Ultrafast Phase Transition Dynamics in GeSb Films

Albert M.-T. Kim, J. Solis*, J.P. Callan, C.A.D. Roeser and E. Mazur

Harvard University, Division of Engineering & Applied Sciences
and Department of Physics

* Instituto de Optica, Madrid, Spain

Introduction

Motivation to study GeSb

Introduction

Motivation to study GeSb

Experimental Technique

Femtosecond time-resolved ellipsometry

OUTLINE

Introduction

Motivation to study GeSb

Experimental Technique

Femtosecond time-resolved ellipsometry

Results

Time resolved $\varepsilon(\omega)$ of GeSb films

Introduction

Motivation to study GeSb

Experimental Technique

Femtosecond time-resolved ellipsometry

Results

Time resolved $\varepsilon(\omega)$ of GeSb films

Analysis

Comparison to previous results

Introduction

Motivation to study GeSb

Experimental Technique

Femtosecond time-resolved ellipsometry

Results

Time resolved $\varepsilon(\omega)$ of GeSb films

Analysis

Comparison to previous results

Conclusions

Motivations to study phase transitions in GeSb films (GeSb)

Sb-rich films of Ge and Sb are interesting for optical data storage

- can optically induce transformation from crystalline to amorphous phase
- $\Delta R/R$ is about 18%

Motivations to study phase transitions in GeSb films (GeSb)

Sb-rich films of Ge and Sb are interesting for optical data storage

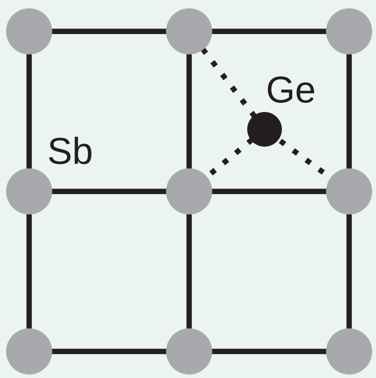
— can optically induce transformation from crystalline to amorphous phase

— $\Delta R/R$ is about 18%

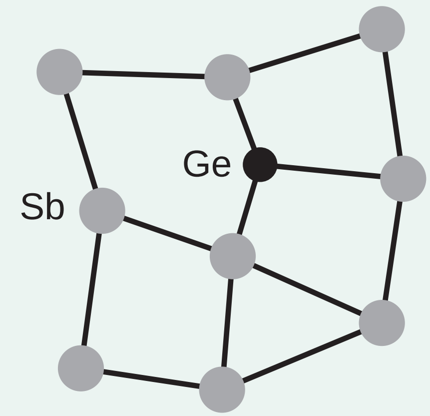
Recently observed ultrafast disorder to order phase transition

— Sokolowski-Tinten et al. reported on crystallization within 200fs

Crystalline vs amorphous phase of GeSb



$Ge_{0.06}Sb_{0.94}$



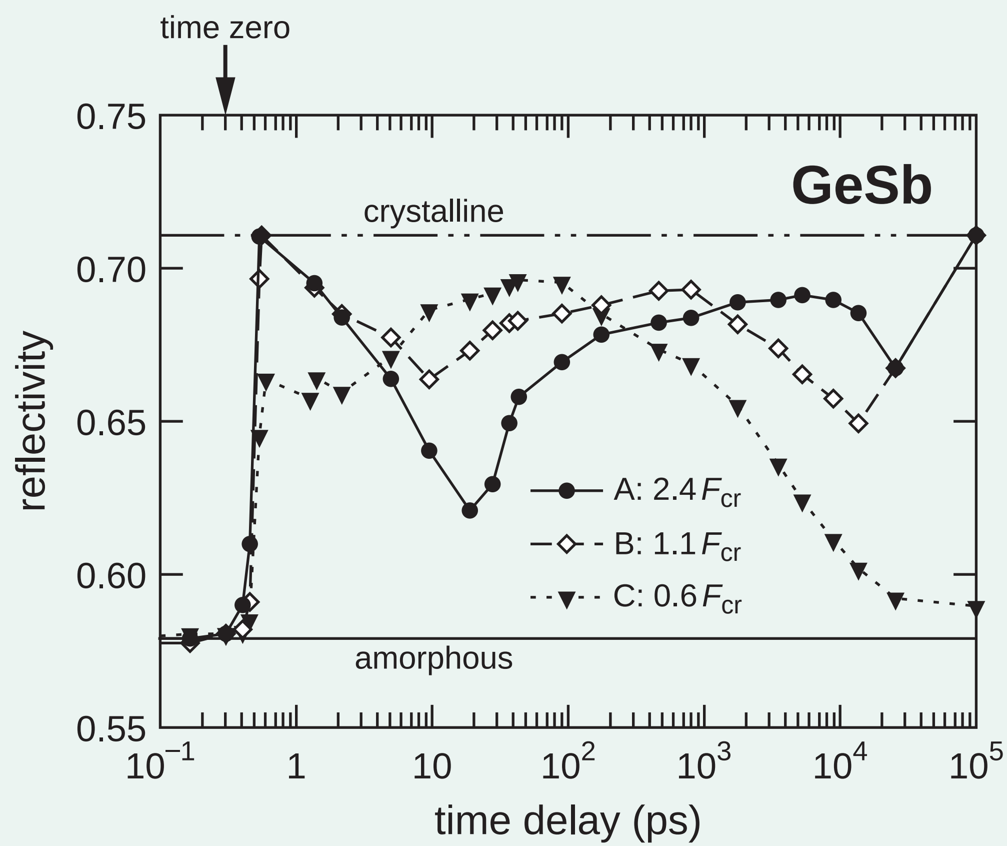
crystalline structure identical to pure Sb — solid solution of Ge in Sb

R ≅ 67%

amorphous phase is stabilized by Ge atoms

R ≅ 55%

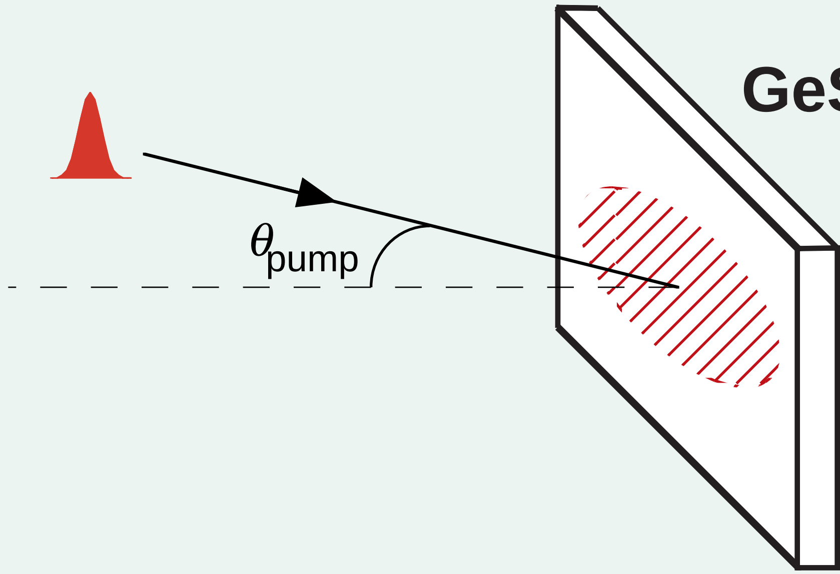
Previous work hints at ultrafast crystallization



Transient reflectivities at 2.01 eV and 0^o angle of incidence

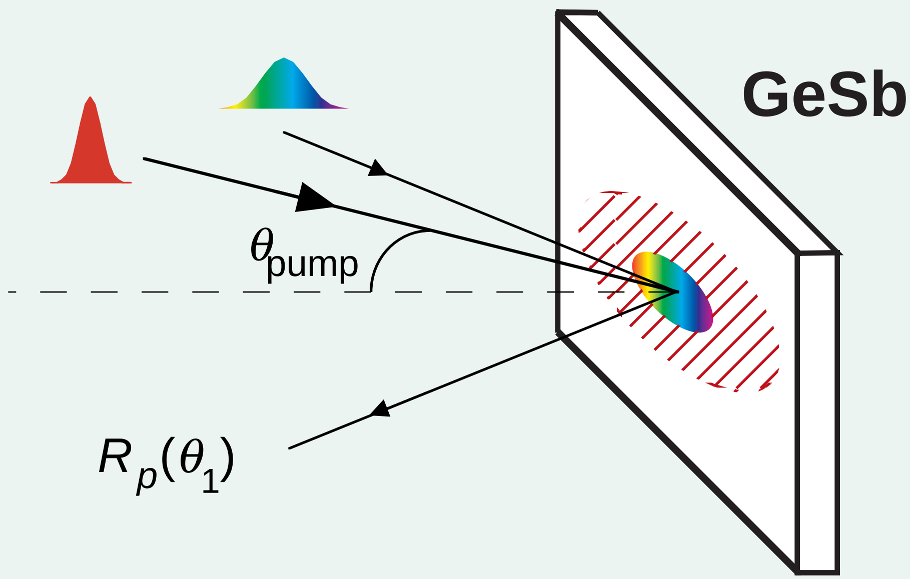
Sokolowski-Tinten et al. PRL, **81**, 3670 (1998)

FS time resolved ellipsometry — FTRE



pump pulse:
- 800 nm
- 35 fs
- $\theta_{pump} = 50^o$

FTRE — first angle reflectivity spectrum



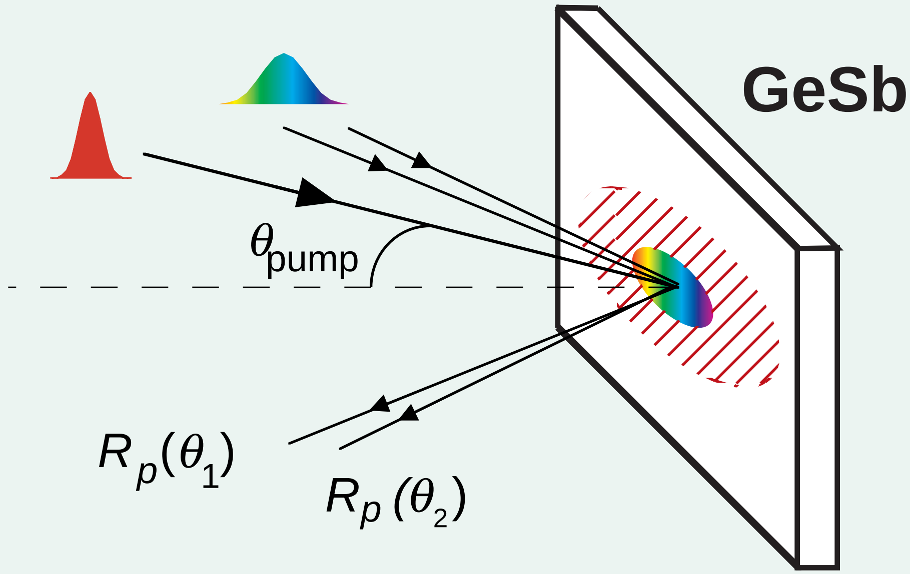
time delayed probe pulse:

