Ultrafast Phase Transition Dynamics in GeSb Films

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Motivation to study GeSb

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Experimental Technique

Femtosecond time-resolved ellipsometry

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

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Motivations to study phase transitions in GeSb films

Applications in optical data storage

— optically induce transitions between crystalline and amorphous phases

— $\Delta R/R \sim 18\%$

Motivations to study phase transitions in GeSb films

Applications in optical data storage

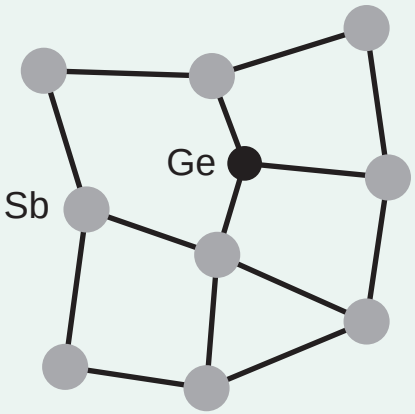
— optically induce transitions between crystalline and amorphous phases

— $\Delta R/R \sim 18\%$

Recently suggested ultrafast disorder-to-order phase transition

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

Amorphous and crystalline phases of GeSb

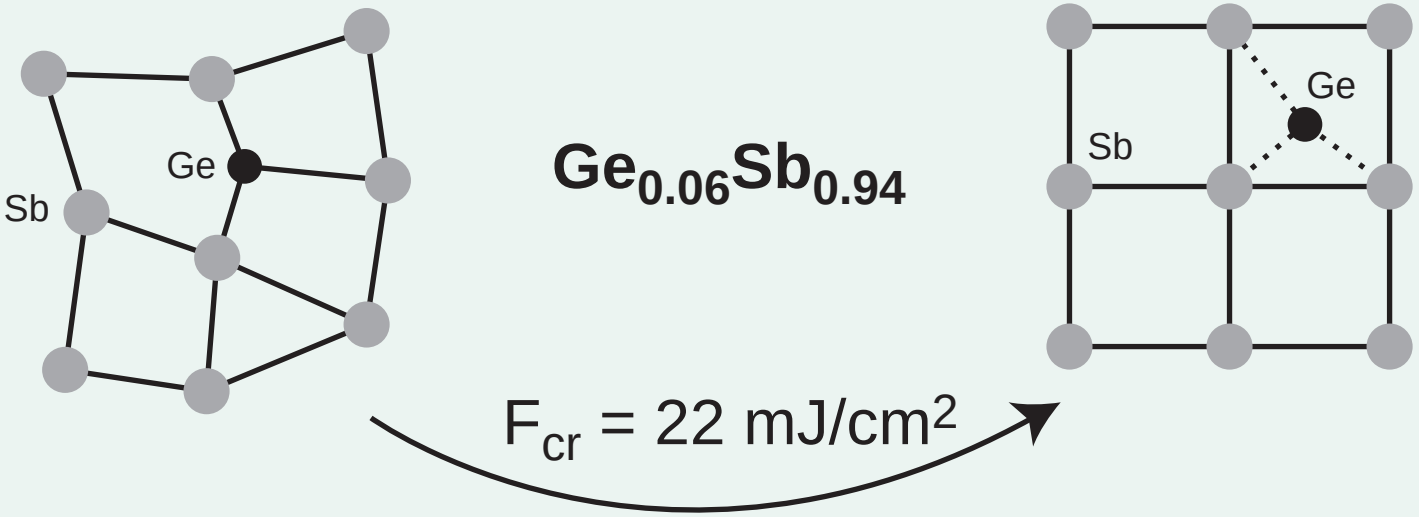


$Ge_{0.06}Sb_{0.94}$

amorphous phase is stabilized by Ge atoms

R ≅ 55%

Amorphous and crystalline phases of GeSb



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crystalline structure identical to pure Sb — solid solution of Ge in Sb
R ≅ 67%

