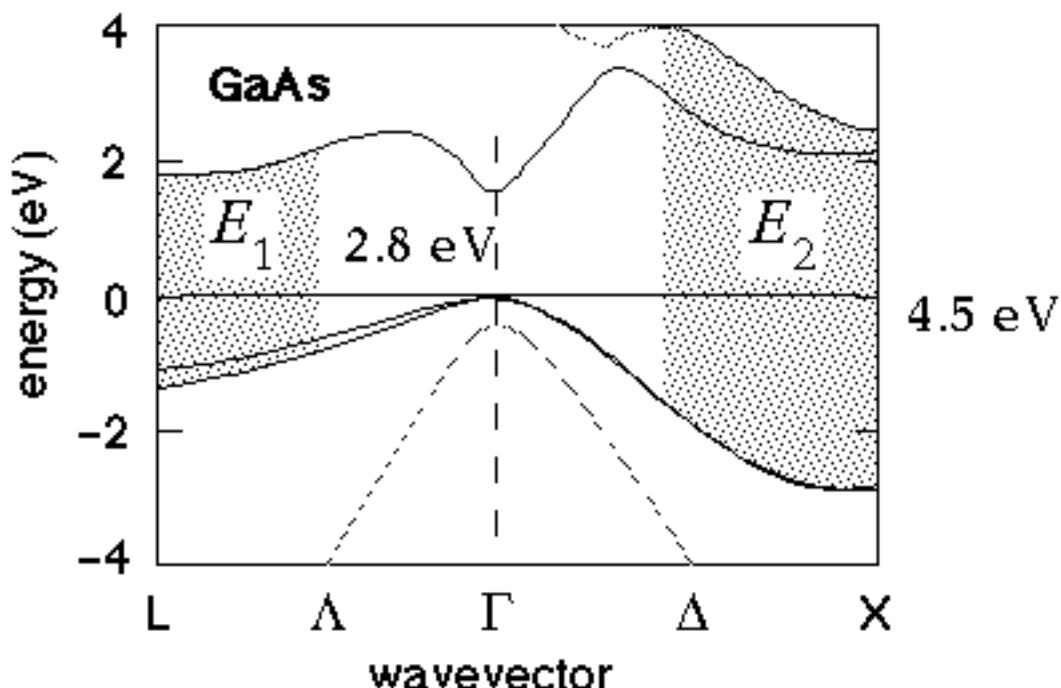
GaAs BANDSTRUCTURE



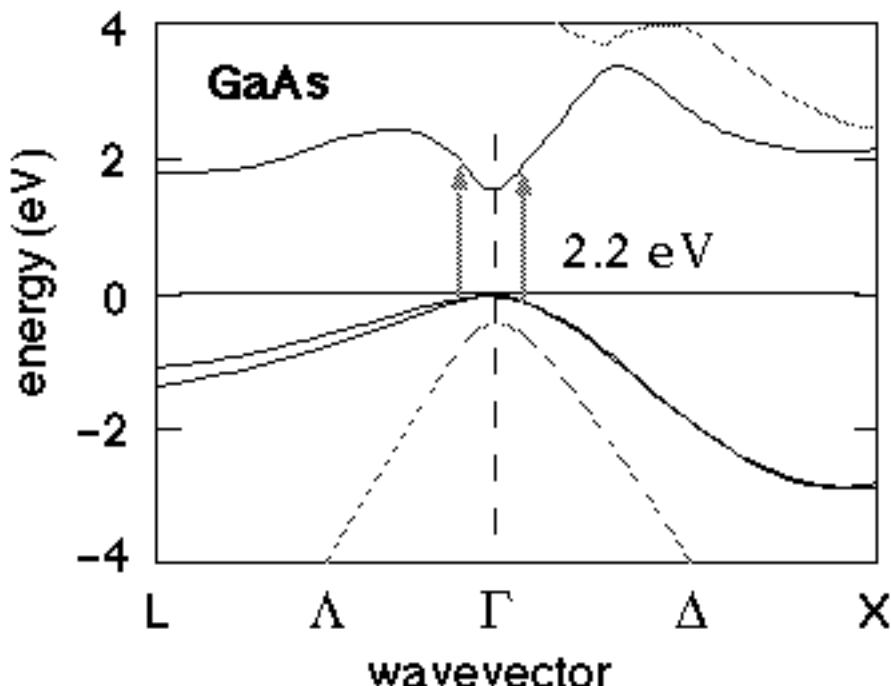
GaAs BANDSTRUCTURE



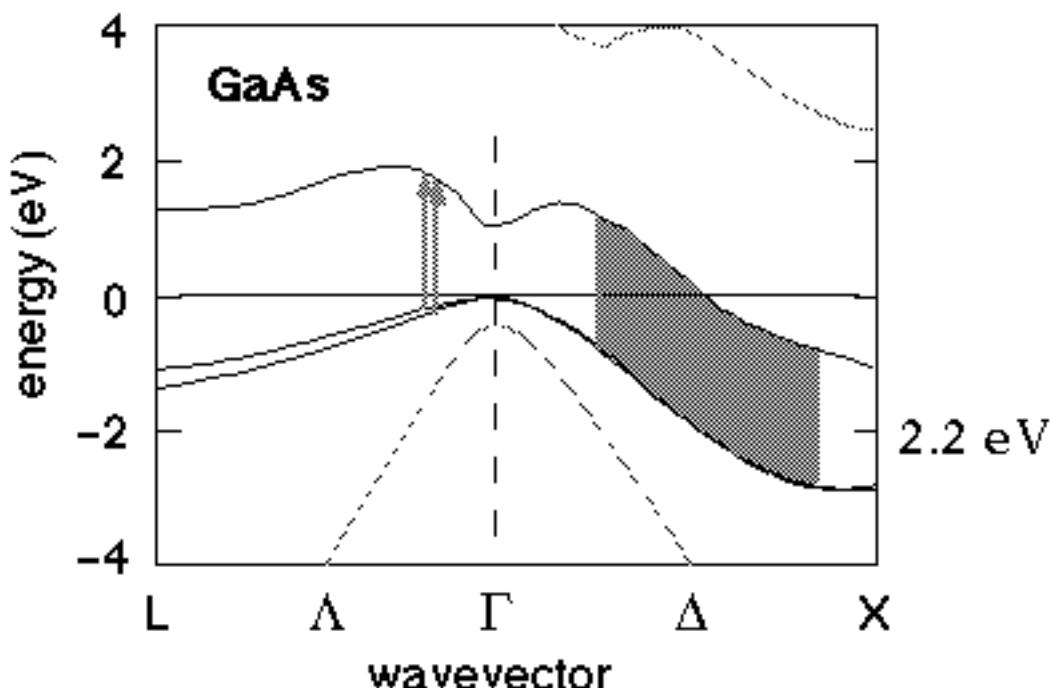
GaAs BANDSTRUCTURE



GaAs BANDSTRUCTURE



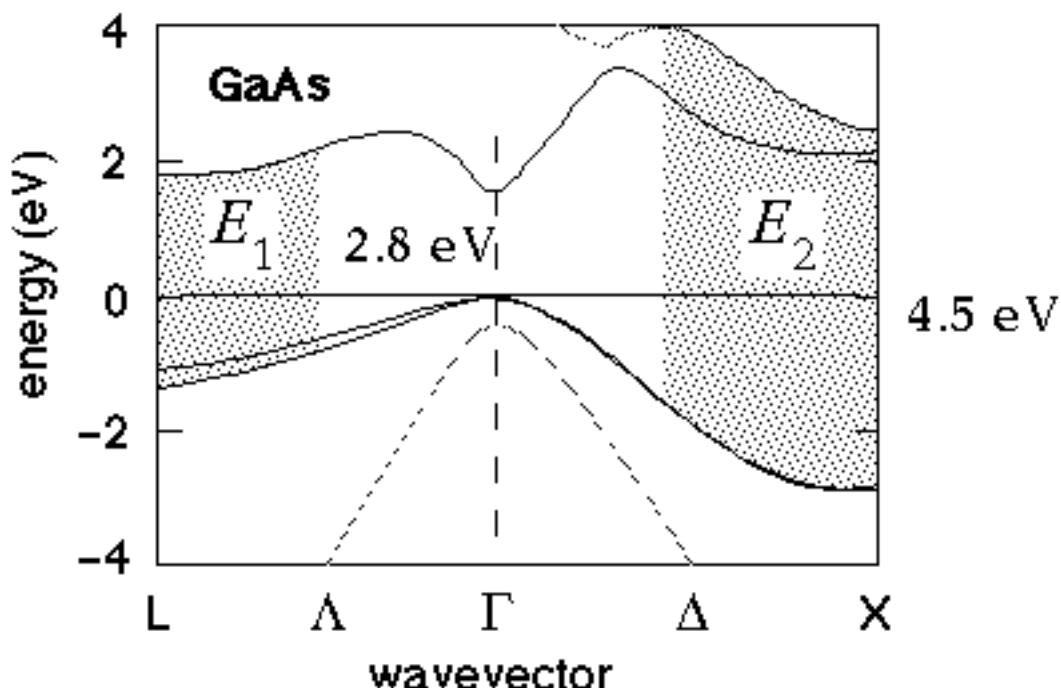
GaAs BANDSTRUCTURE



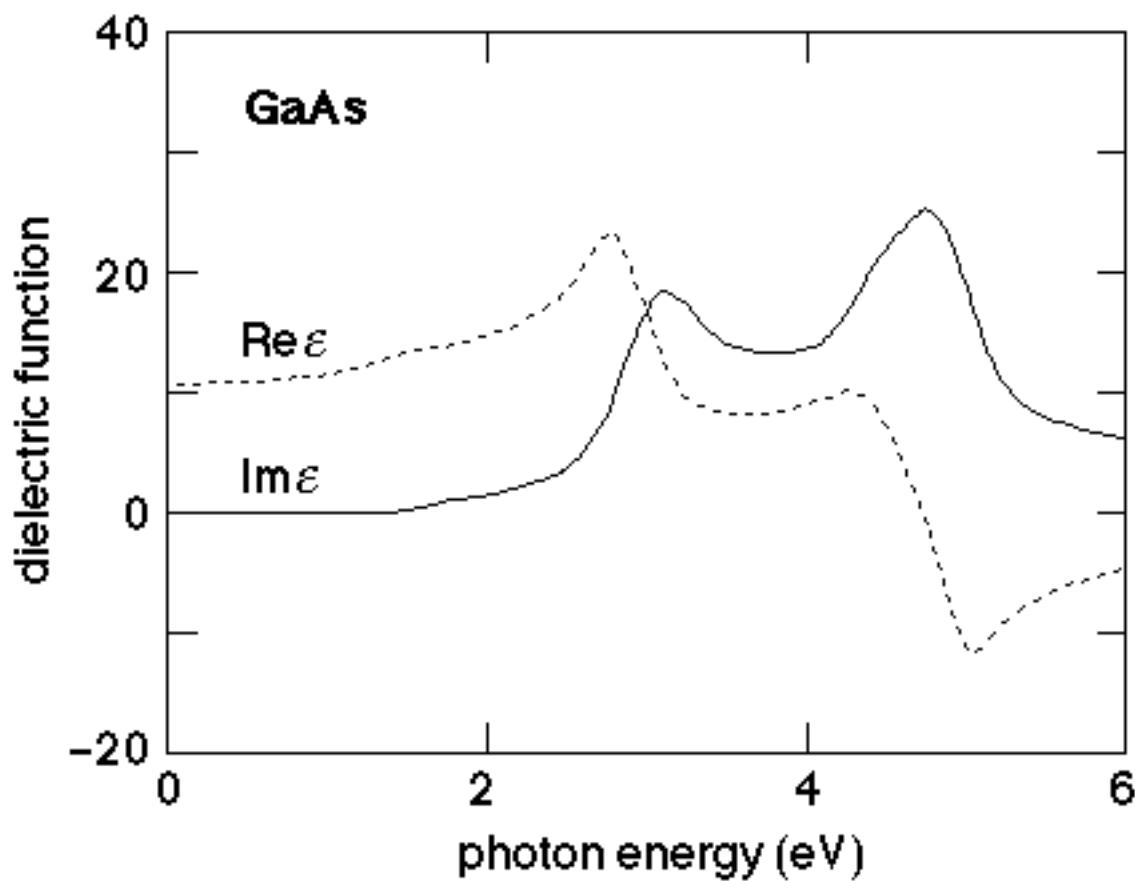
S. Froyen *et al.*, Phys Rev. B **28** (1983) 3258