— 1.7 — 3.5 eV (350nm — 750nm)

— < 0.1 μJ

— θ_1=53^o

FTRE — second angle reflectivity spectrum



time delayed probe pulse:

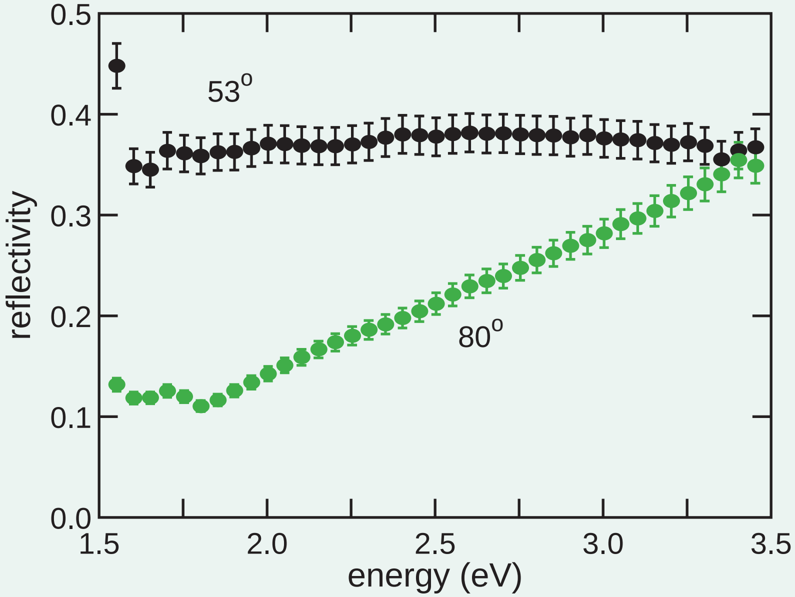
— 1.7 — 3.5 eV (350nm — 750nm)

— < 0.1 µJ

— θ_1=53^o, θ_2=80^o

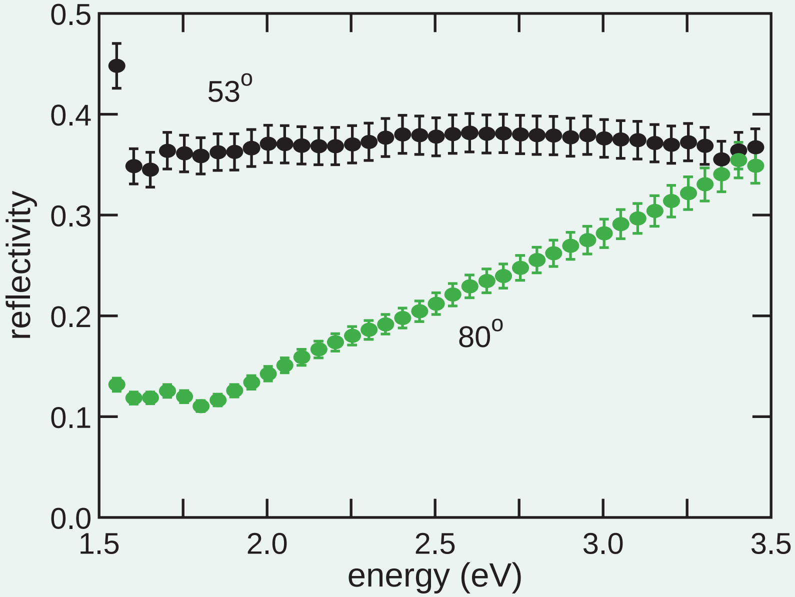
FTRE — extracting the dielectric function