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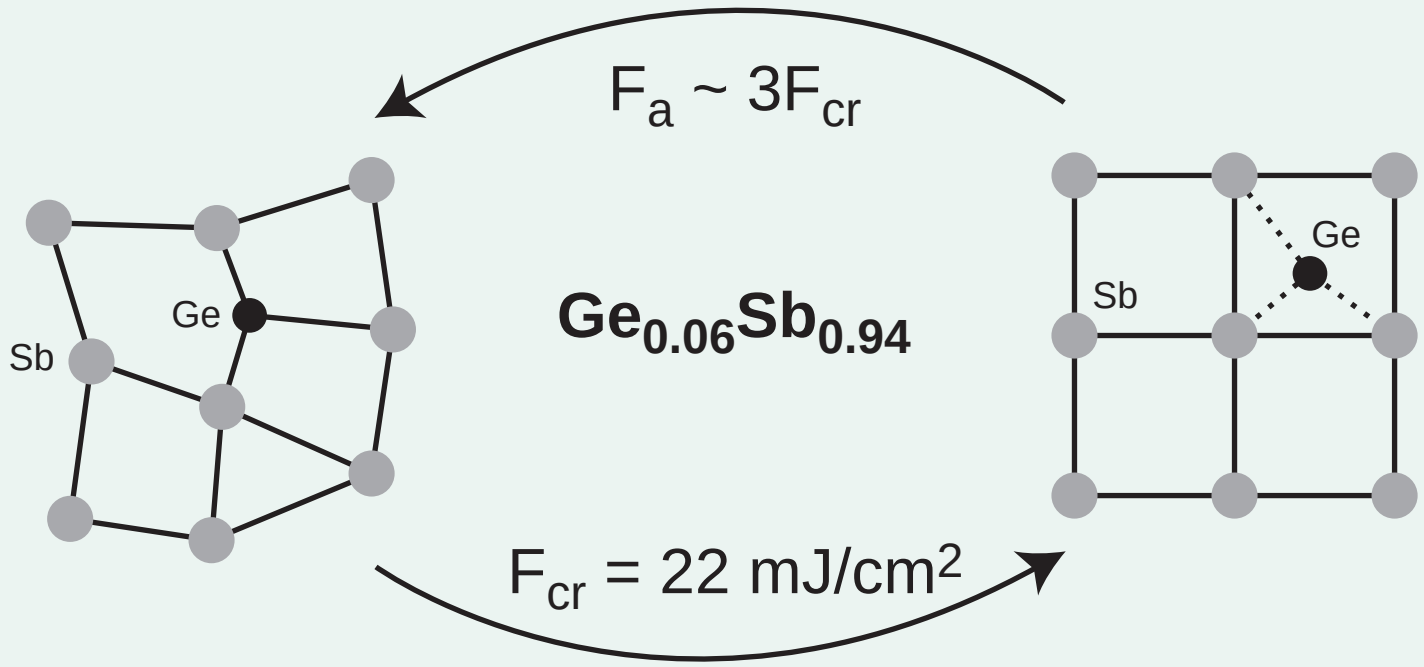

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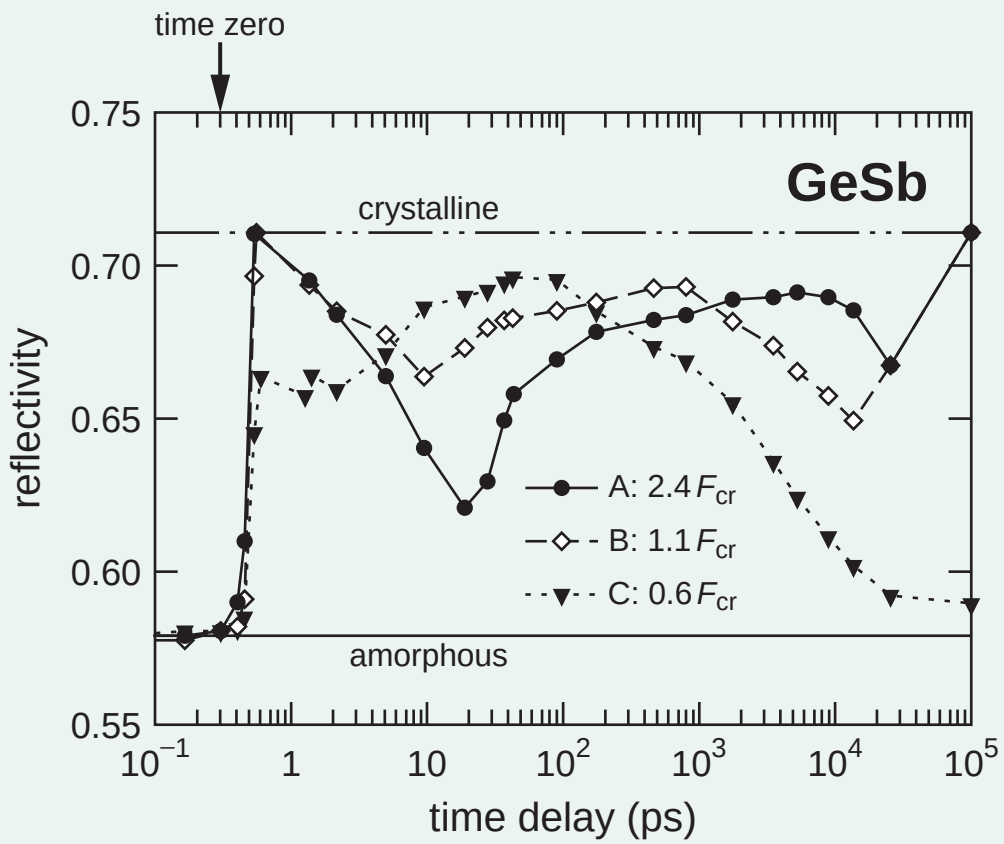
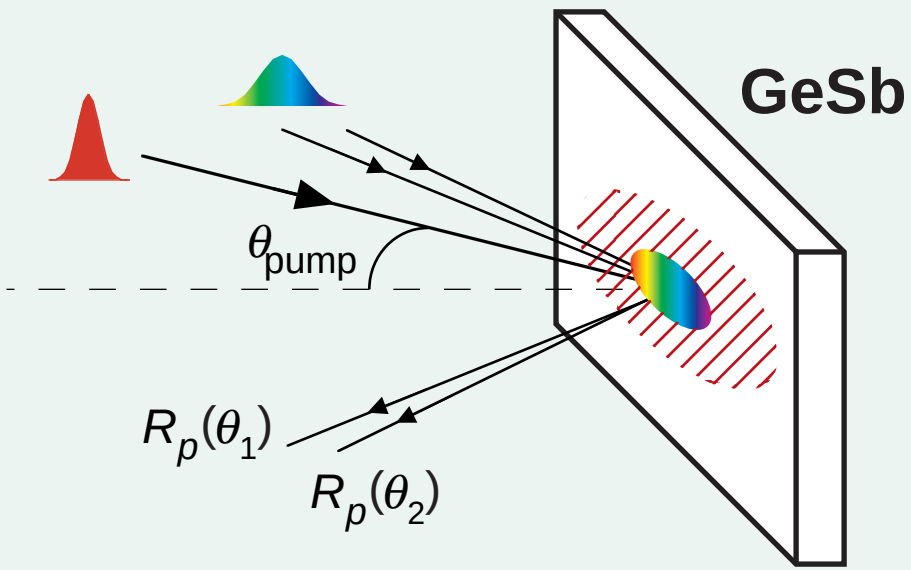
Previous work suggests ultrafast crystallization