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

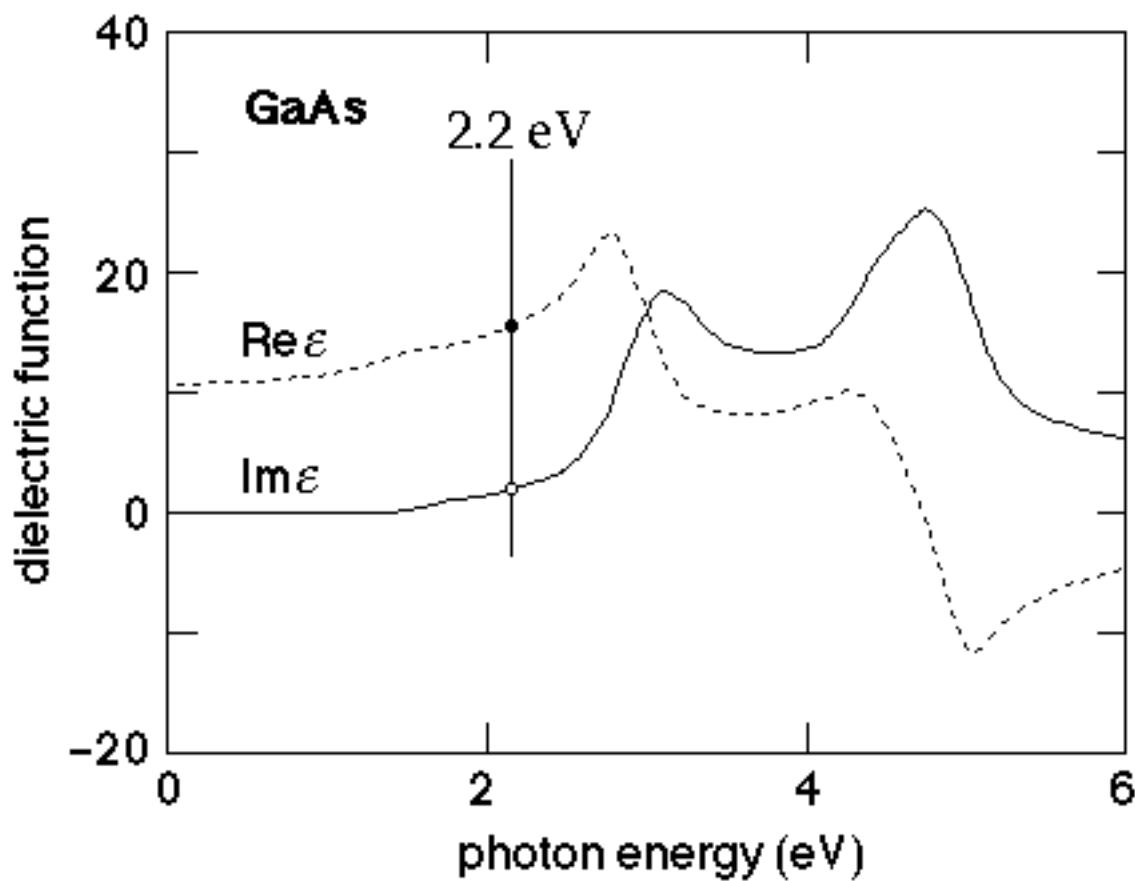
GaAs BANDSTRUCTURE



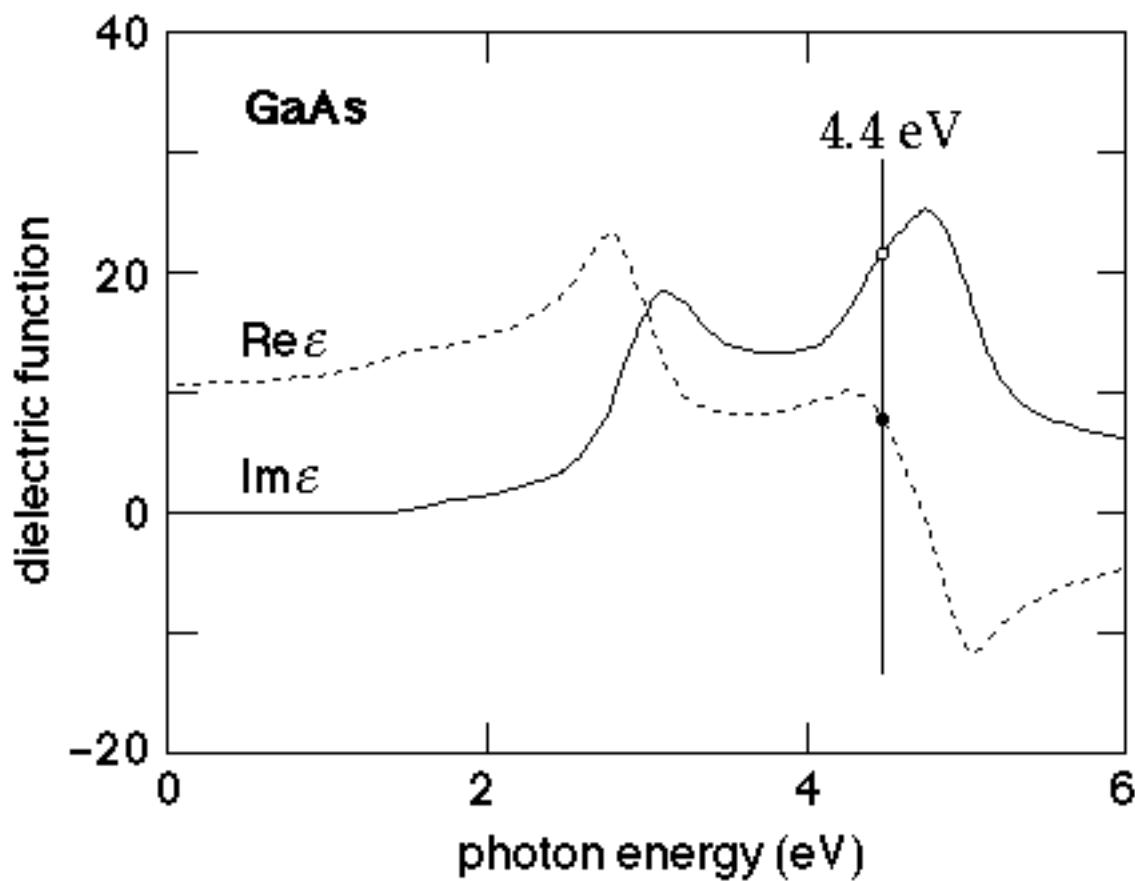
GaAs DIELECTRIC FUNCTION



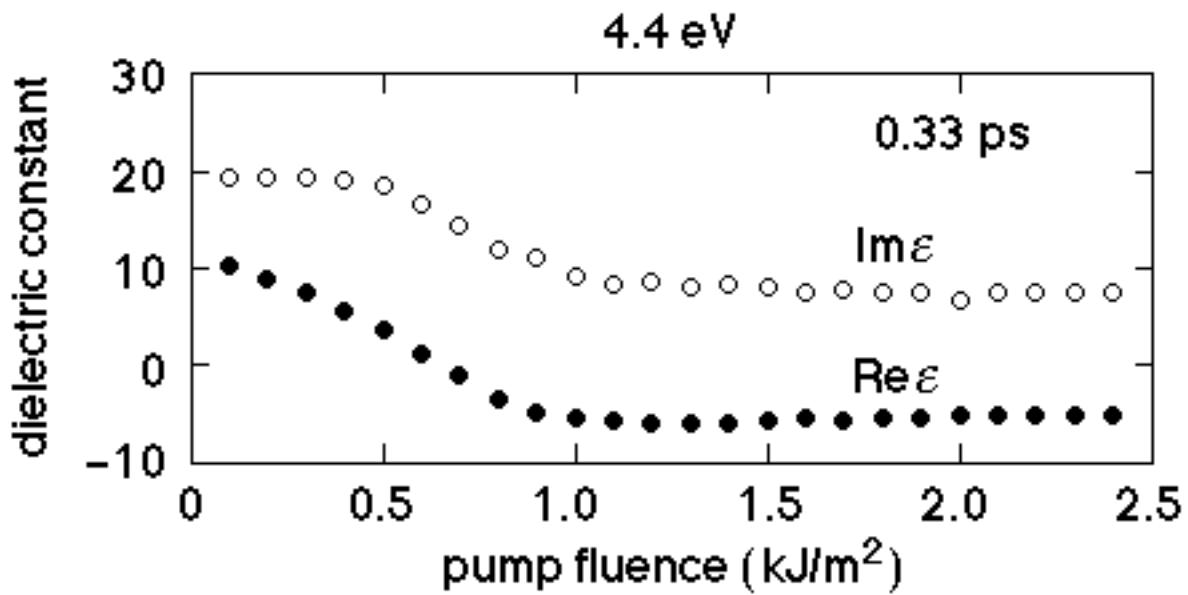
GaAs DIELECTRIC FUNCTION



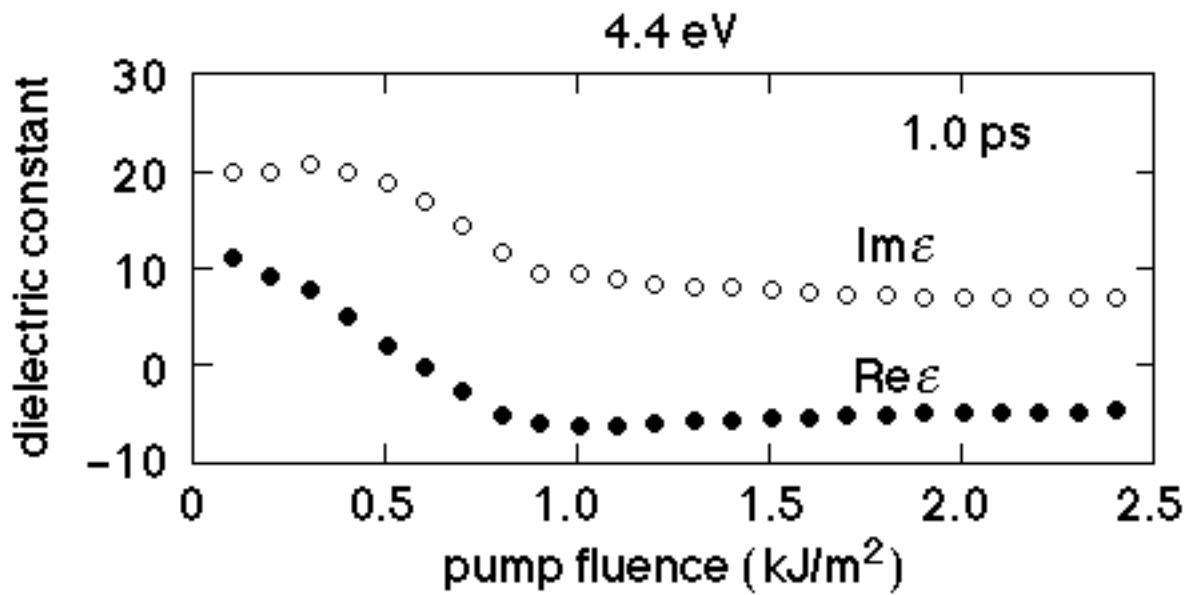
GaAs DIELECTRIC FUNCTION



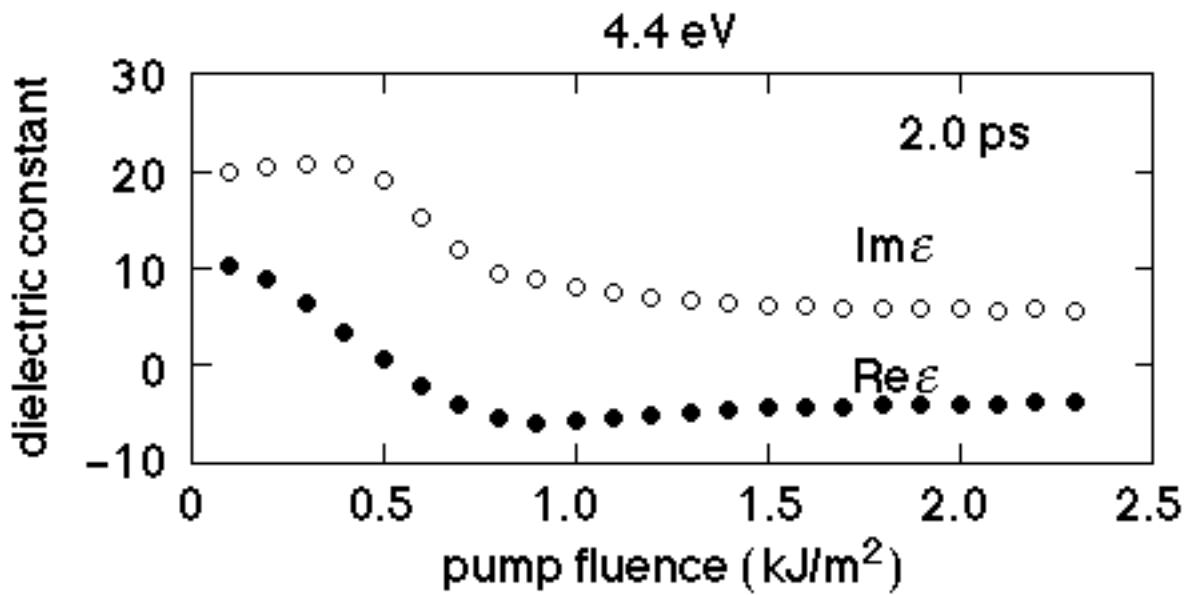
FLUENCE DEPENDENCE



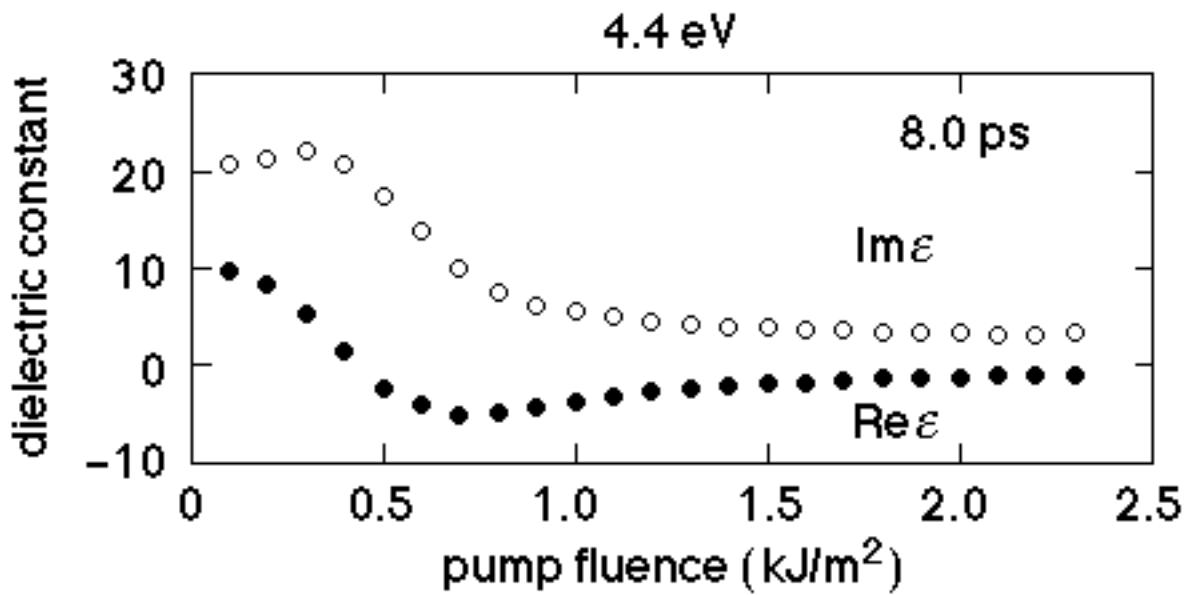
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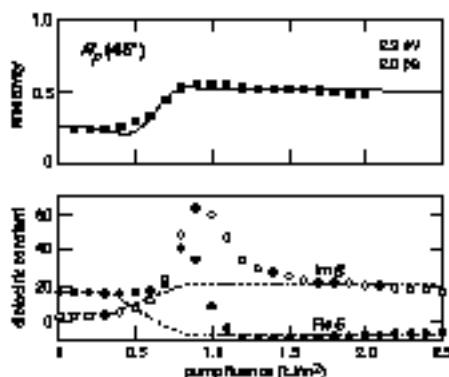
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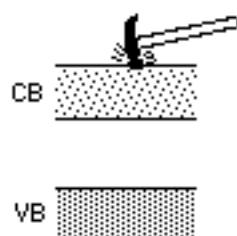
FLUENCE DEPENDENCE