Reflectivity spectra



FTRE — extracting the dielectric function

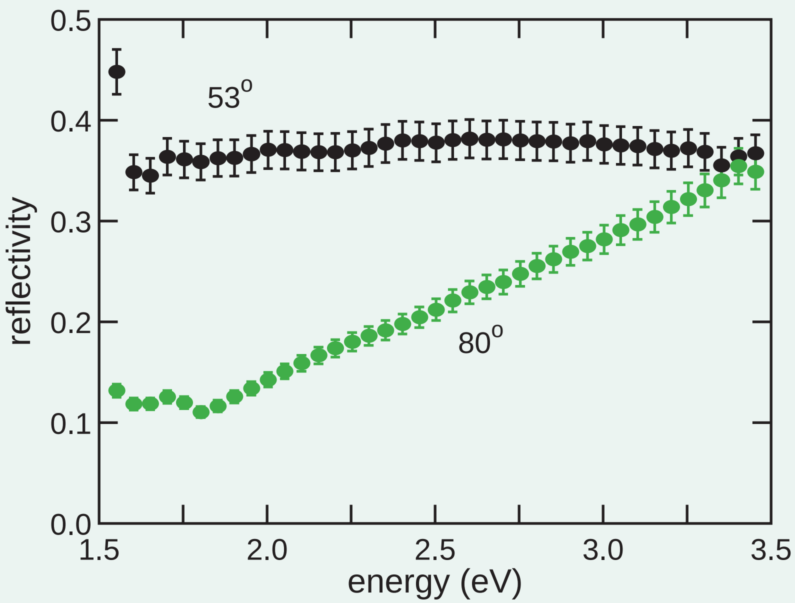
Reflectivity spectra



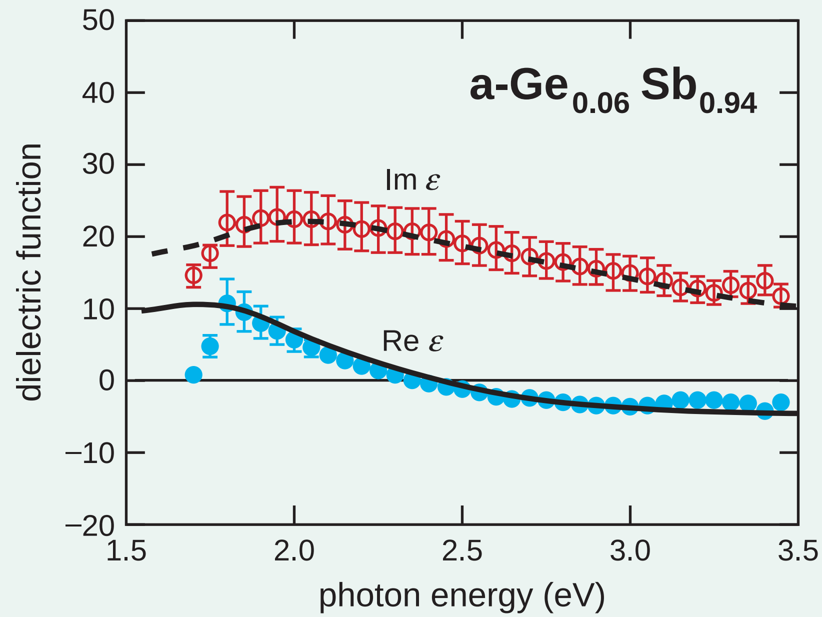
Numerically invert Fresnel formula

FTRE — extracting the dielectric function

Reflectivity spectra

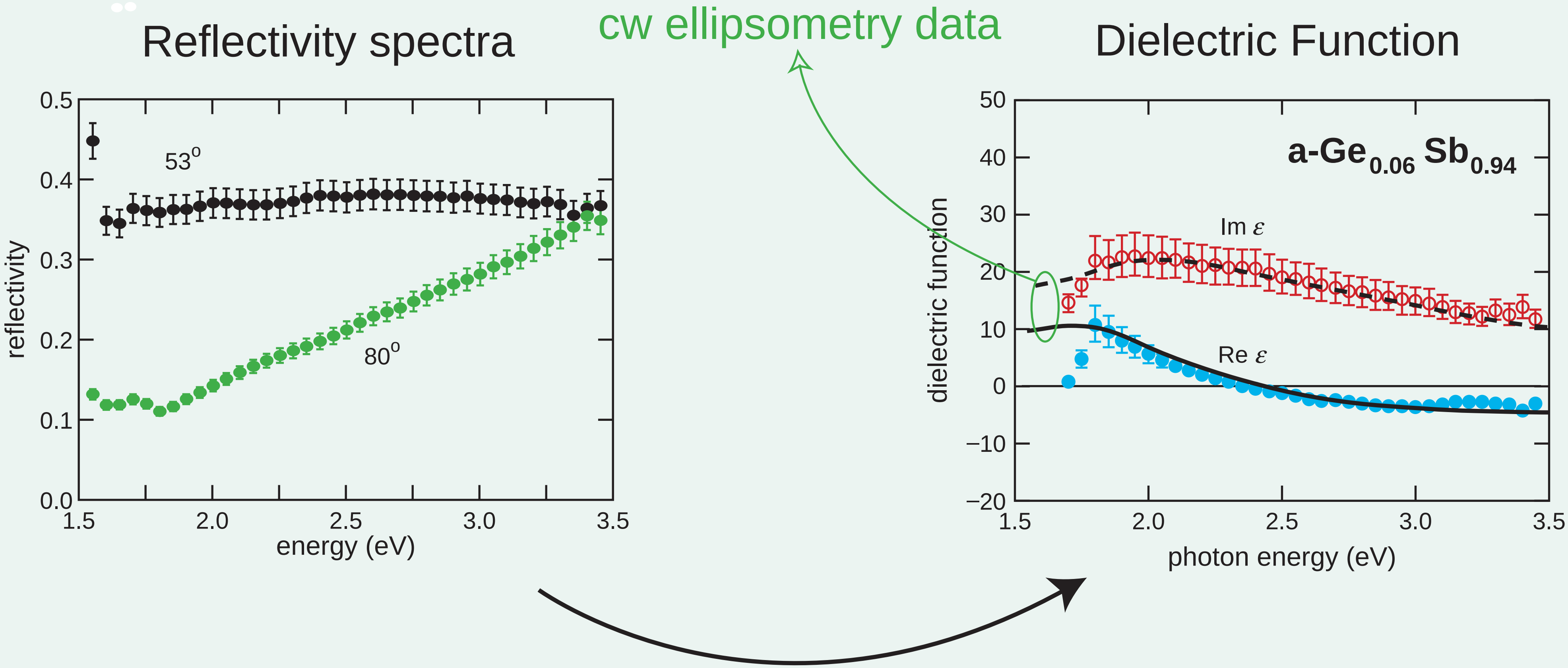


Dielectric Function



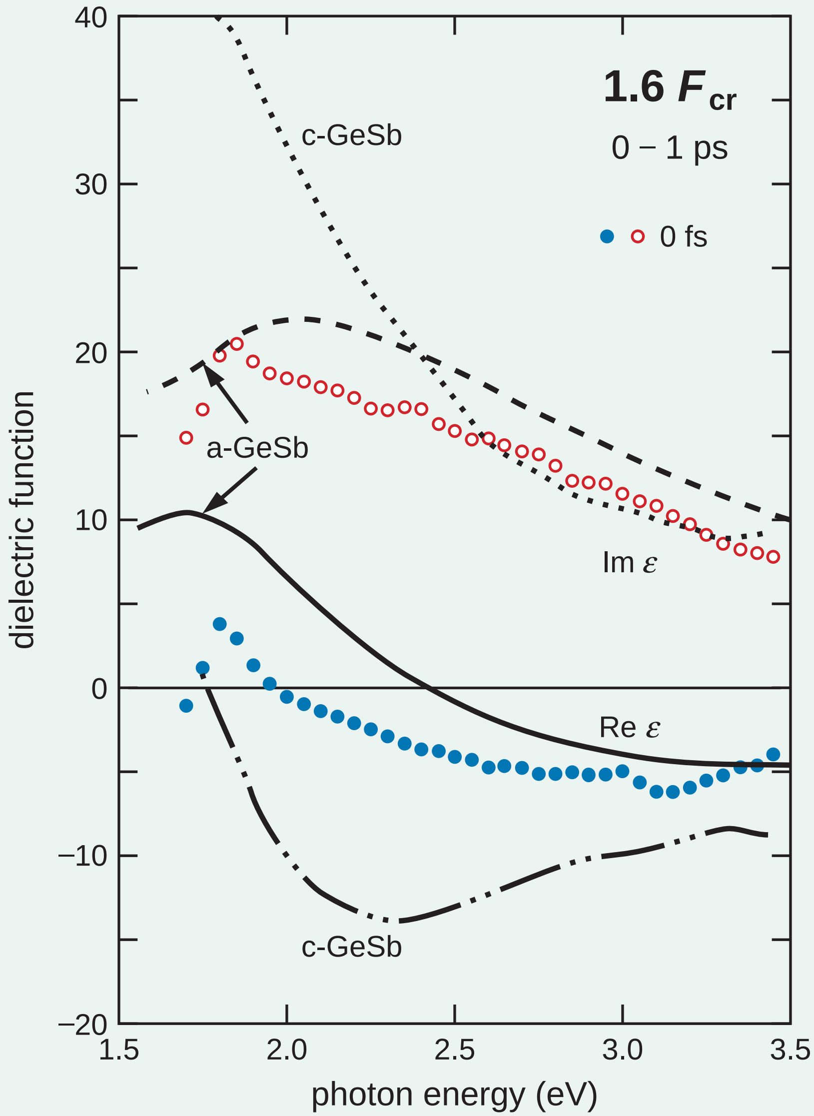
Numerically invert Fresnel formula