Transient reflectivities at 2.01 eV and 0° angle of incidence

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

Time-Resolved Ellipsometry

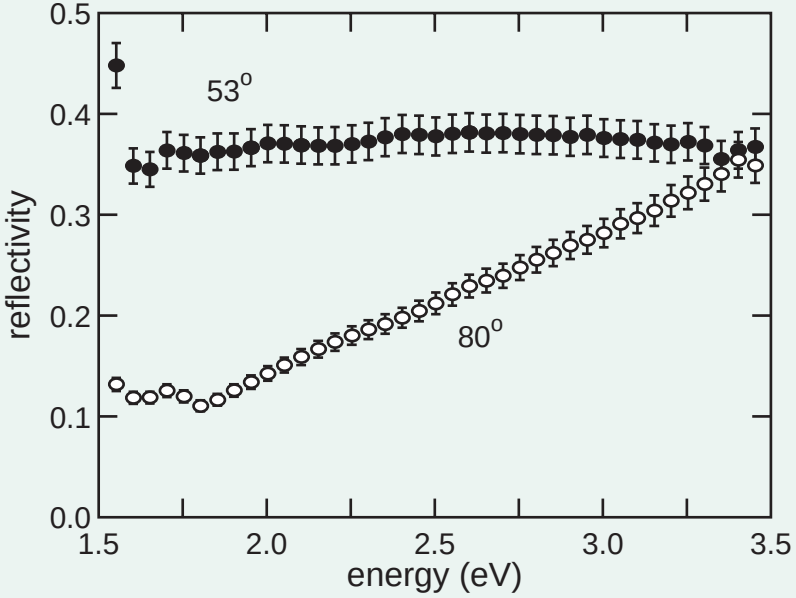


pump pulse:
- 1.5 eV (800nm)
- up to 500 μJ
- $\theta_{pump} < \theta_1, \theta_2$

probe pulse:
- 1.7 — 3.5 eV (350nm — 750nm)
- < 0.1 μJ
- $\theta_1 = 53^o$, $\theta_2 = 80^o$