SUMMARY

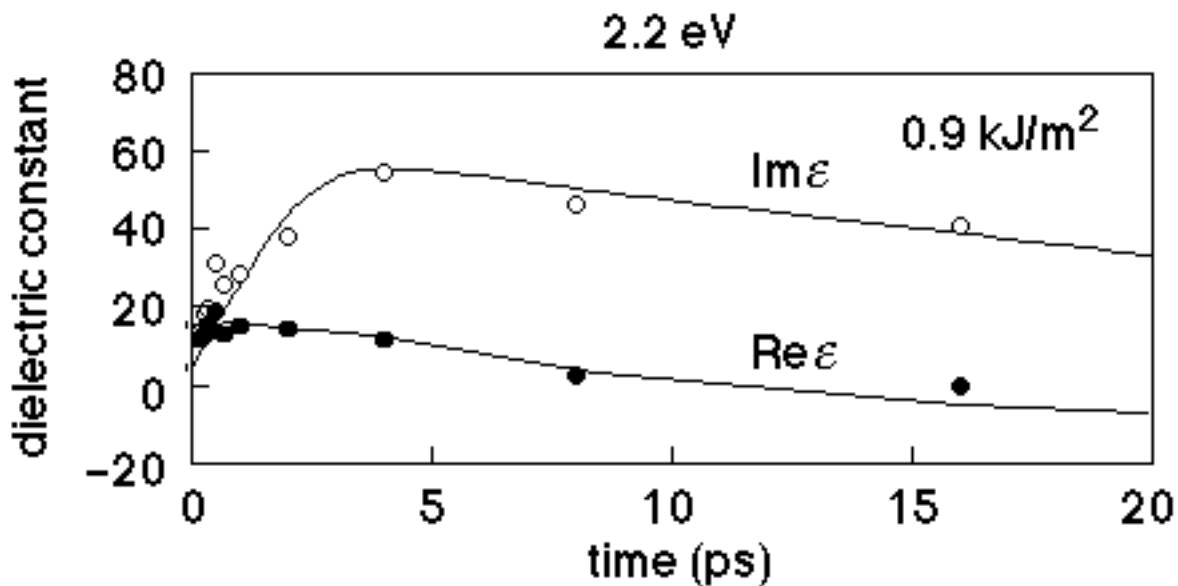


single-angle reflectivity not a good indicator of what happens



laser pulse causes bandgap collapse

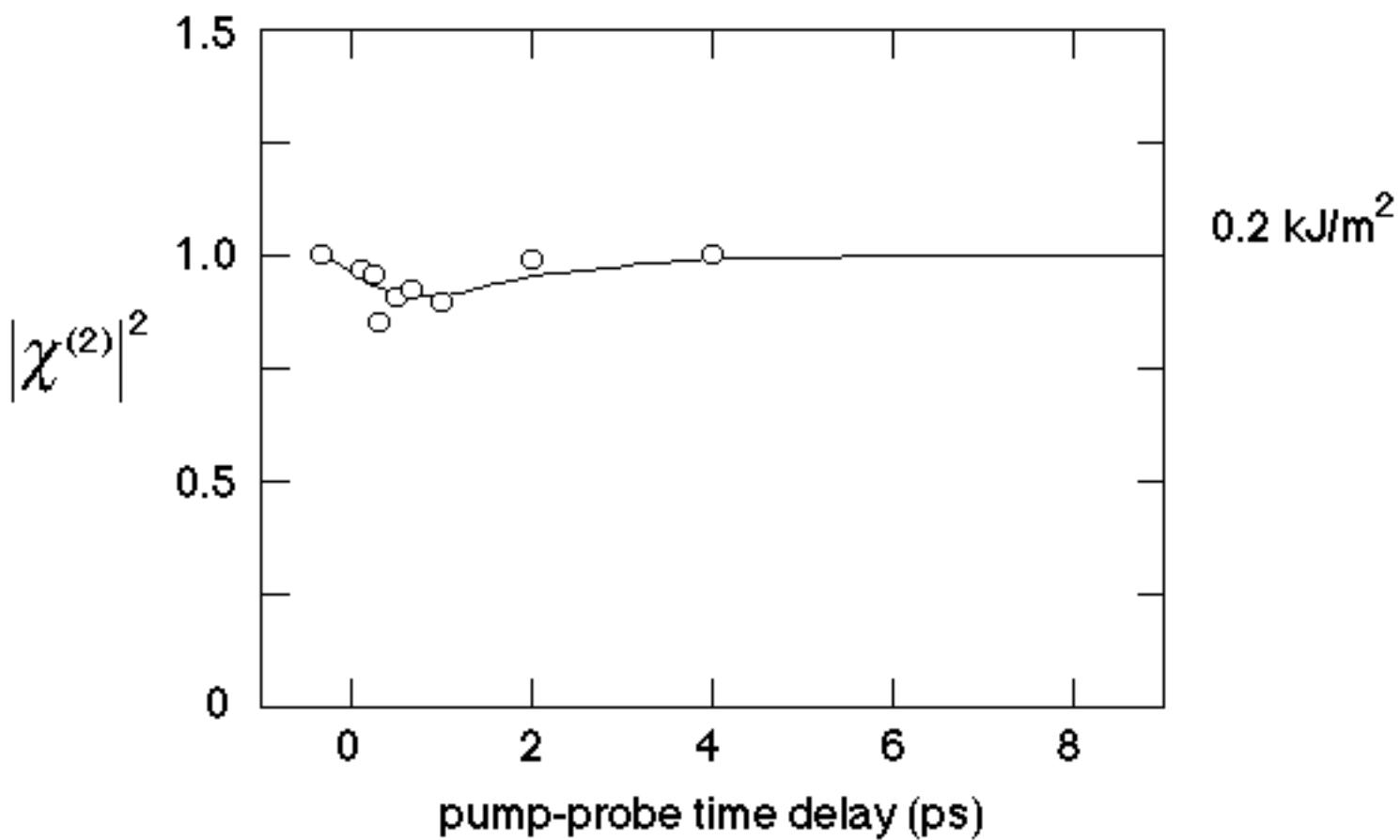
CAUSE OF COLLAPSE



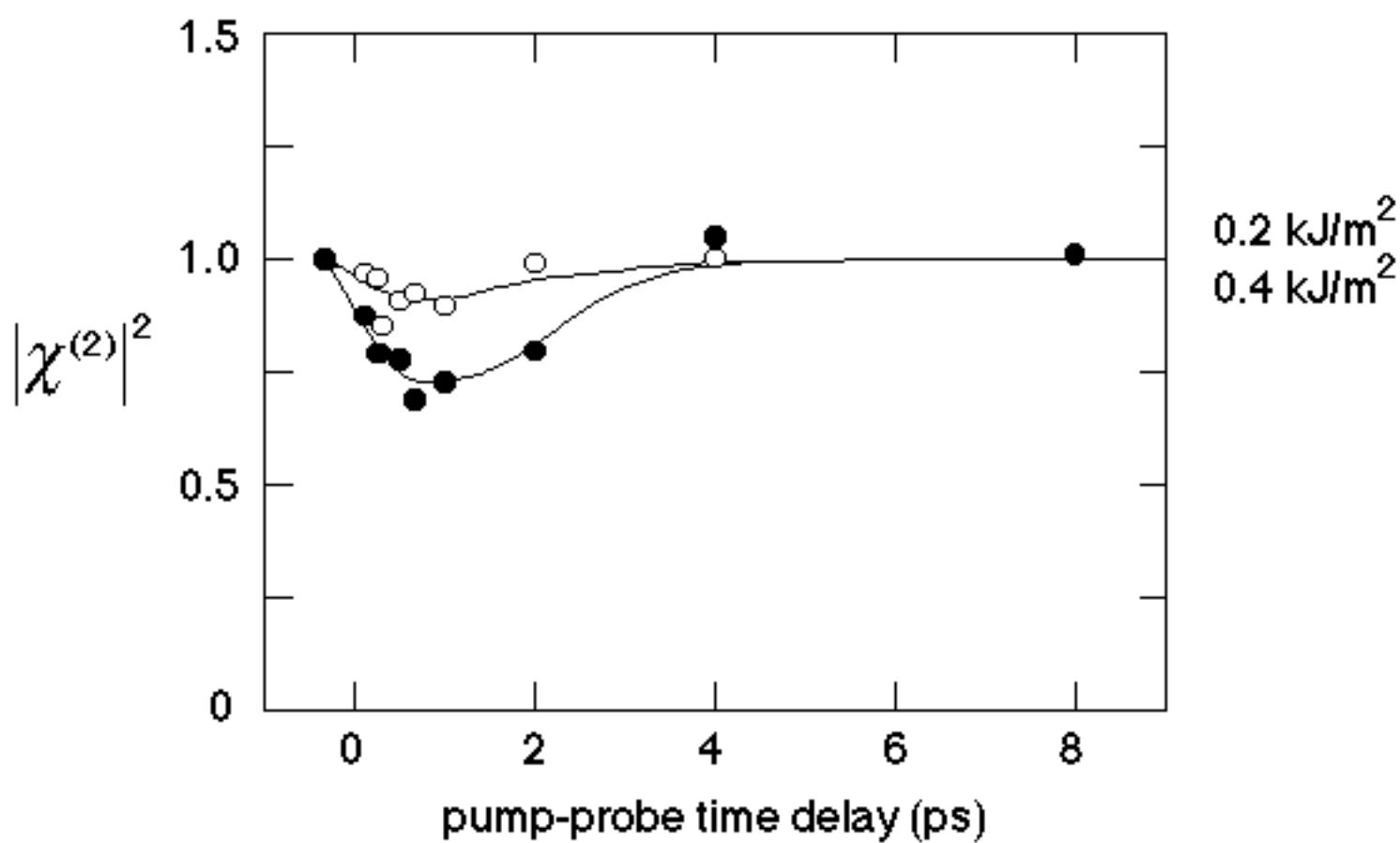
slow rise time → collapse cannot be caused by electronic effects alone
structural change?