FTRE — extracting the dielectric function



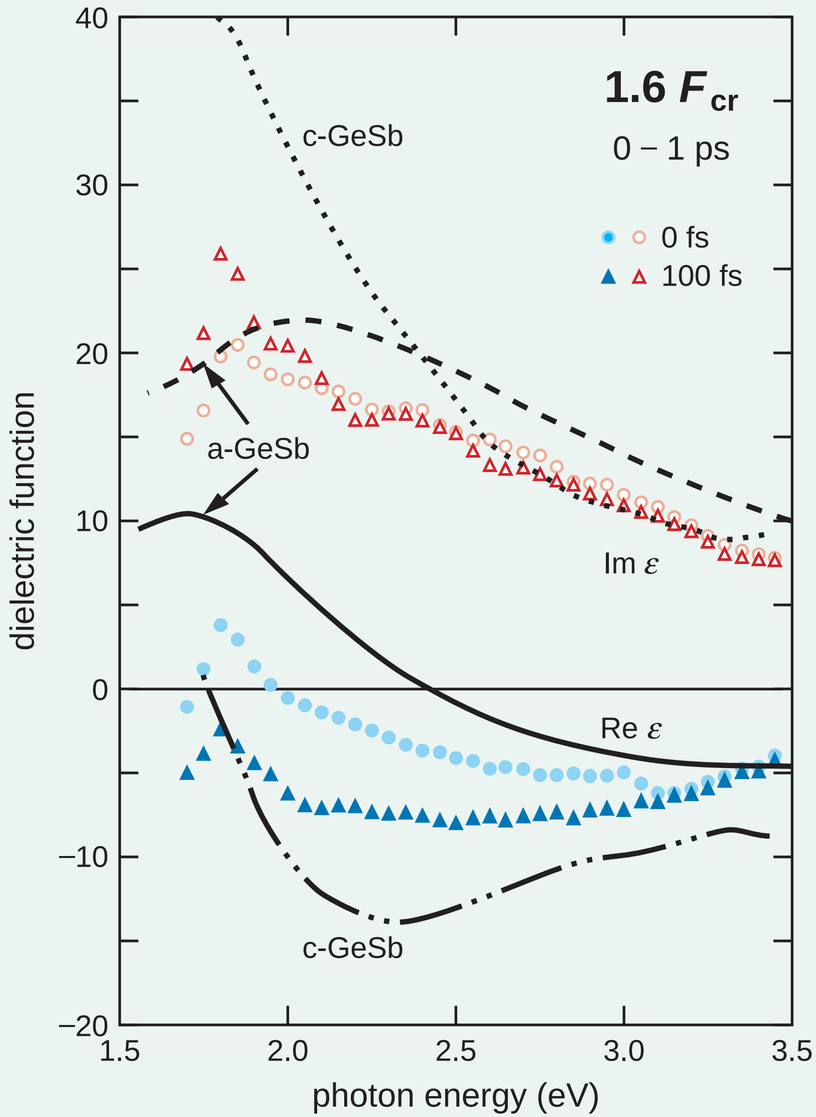
Numerically invert Fresnel formula

Evolution of ε(ω) after excitation at 1.6 F_{cr} = 0.22 kJ/m^2



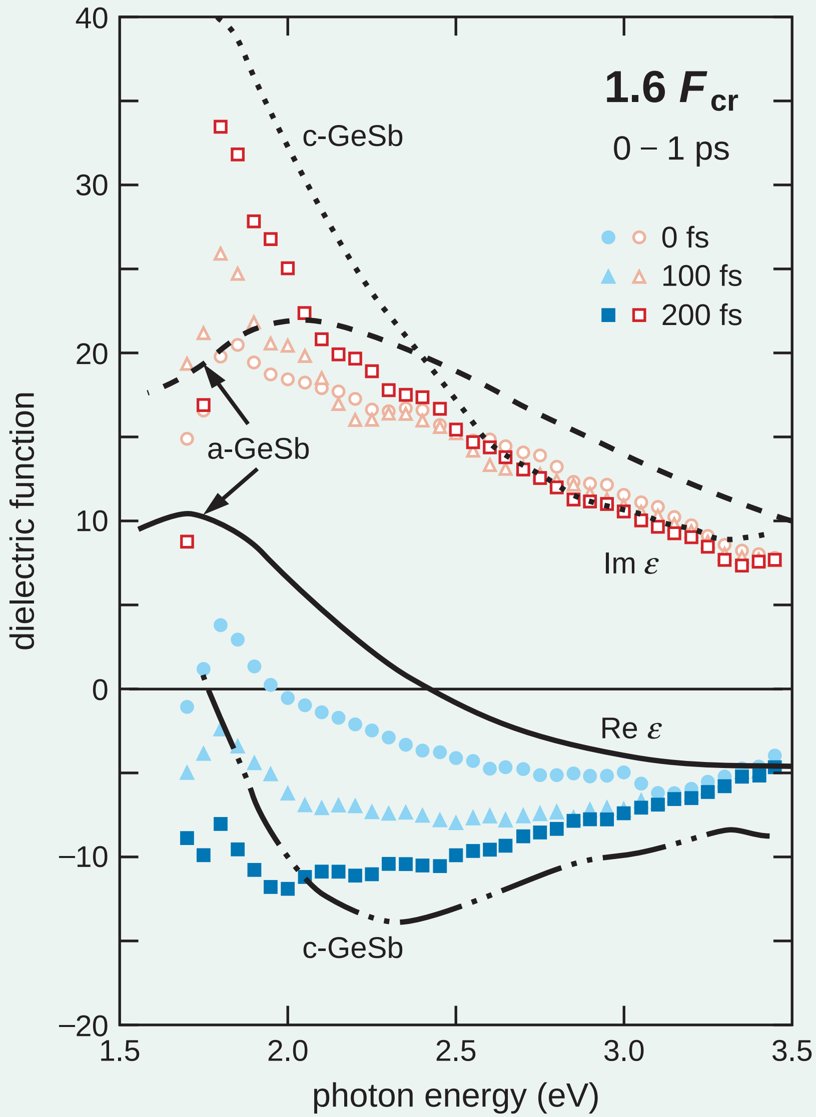
ε(ω) gradually shifts downwards

Evolution of ε(ω) after excitation at 1.6 F_{cr}



ε(ω) gradually shifts downwards

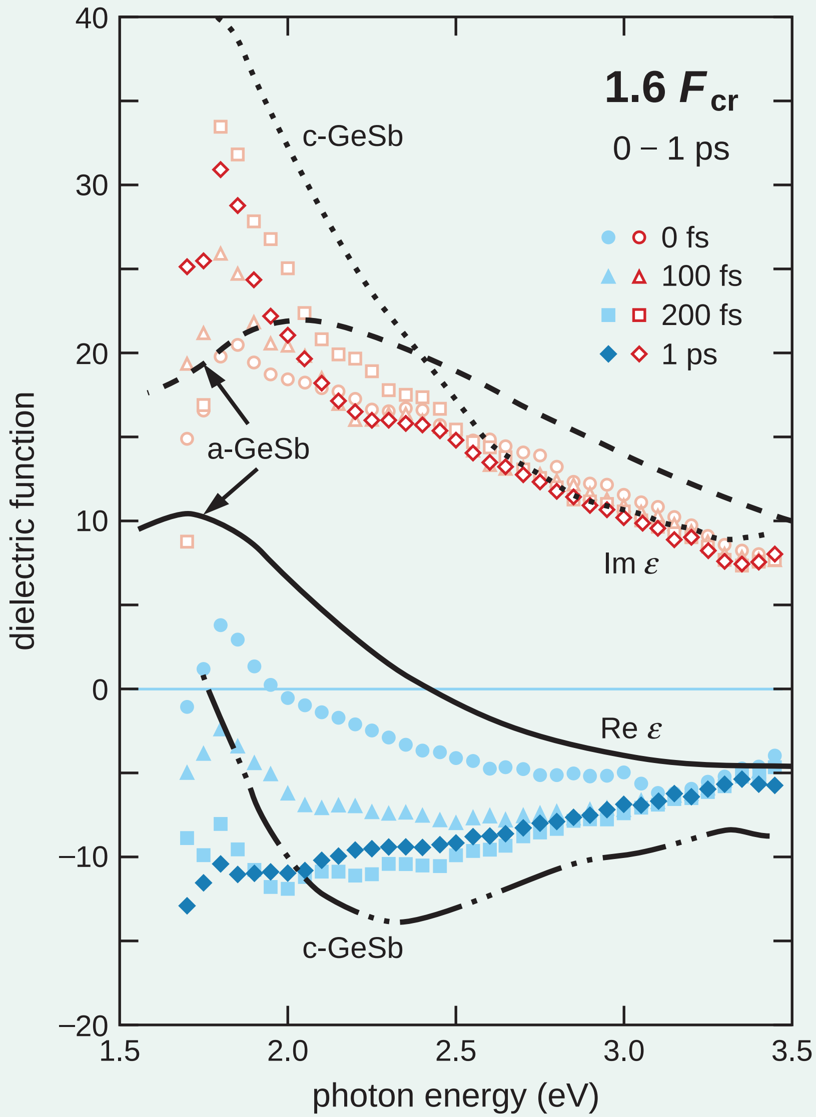
Evolution of ε(ω) after excitation at 1.6 F_{cr}



ε(ω) gradually shifts downwards

ε(ω) approaches c-GeSb

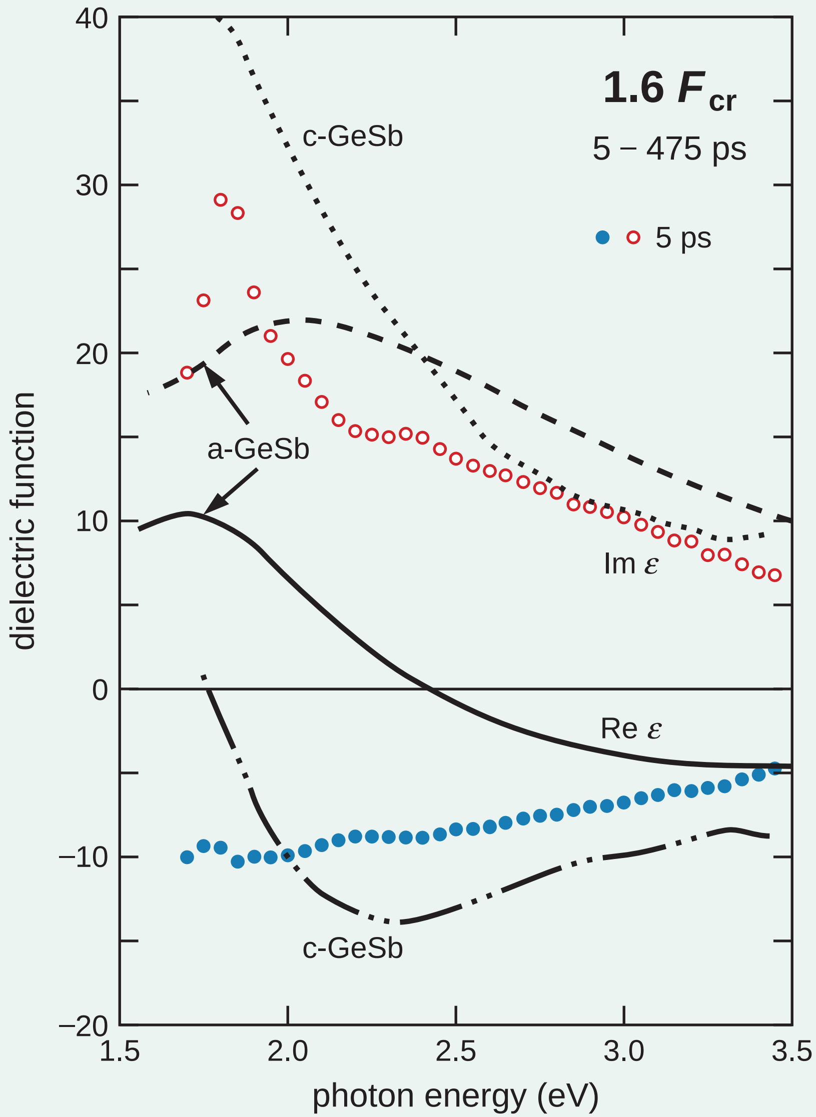
Evolution of ε(ω) after excitation at 1.6 F_{cr}



ε(ω) gradually shifts downwards

ε(ω) approaches c-GeSb

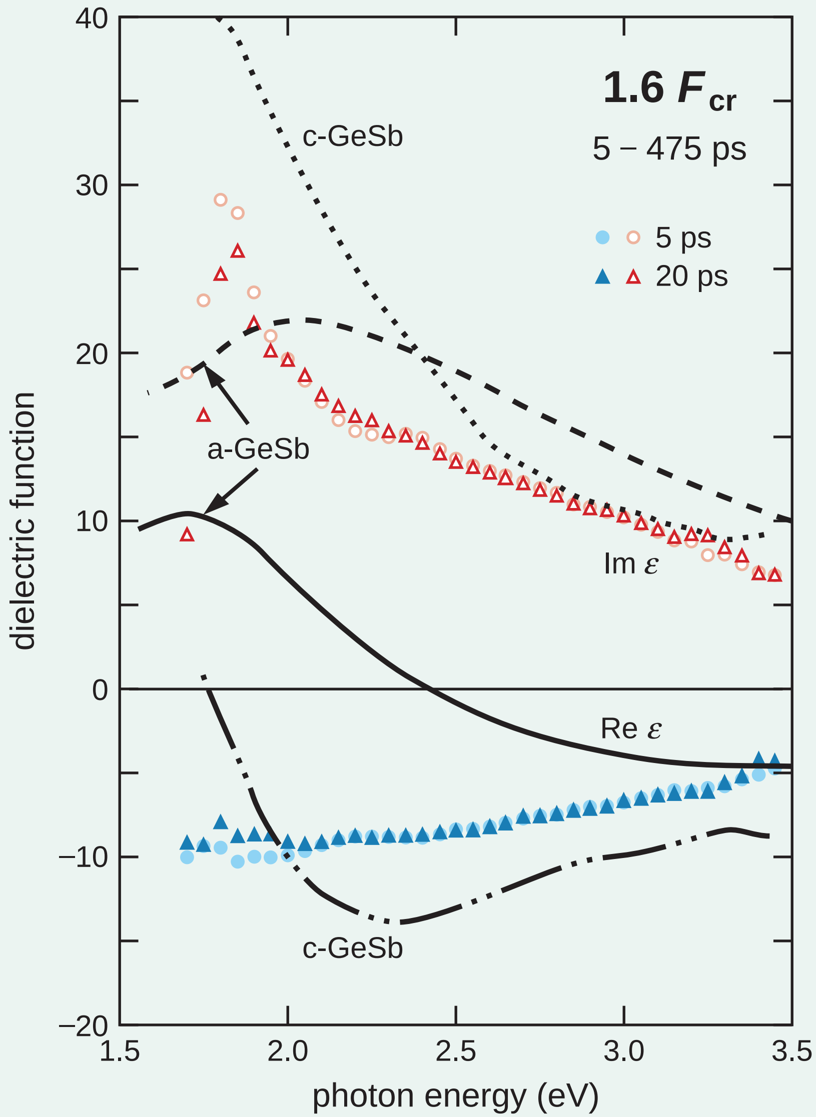
Evolution of ε(ω) after excitation at 4.0 F_{cr}