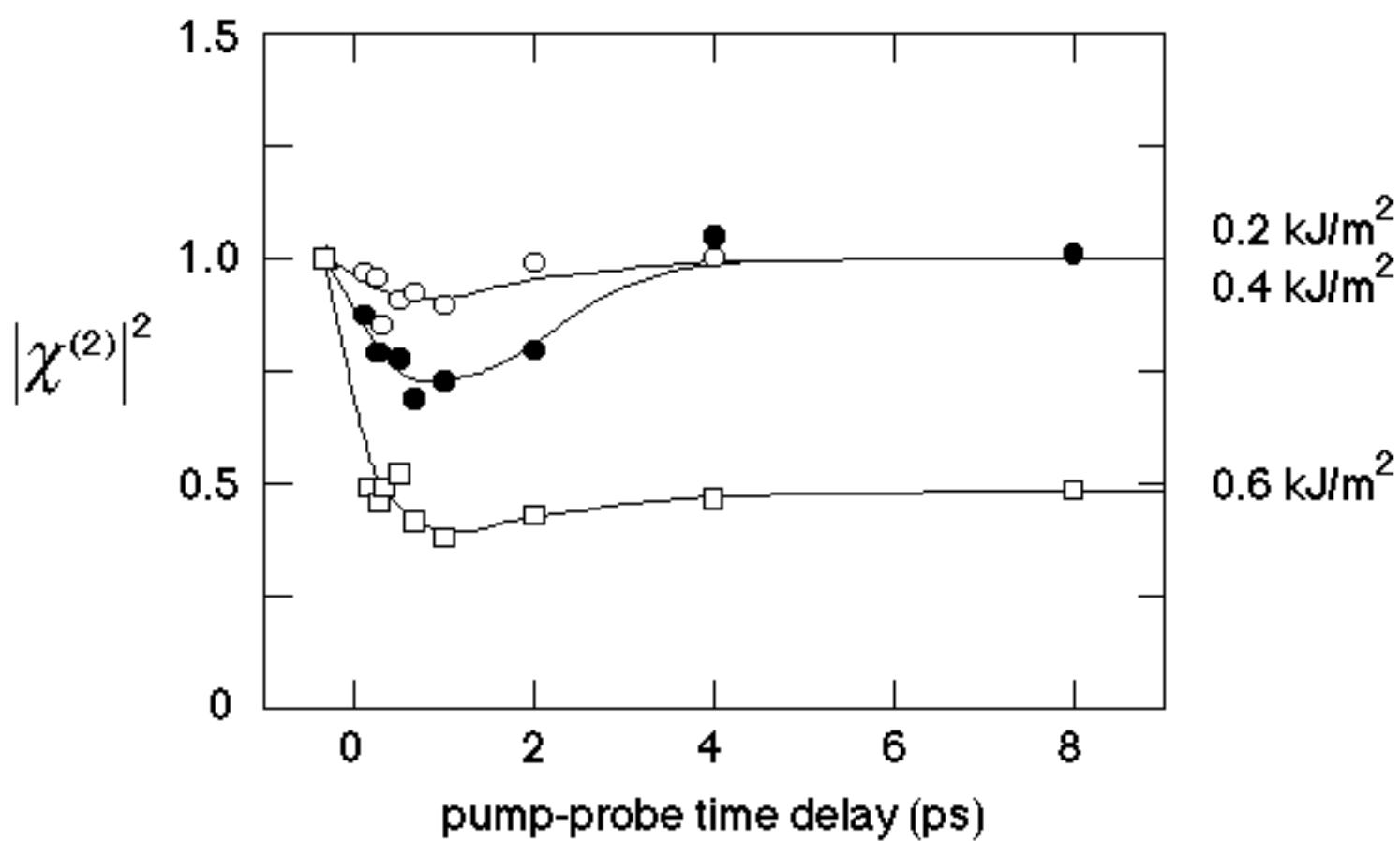
SECOND HARMONIC DATA



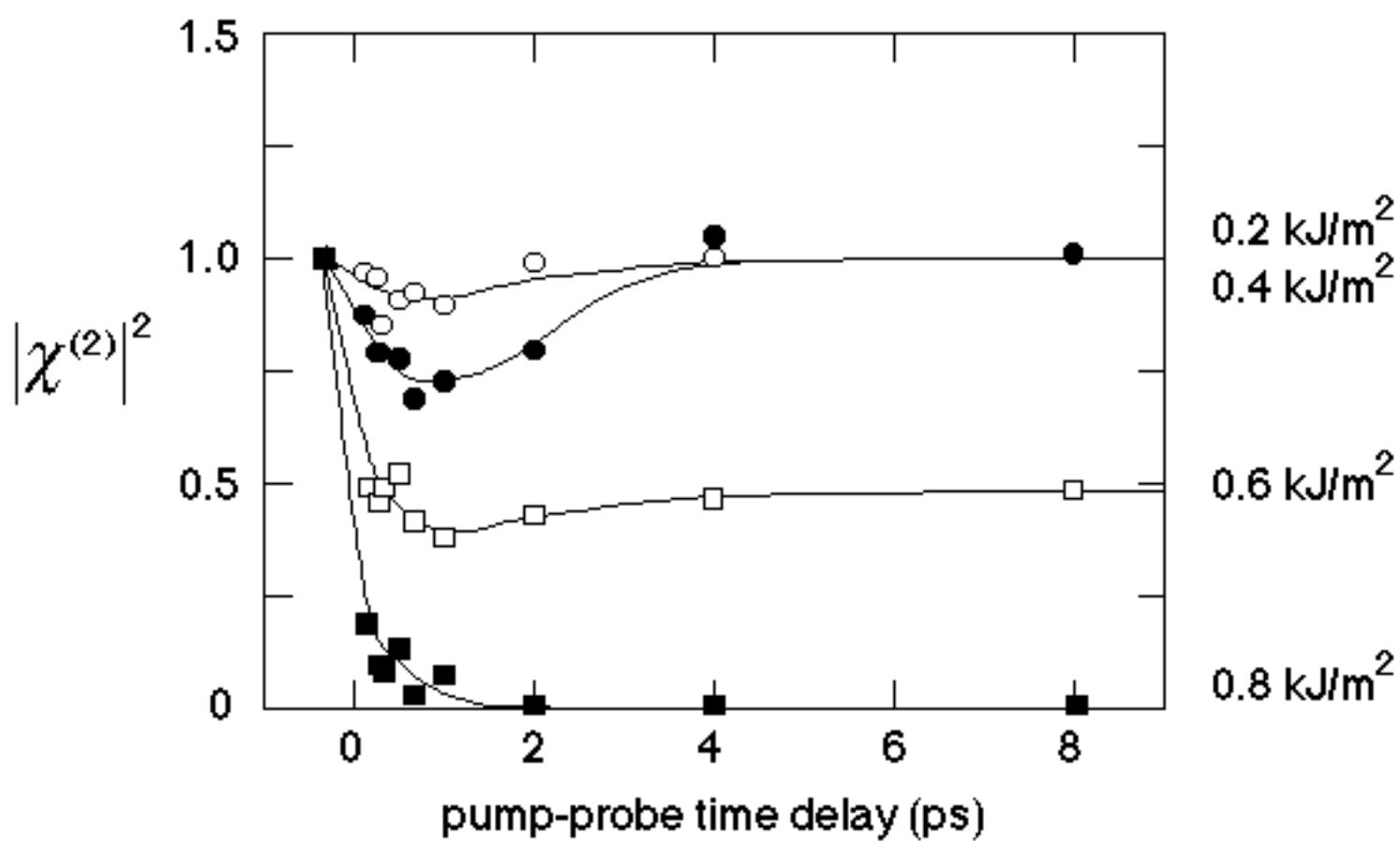
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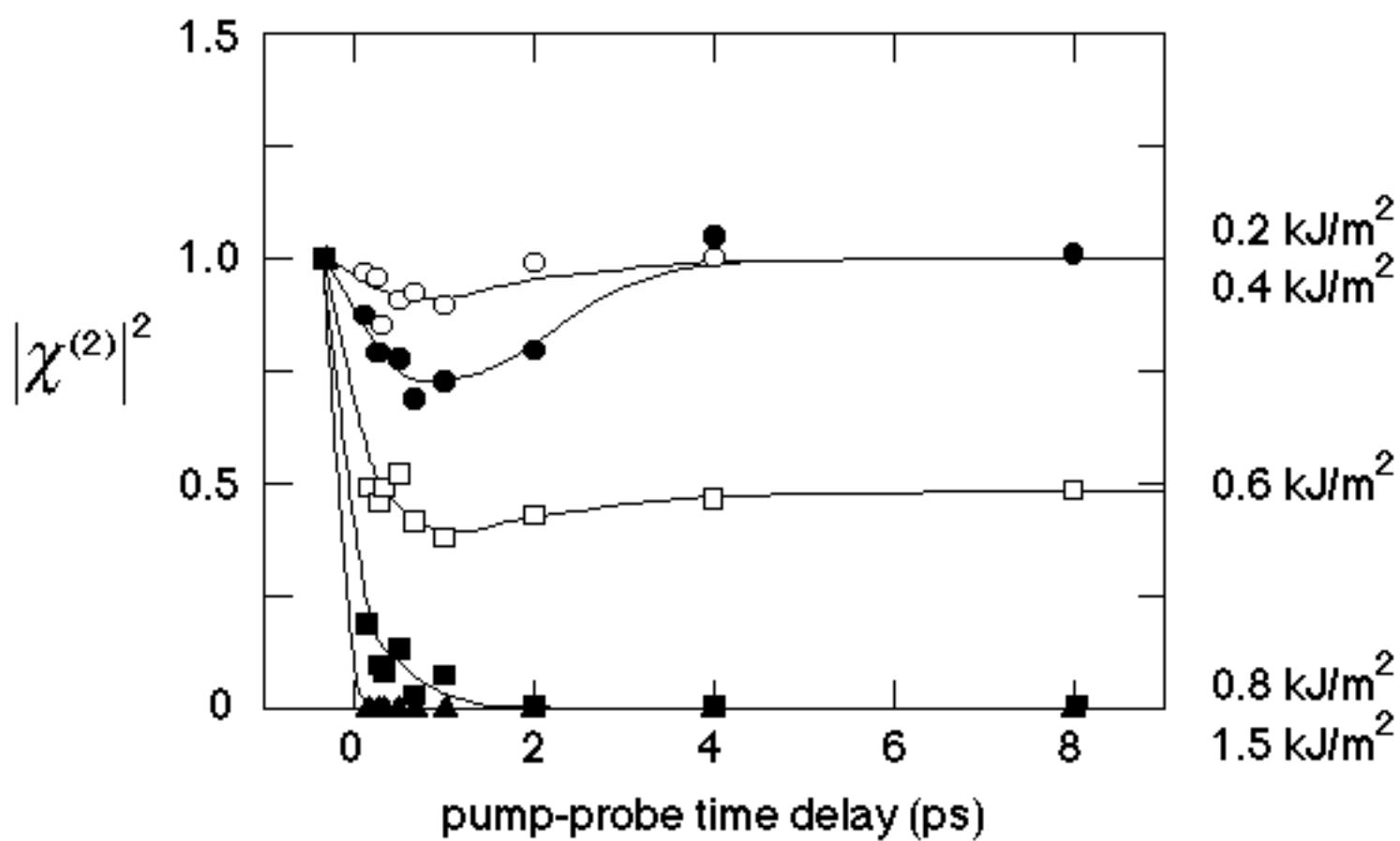
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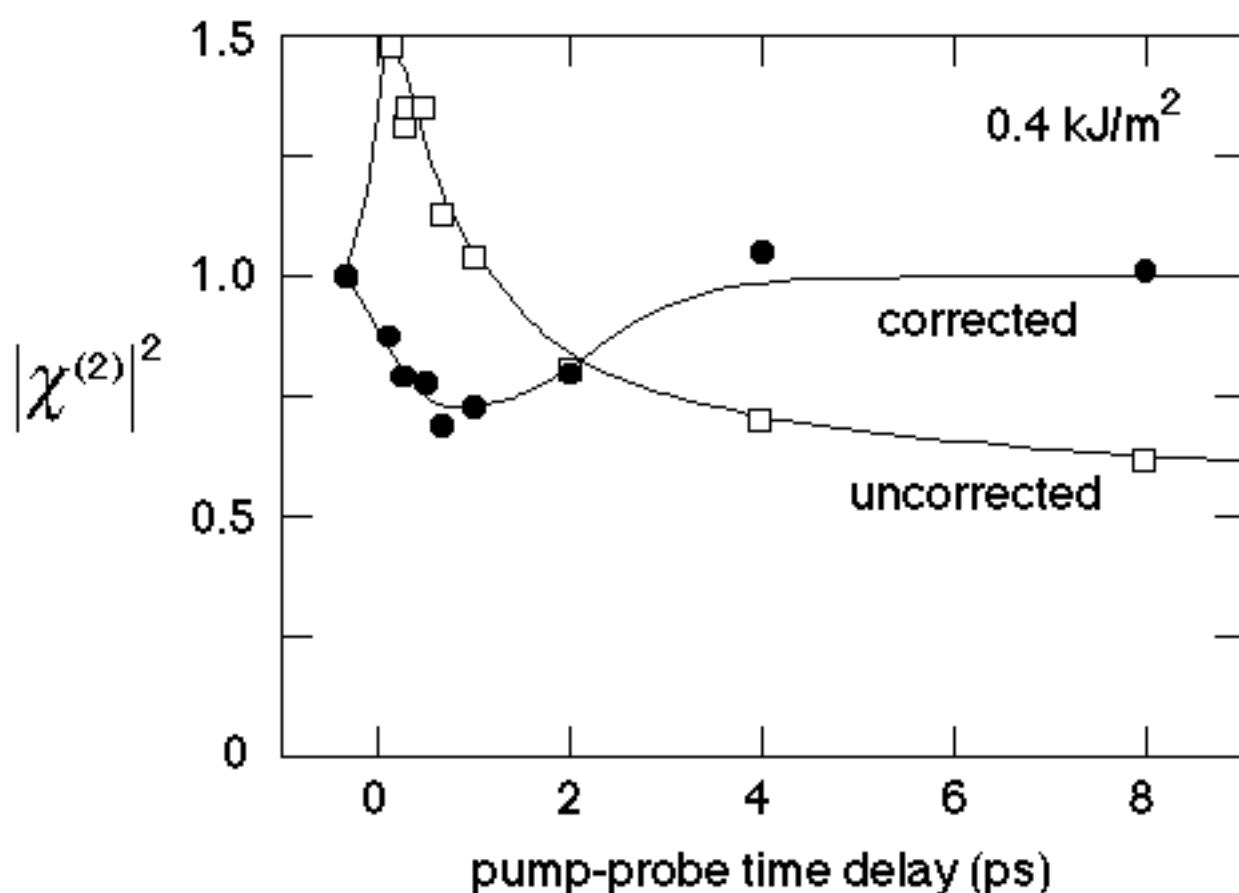
SECOND HARMONIC DATA