ε(ω) gradually shifts downwards

ε(ω) approaches c-GeSb

Evolution of ε(ω) after excitation at 1.6 F_{cr}

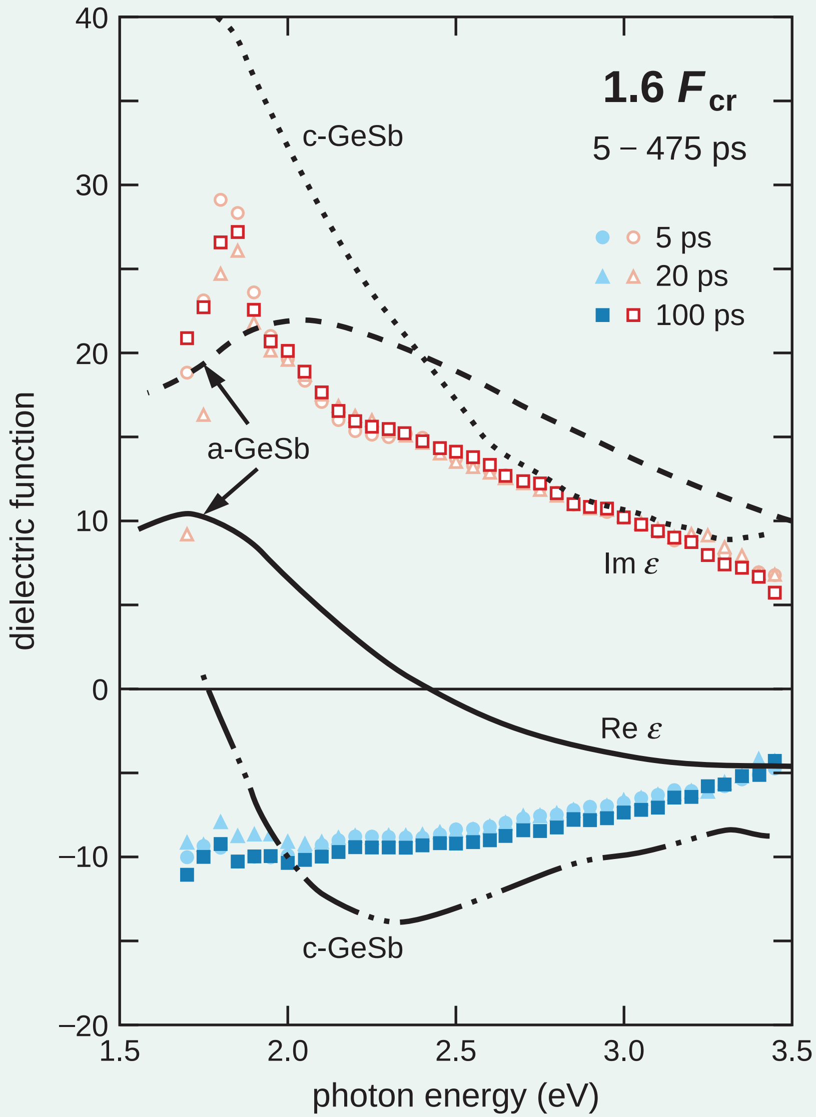


ε(ω) gradually shifts downwards

ε(ω) approaches c-GeSb

ε(ω) reaches intermediate state

Evolution of ε(ω) after excitation at 1.6 F_{cr}

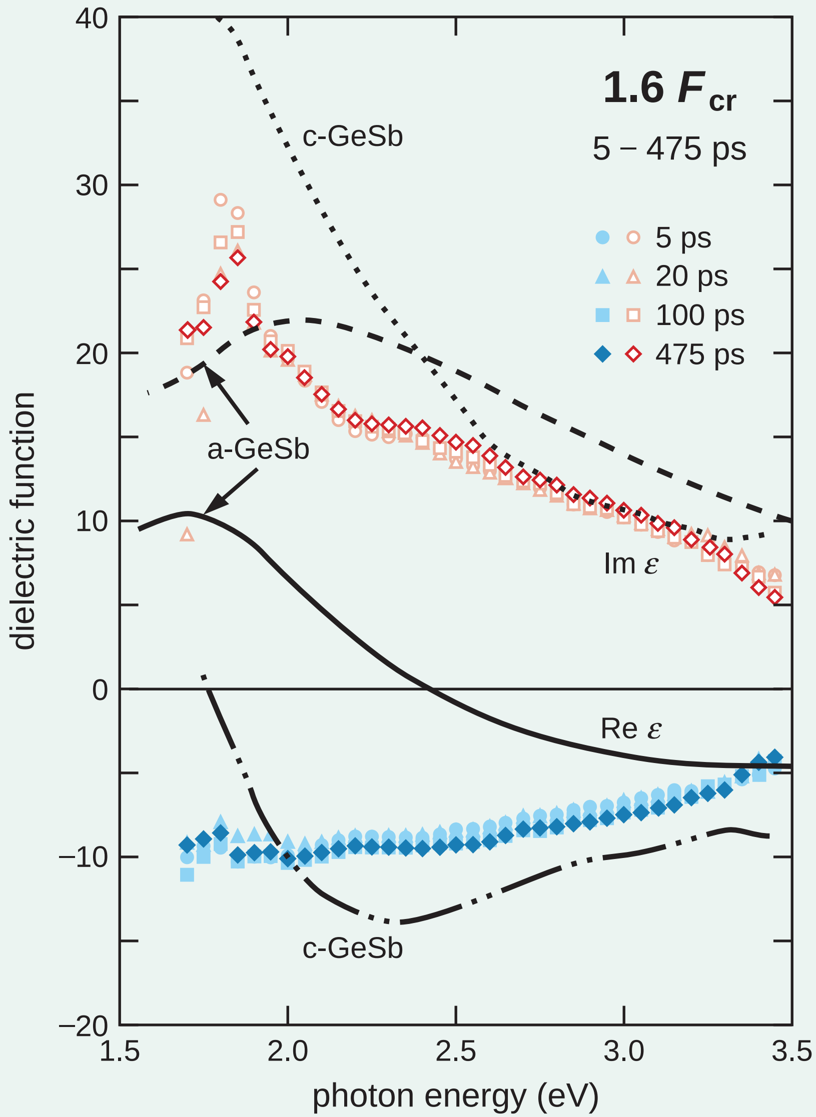


ε(ω) gradually shifts downwards

ε(ω) approaches c-GeSb

ε(ω) reaches intermediate state

Evolution of ε(ω) after excitation at 1.6 F_{cr}



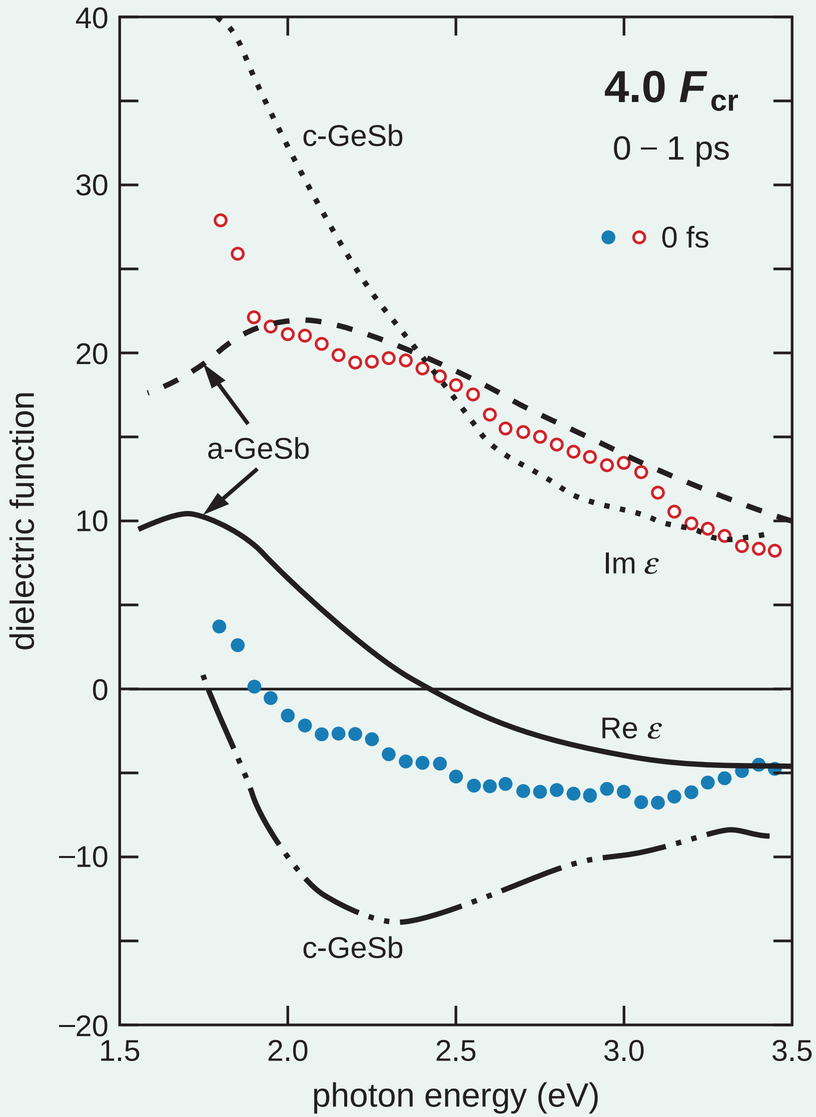
ε(ω) gradually shifts downwards

ε(ω) approaches c-GeSb

ε(ω) reaches intermediate state

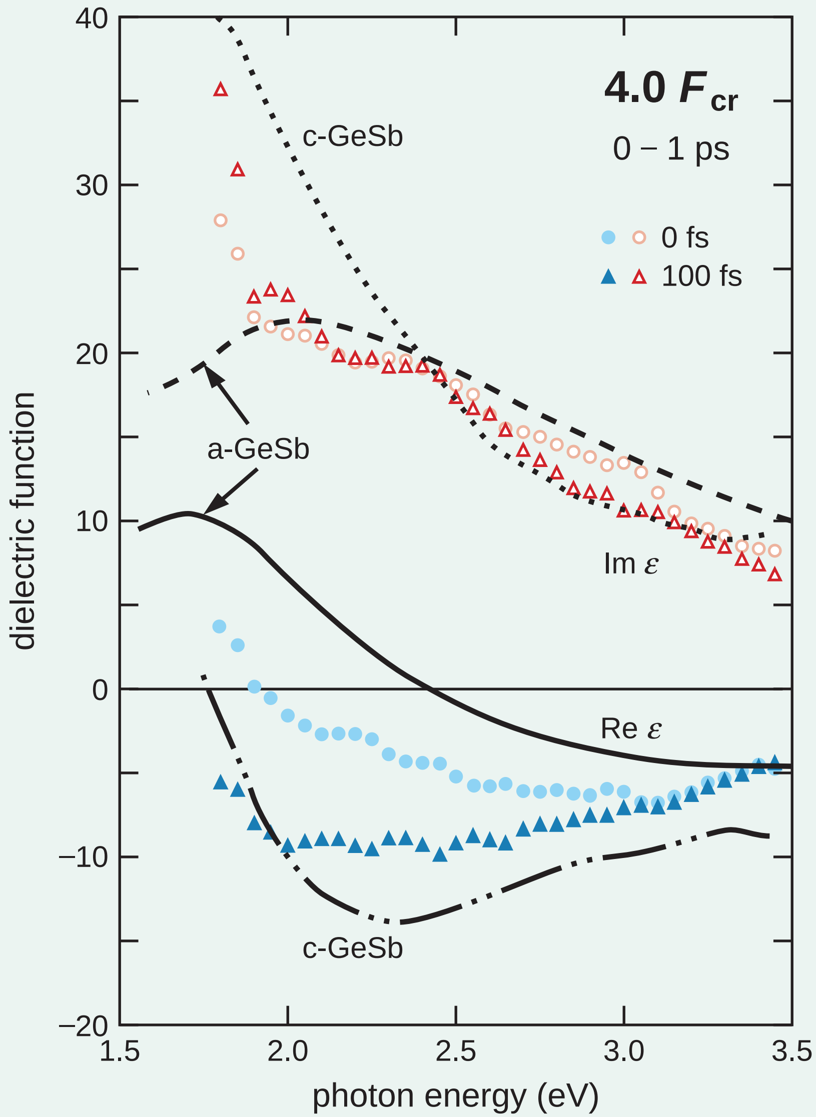
state is NOT c-GeSb

Evolution of ε(ω) after excitation at 4.0 F_{cr}



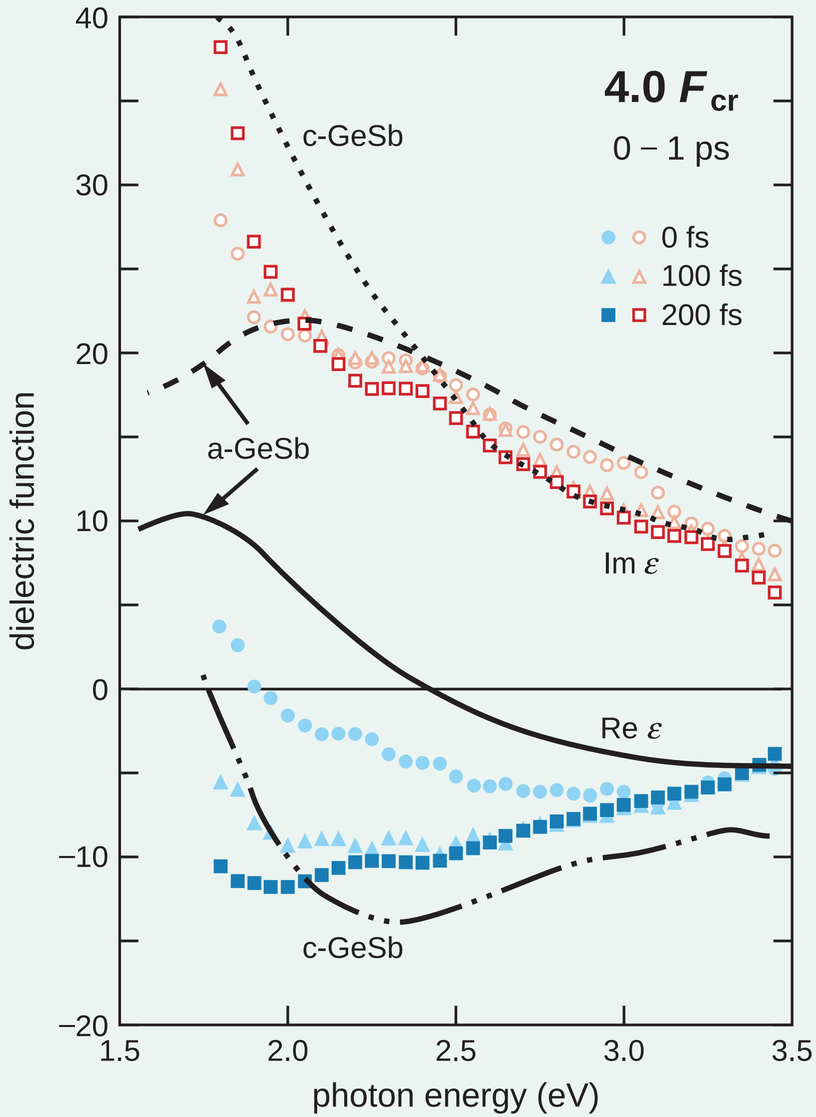
ε(ω) gradually shifts downwards

Evolution of ε(ω) after excitation at 4.0 F_{cr}



ε(ω) gradually shifts downwards

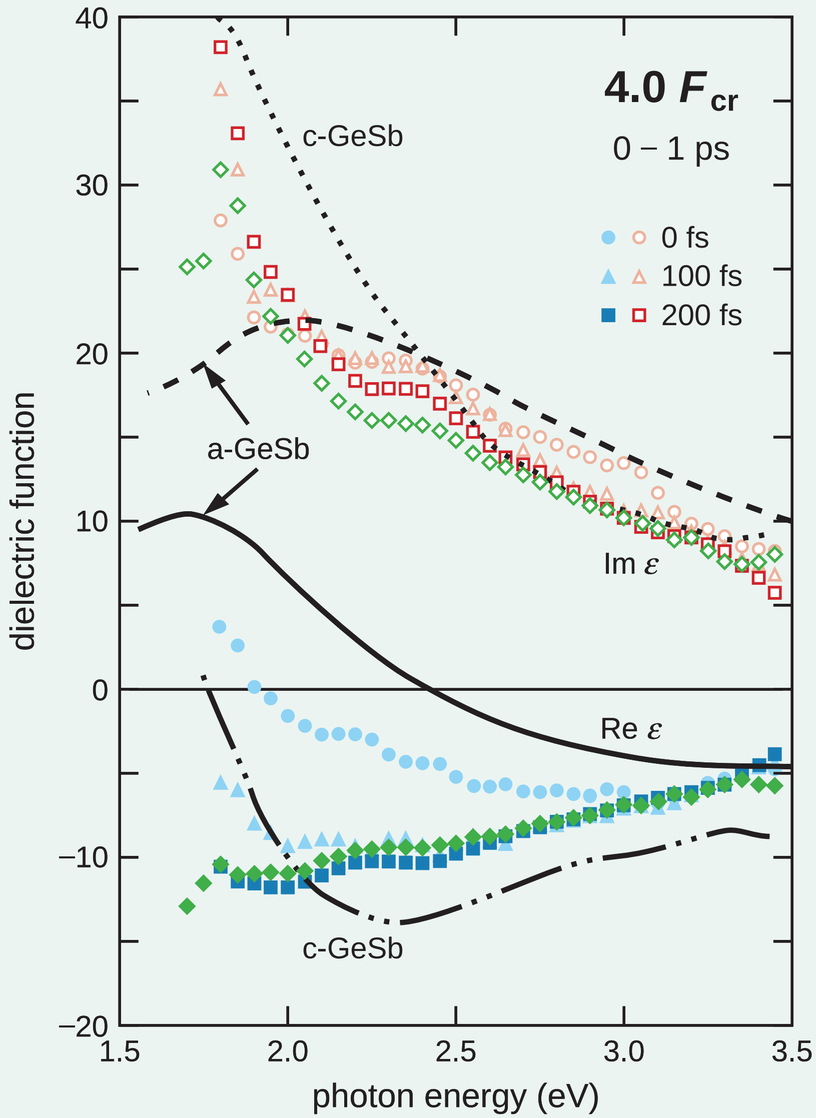
Evolution of ε(ω) after excitation at 4.0 F_{cr}