SECOND HARMONIC DATA



EFFECT OF CHANGES IN ϵ



FEMTOSECOND WORK

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

long-range order disappears even below threshold

structural change due to destabilization of covalent bonds

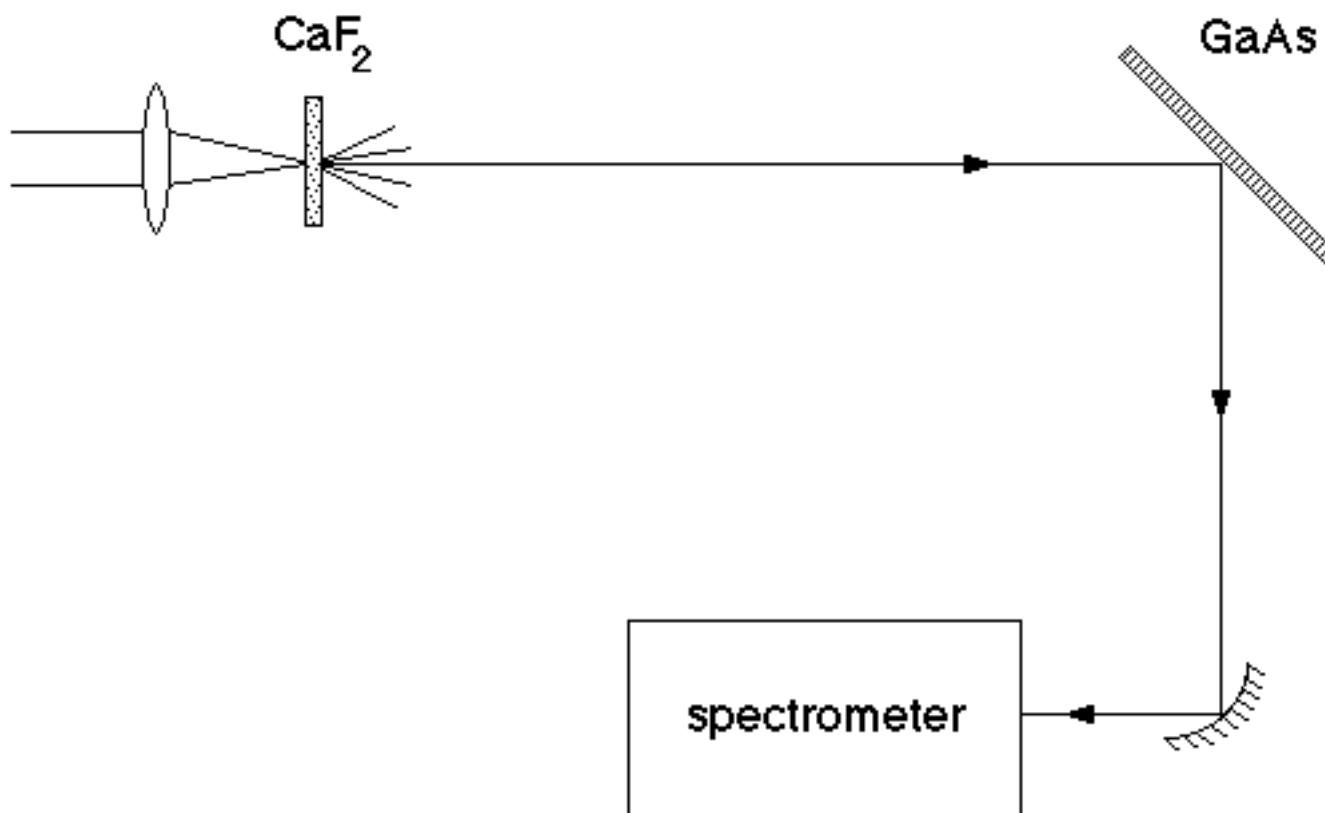


OUTLINE

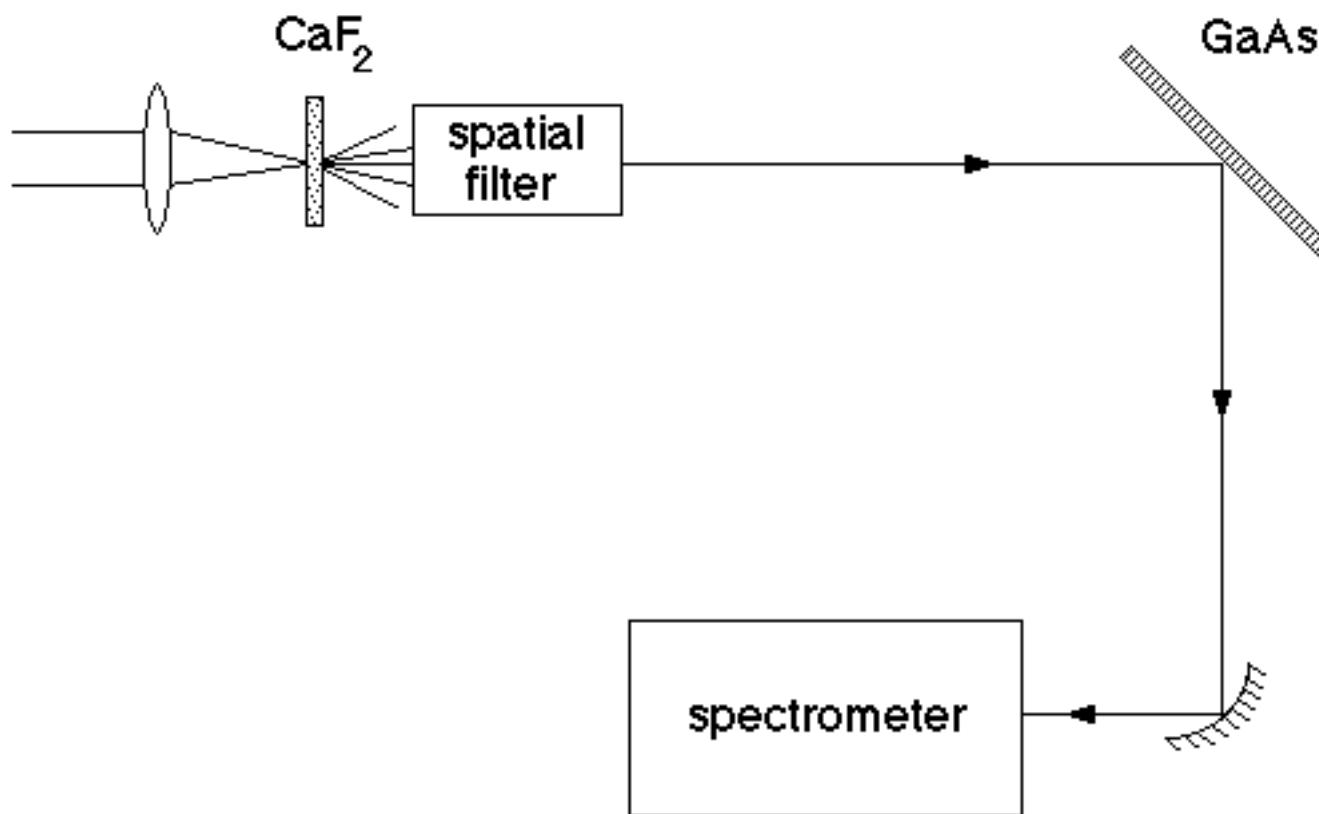
- ① Background
- ② Femtosecond work
- ③ Future



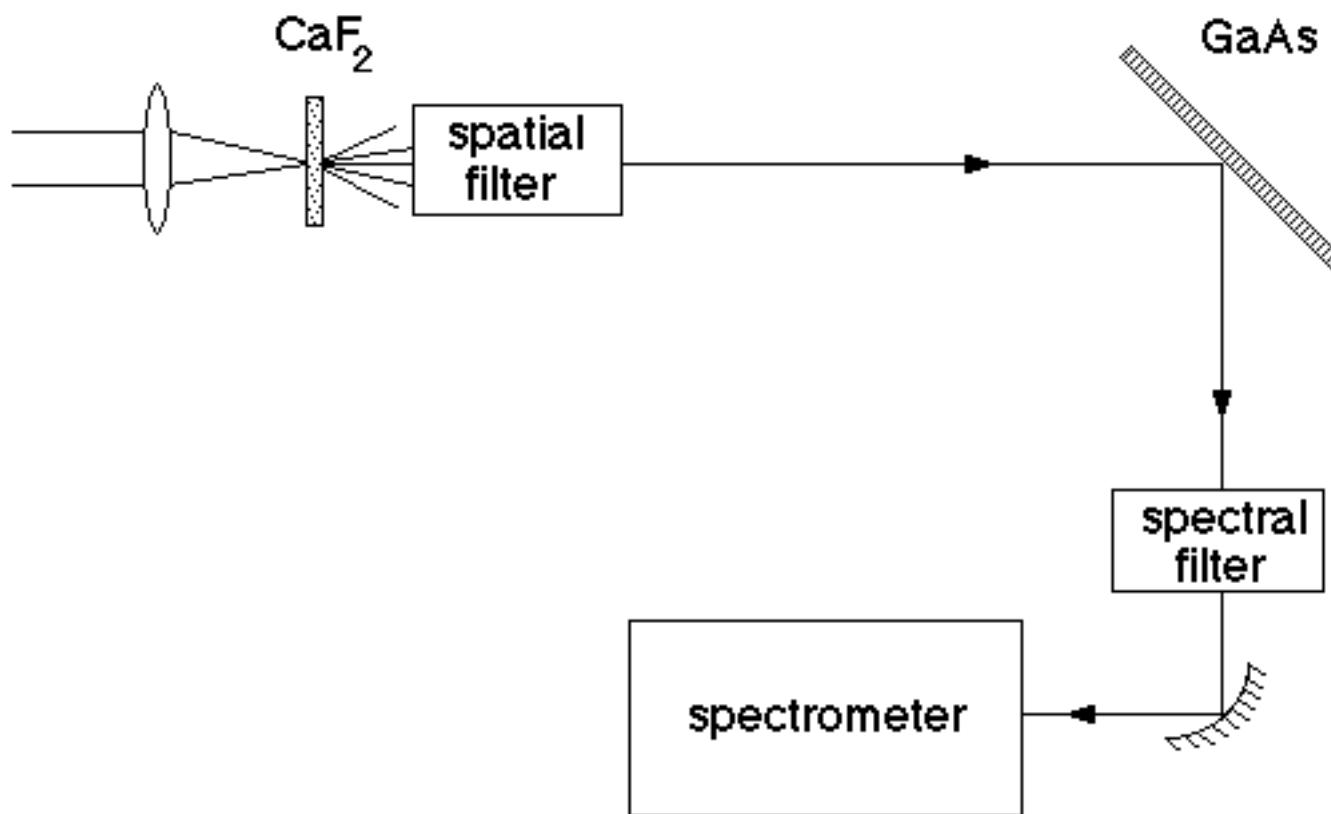
BROADBAND PROBE



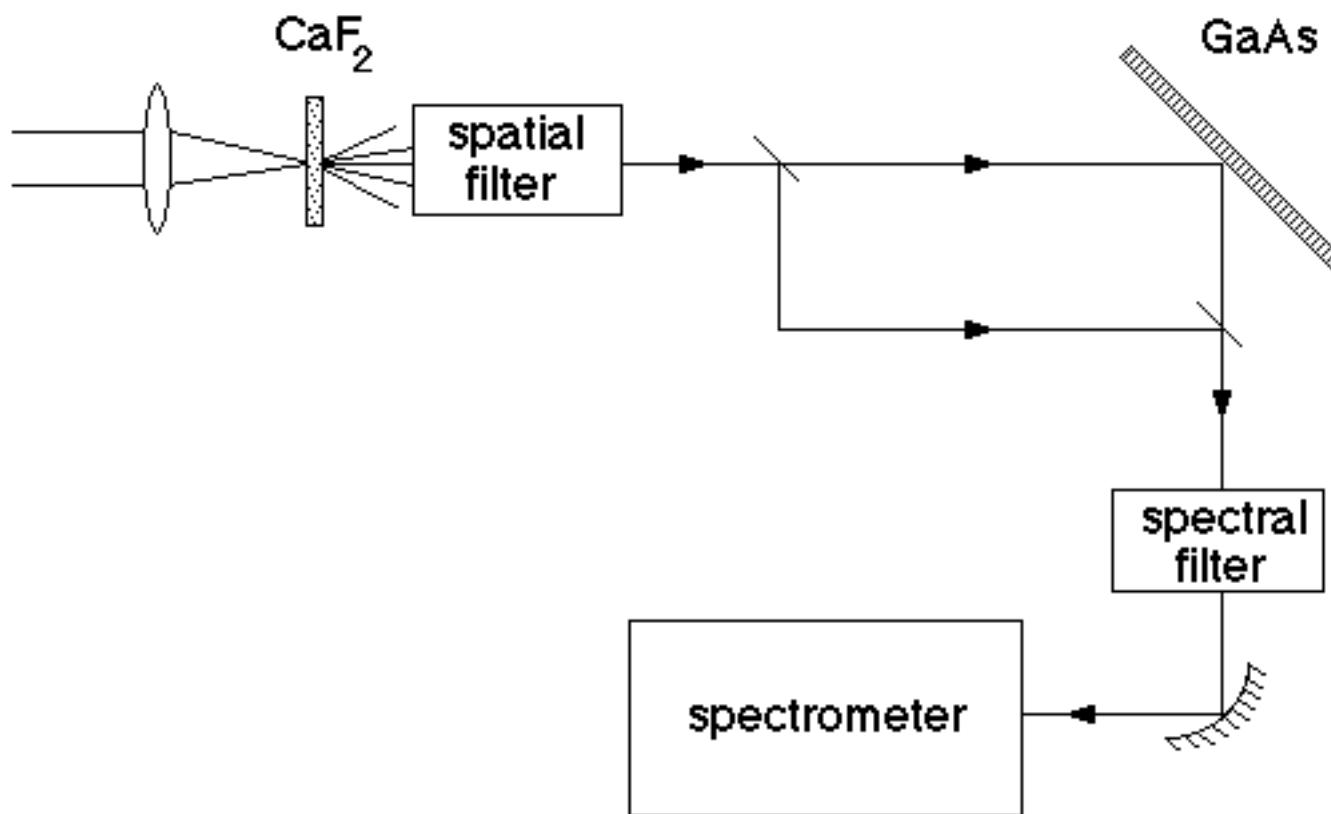
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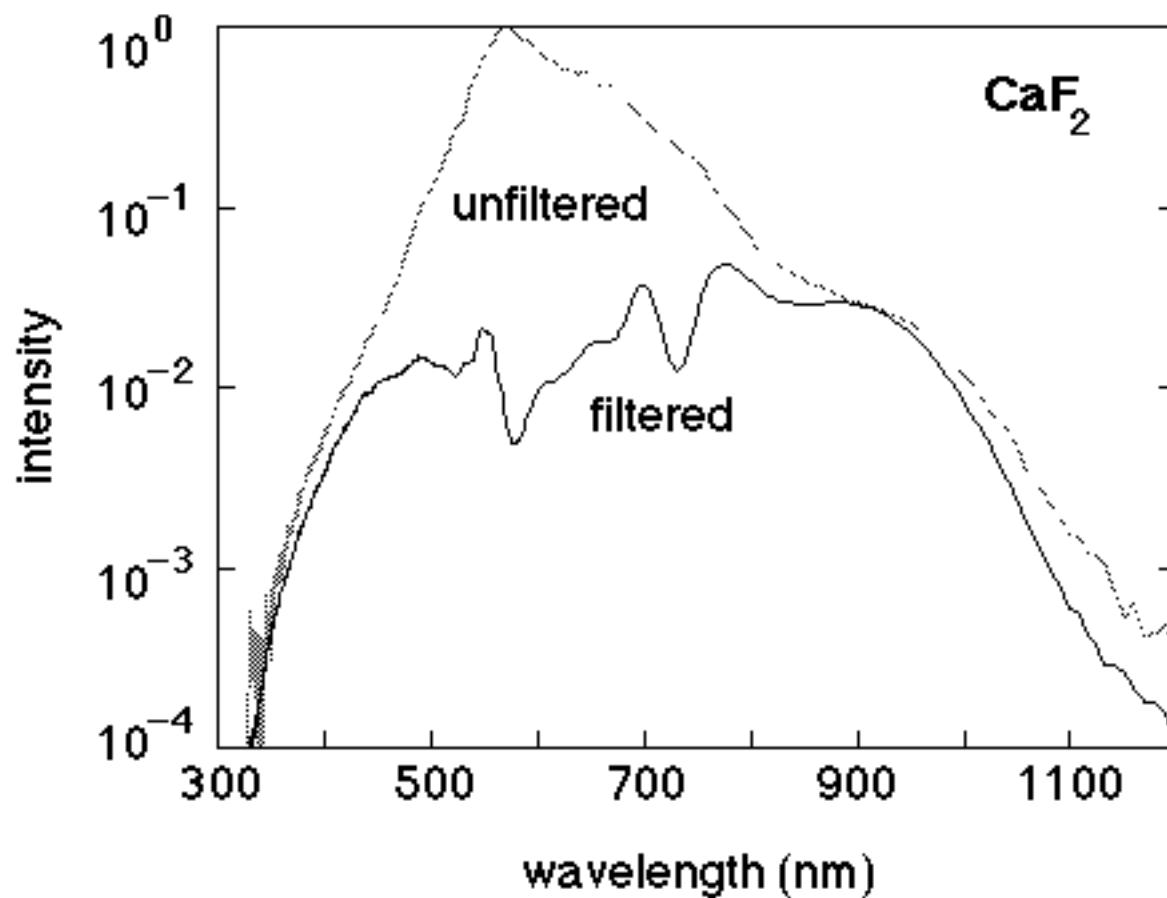