ε(ω) gradually shifts downwards

ε(ω) reaches intermediate state faster

Evolution of ε(ω) after excitation at 4.0 F_{cr}



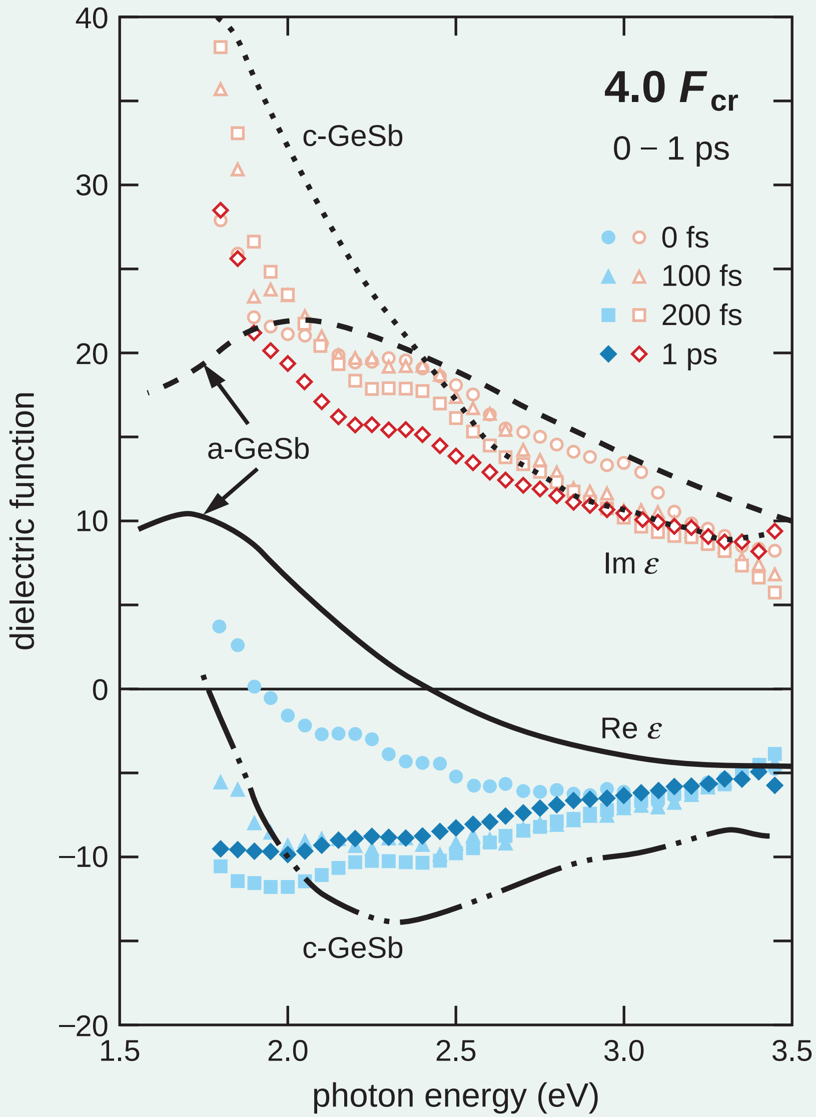
ε(ω) gradually shifts downwards

ε(ω) reaches intermediate state faster

intermediate state is fluence independent → new phase

◇ ◆ 1ps at 1.6 F_{cr}

Evolution of ε(ω) after excitation at 4.0 F_{cr}



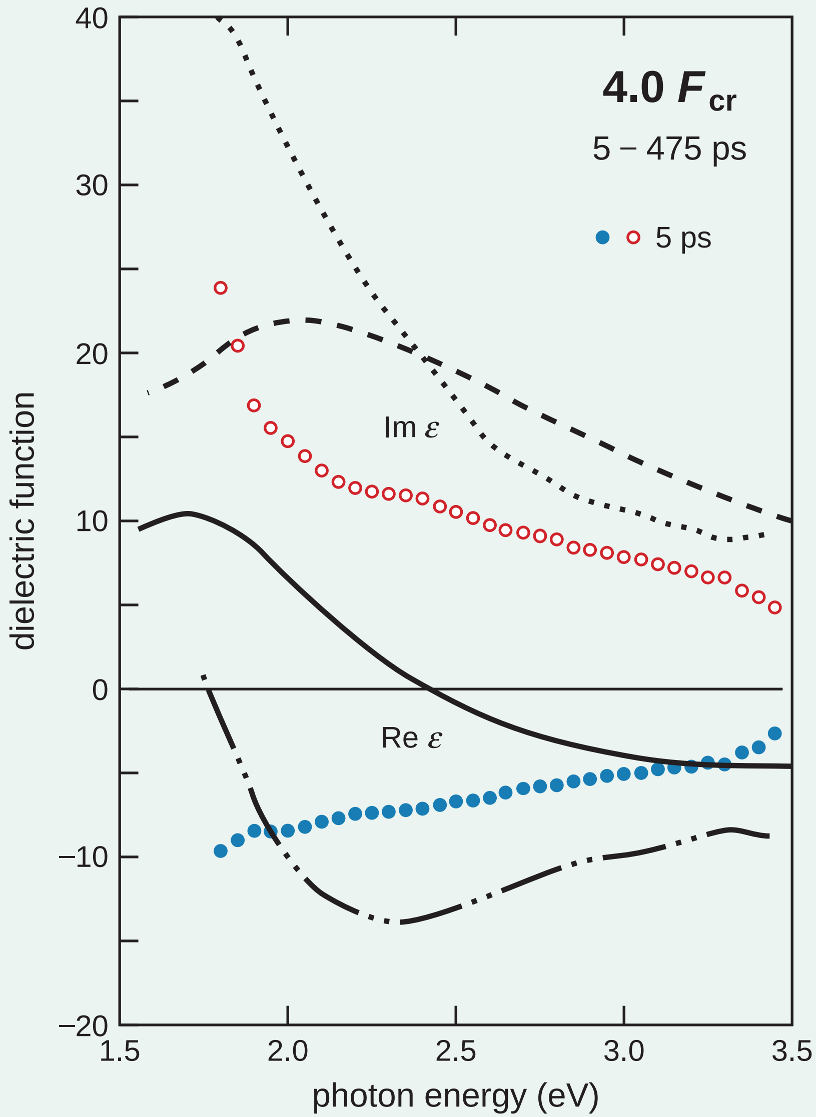
ε(ω) gradually shifts downwards

ε(ω) reaches intermediate state faster

intermediate state is fluence independent → new phase

RESULTS

Evolution of ε(ω) after excitation at 4.0 F_{cr}



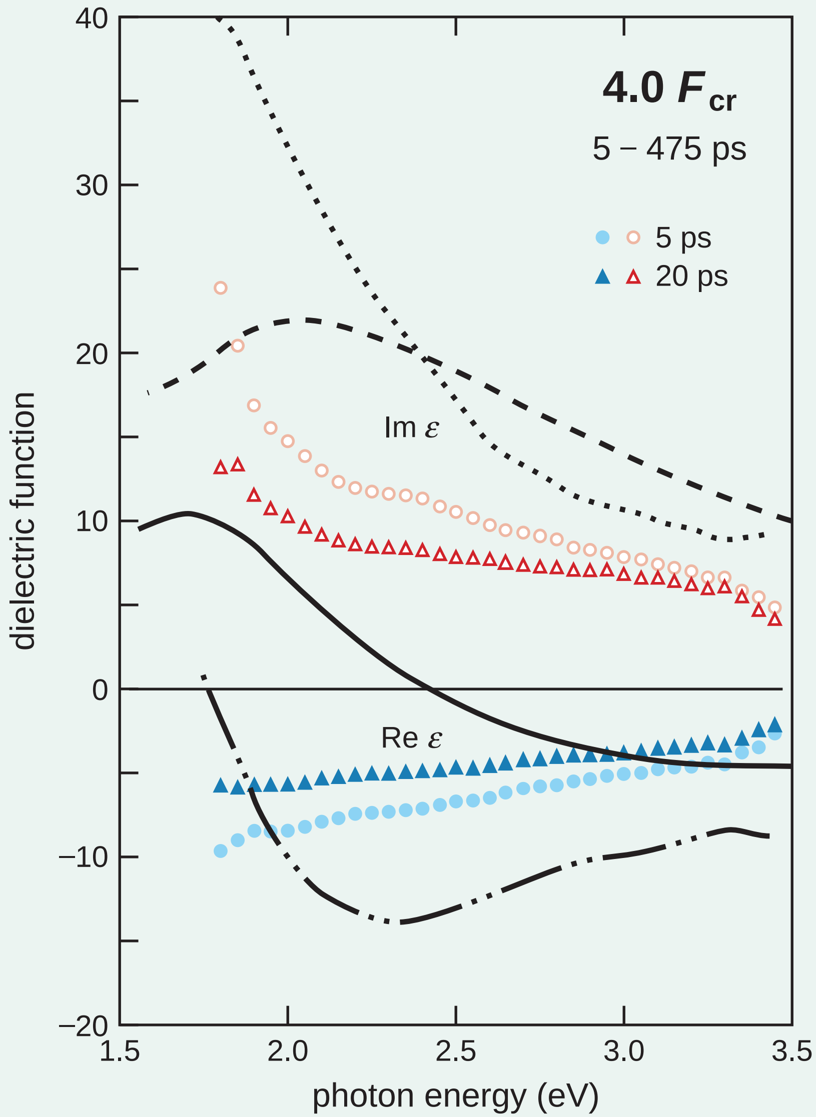
ε(ω) gradually shifts downwards

ε(ω) reaches intermediate state faster

intermediate state is fluence independent → new phase

RESULTS

Evolution of ε(ω) after excitation at 4.0 F_{cr}



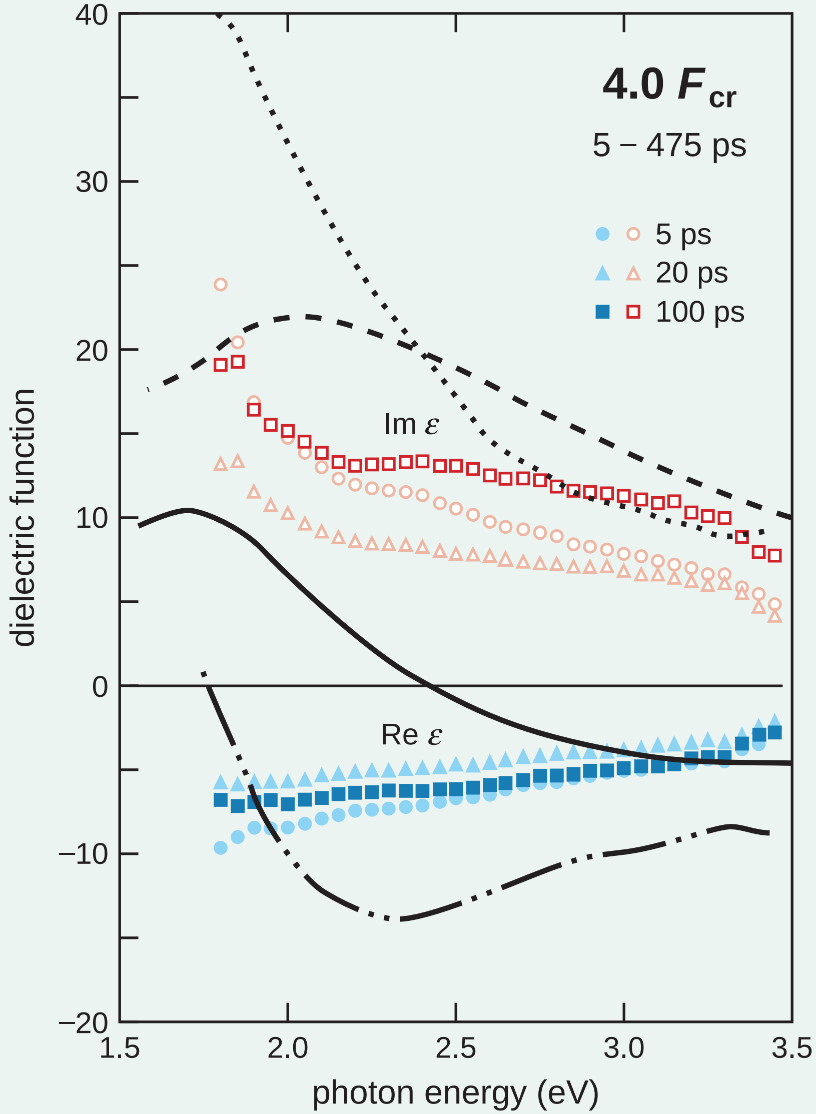
ε(ω) gradually shifts downwards

ε(ω) reaches intermediate state faster

intermediate state is fluence independent → new phase

dynamics at late time delays

Evolution of ε(ω) after excitation at 4.0 F_{cr}



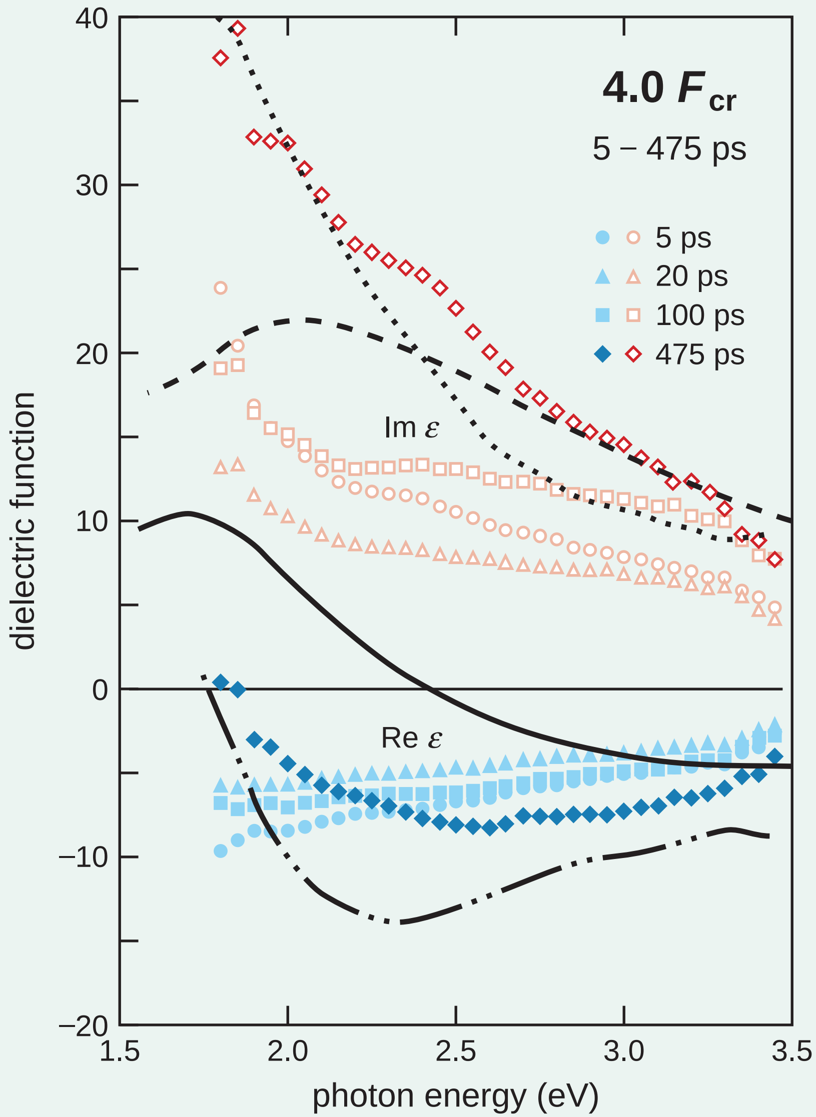
ε(ω) gradually shifts downwards

ε(ω) reaches intermediate state faster

intermediate state is fluence independent → new phase

dynamics at late time delays

Evolution of ε(ω) after excitation at 4.0 F_{cr}



ε(ω) gradually shifts downwards

ε(ω) reaches intermediate state faster

intermediate state is fluence independent → new phase

dynamics at late time delays

first signs of recrystallization after 0.5 ns

Comparison with previous results

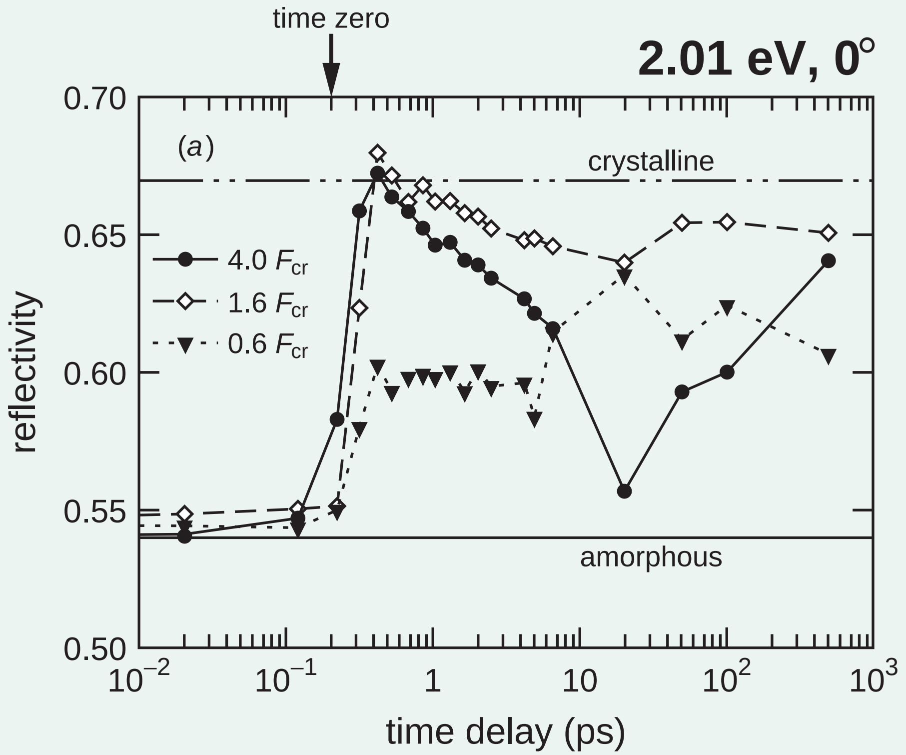
Given time resolved $\varepsilon(\omega)$

Comparison with previous results

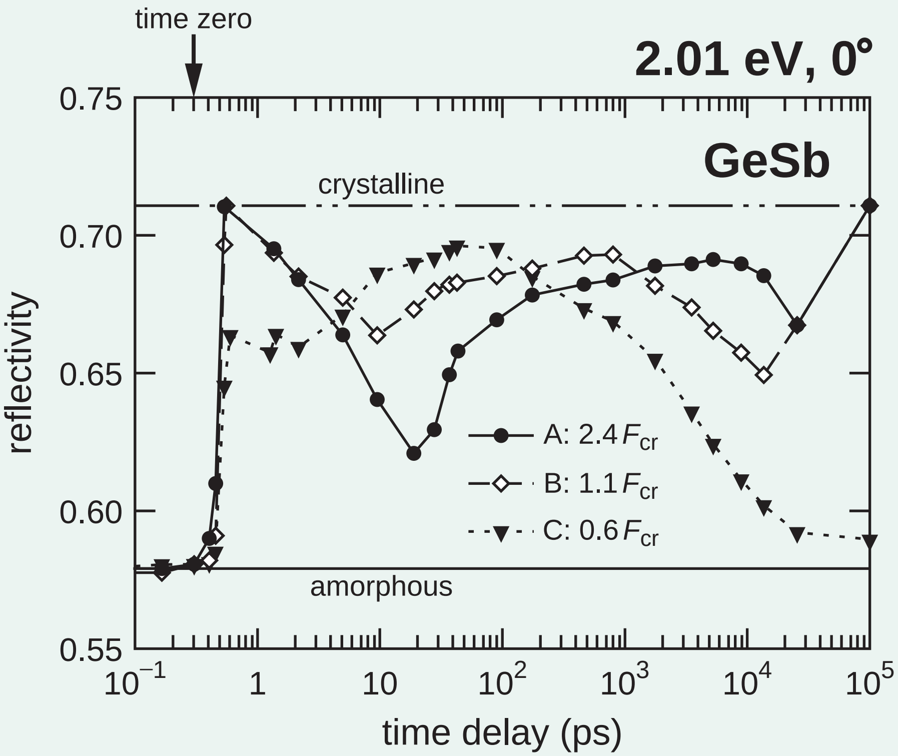
Given time resolved $\varepsilon(\omega)$ —Fresnel Formulae→

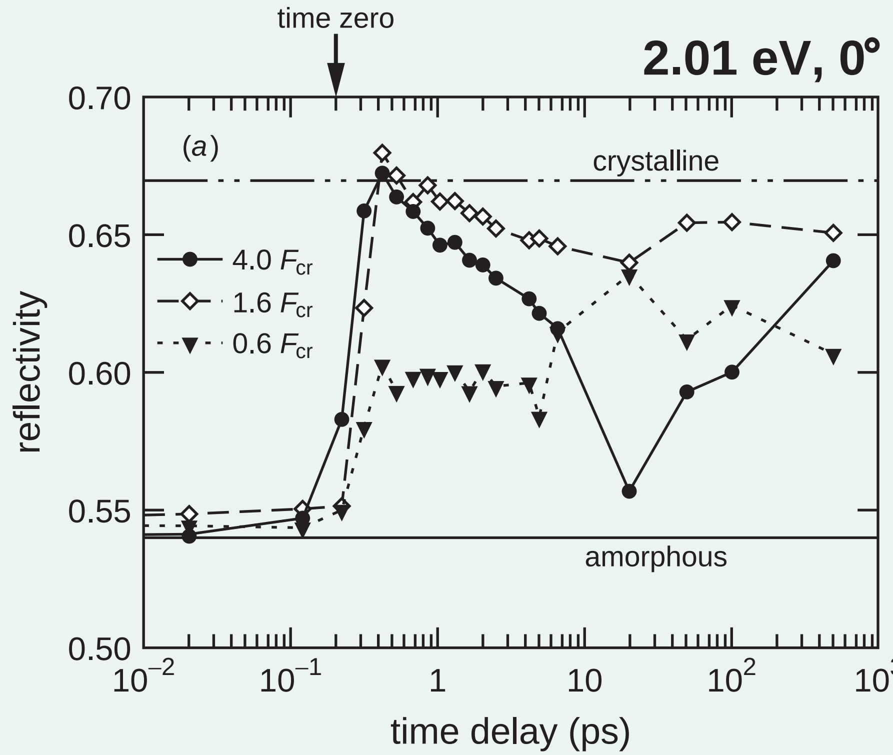
Comparison with previous results

Given time resolved ε(ω) → Fresnel Formulae



Comparison with previous results

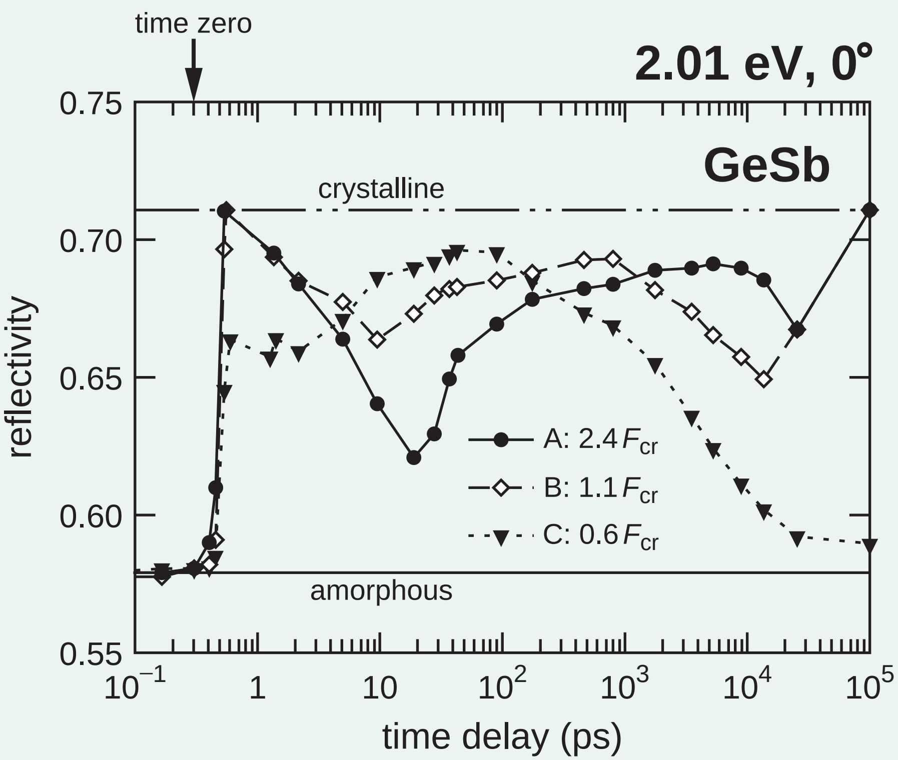


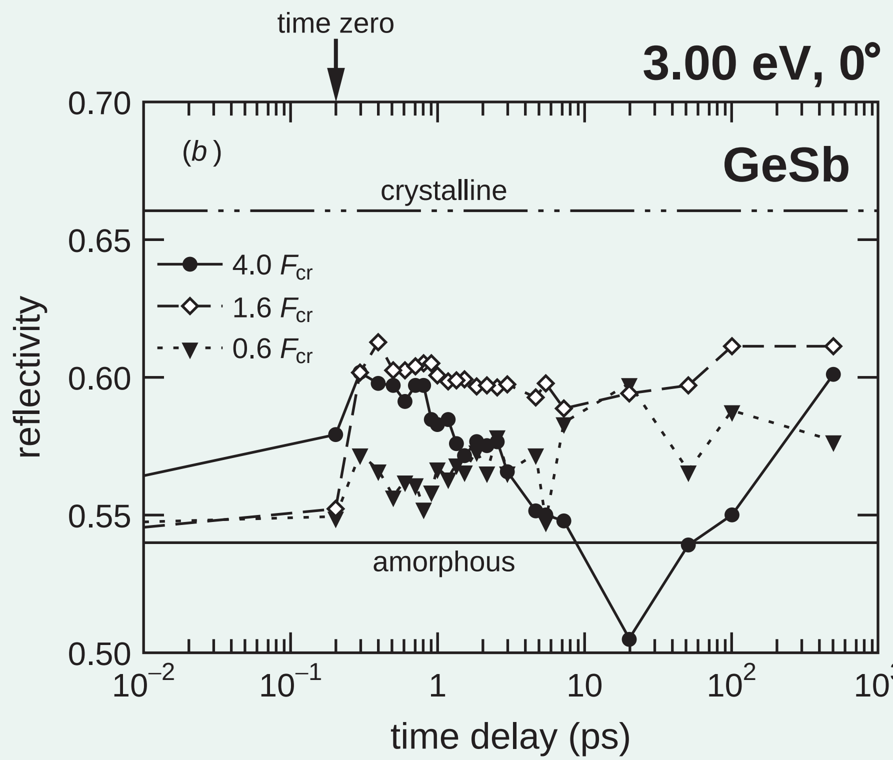


Excellent agreement at 2.01 eV and 0^{o} angle of incidence.

Sokolowski-Tinten et al. PRL, **81**, 3670 (1998)

Comparison with previous results





For other parameters distinction of new phase from c-GeSb becomes evident.

Sokolowski-Tinten et al. PRL, **81**, 3670 (1998)

CONCLUSION

New non-thermal phase of Ge-rich Sb films

CONCLUSION

New non-thermal phase of Ge-rich Sb films

No ultrafast disorder to order transition in GeSb

CONCLUSION

New non-thermal phase of Ge-rich Sb films

No ultrafast disorder to order transition in GeSb

Femtosecond time-resolved ellipsometry is very powerful tool for probing ultrafast phase changes

ACKNOWLEDGMENTS

Dr. K. Sokolowski-Tinten

Dr. Craig Arnold

For a copy of this talk and

additional information, please visit

http://mazur-www.harvard.edu