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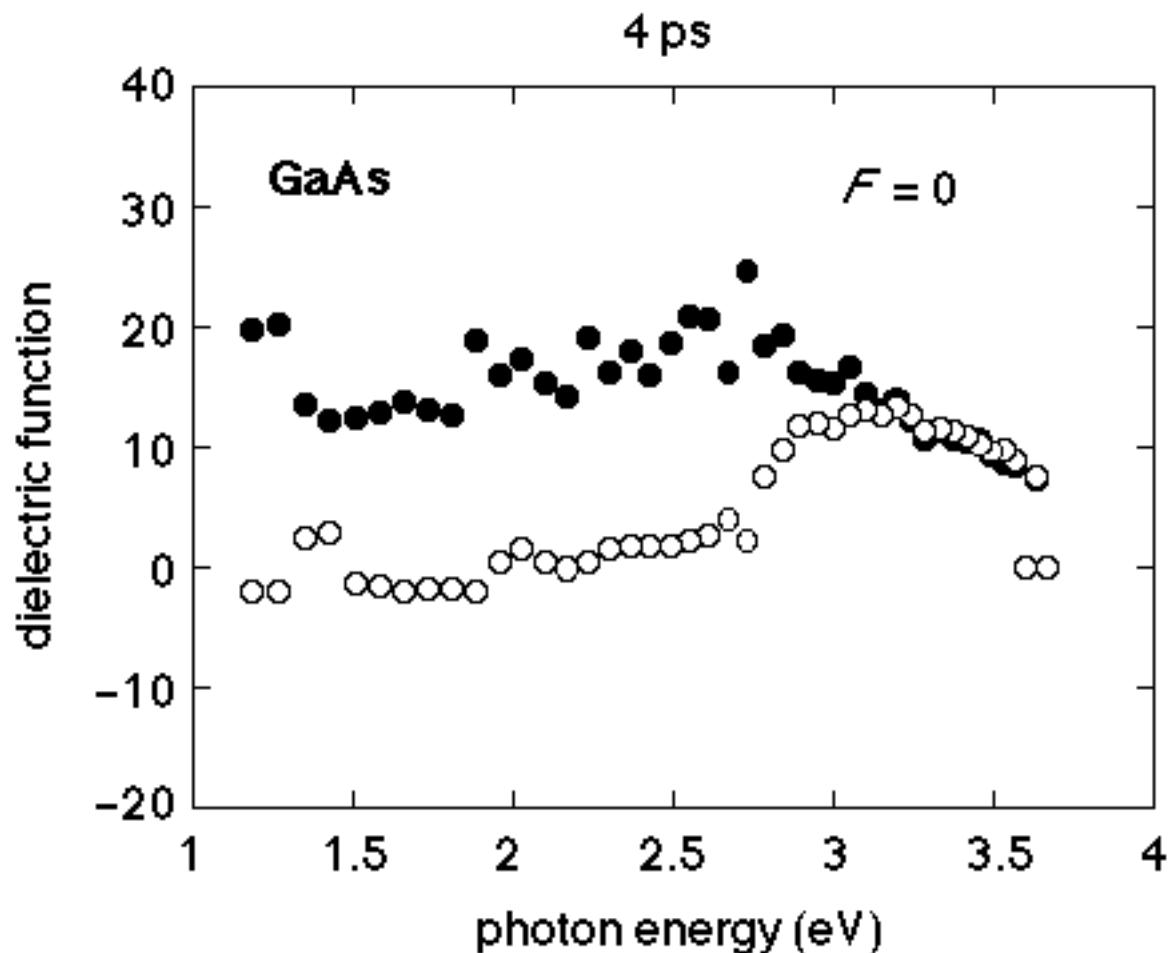
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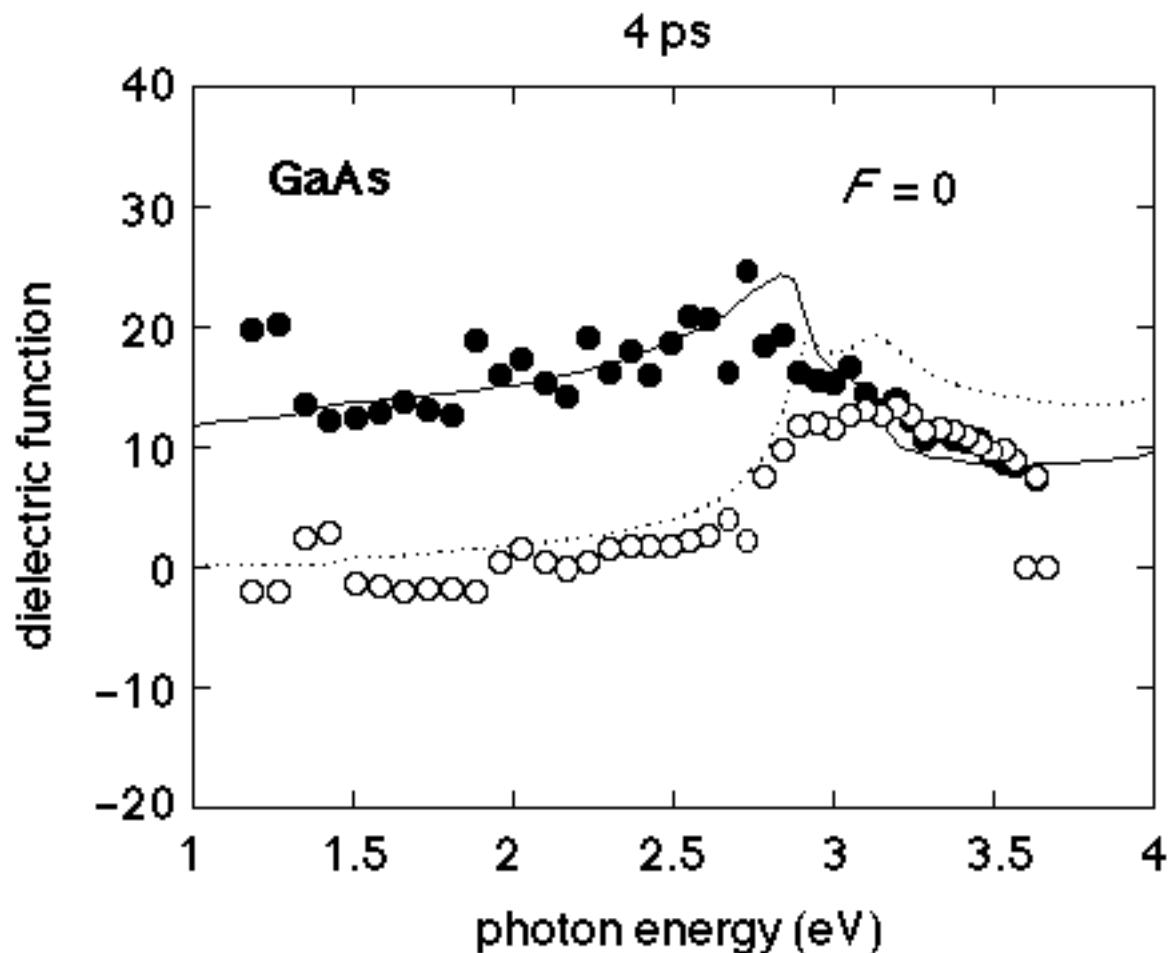
WHITE LIGHT SPECTRUM



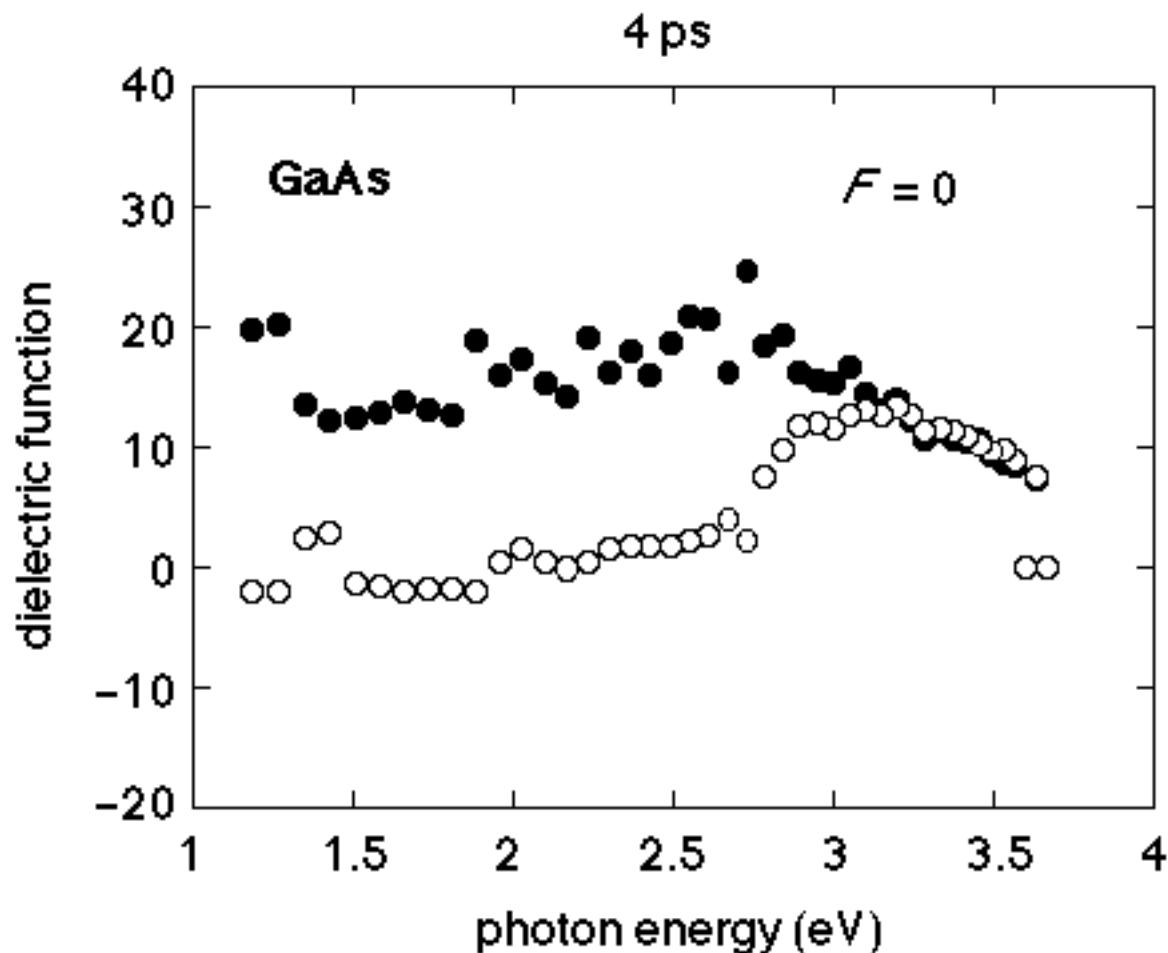
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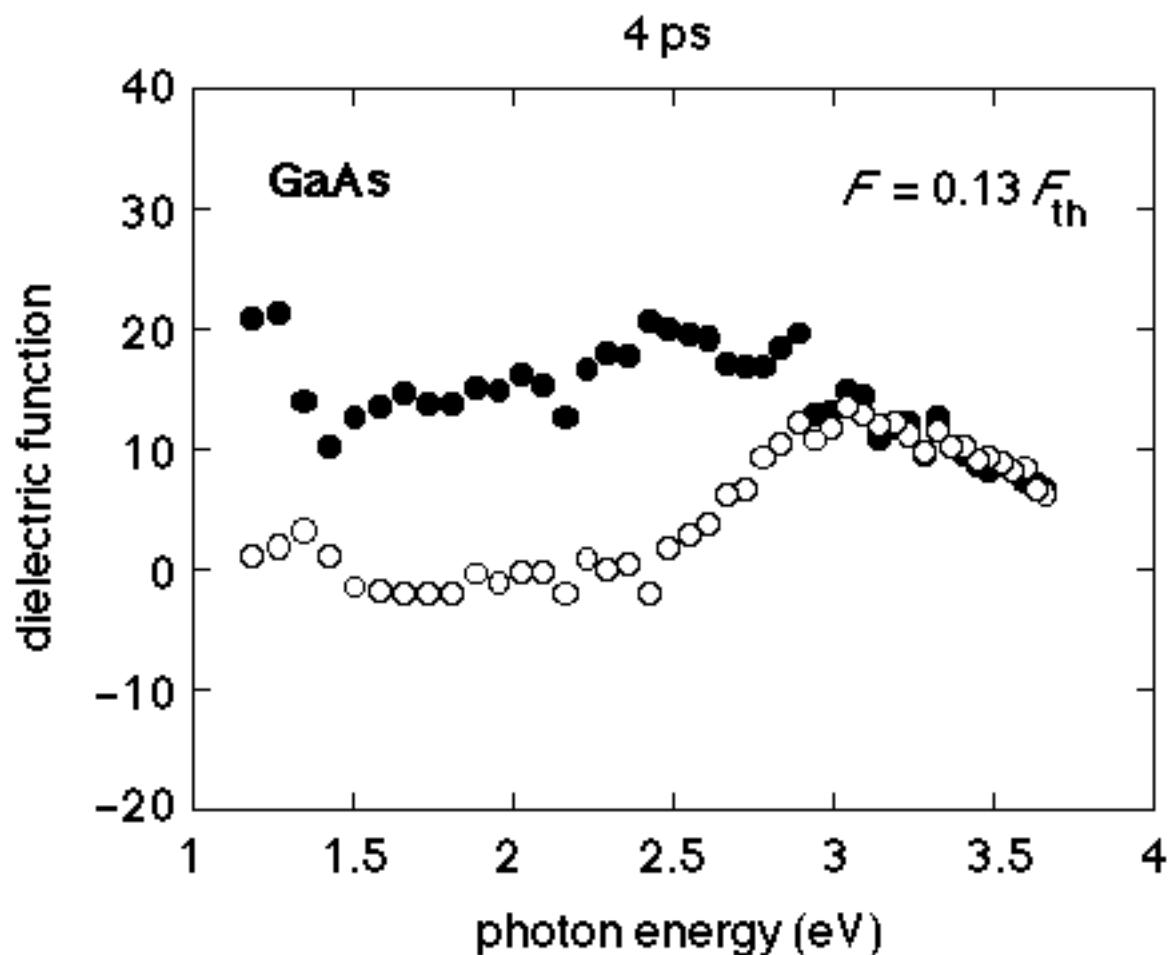
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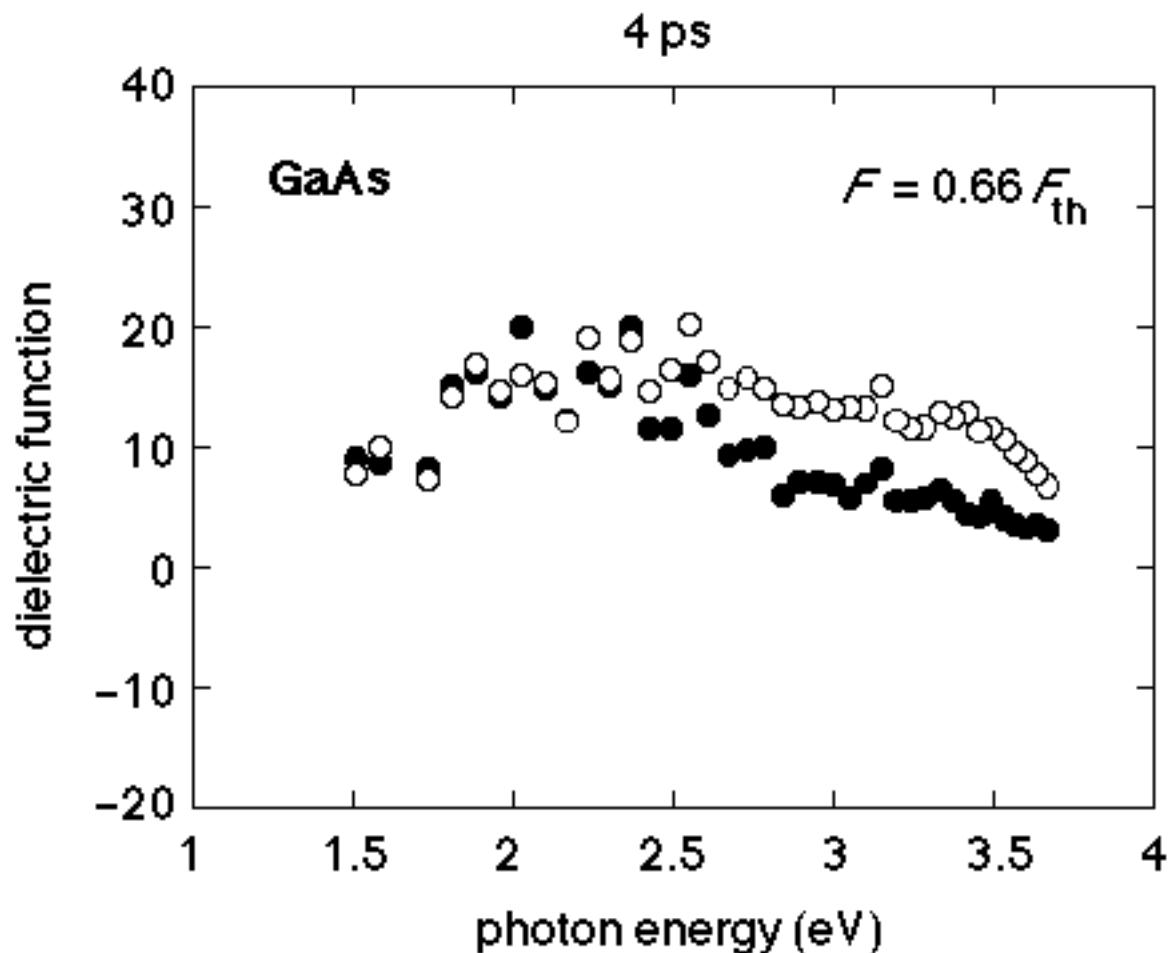
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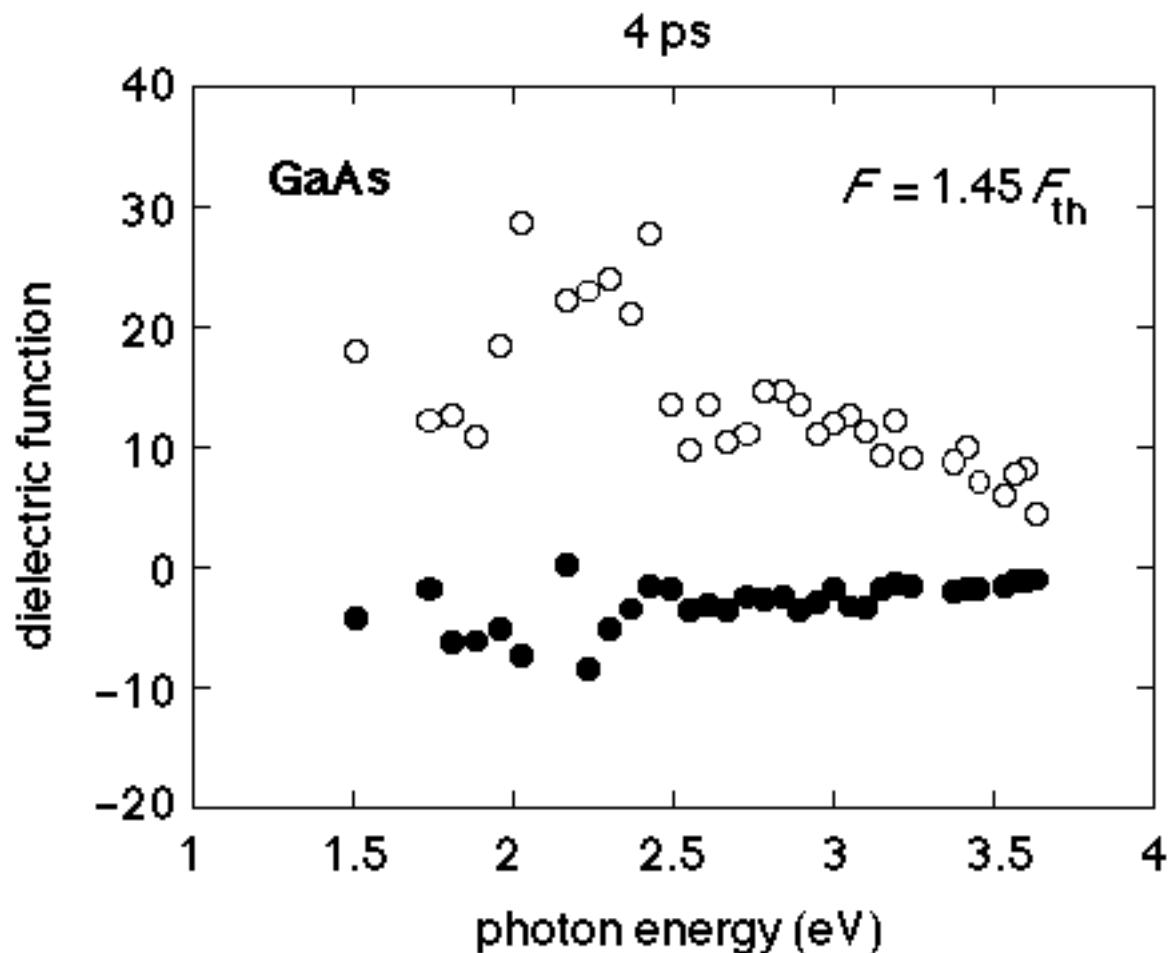
BROADBAND DATA



BROADBAND DATA



BROADBAND DATA



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

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