Extracting the Dielectric Function

Reflectivity Spectra



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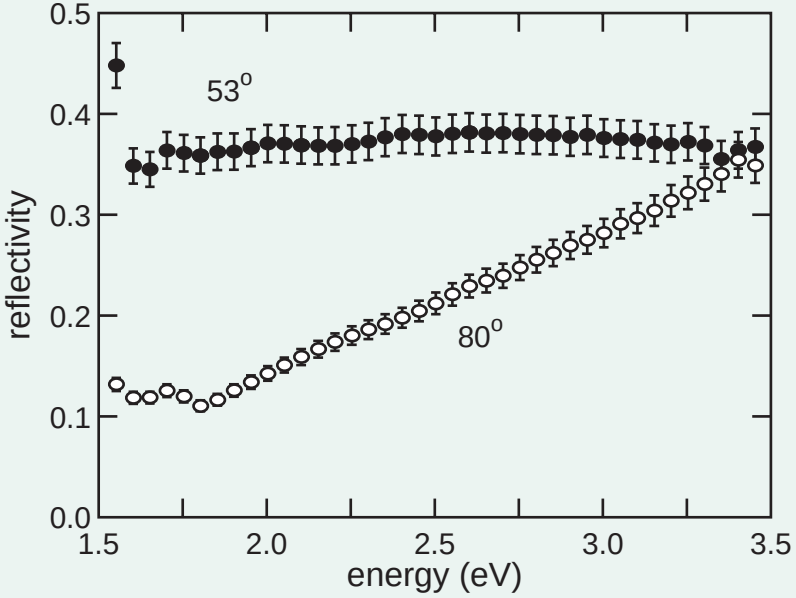

Numerically invert Fresnel formulae

Extracting the Dielectric Function

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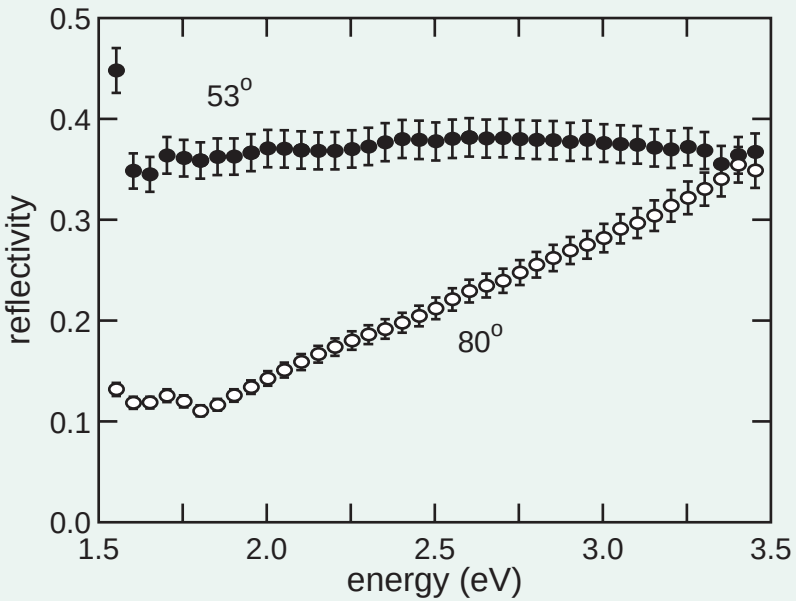
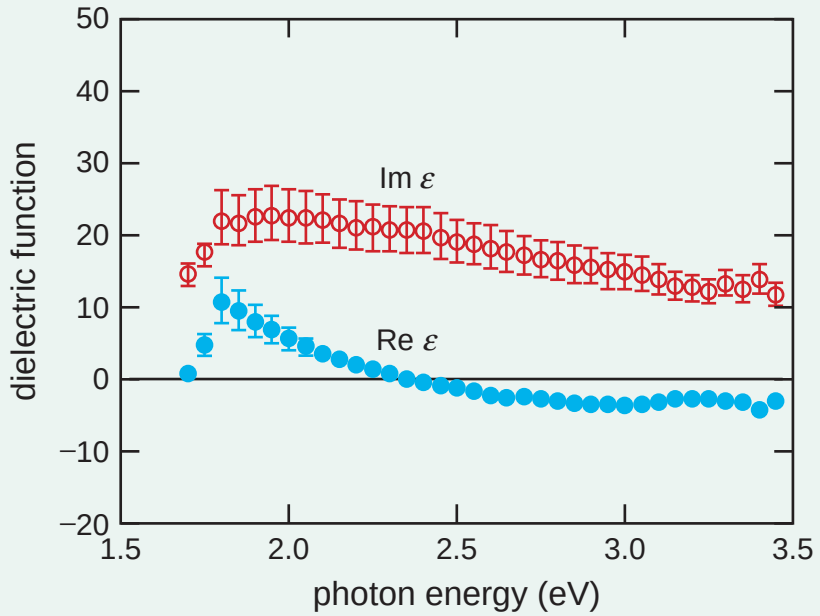


Dielectric Function



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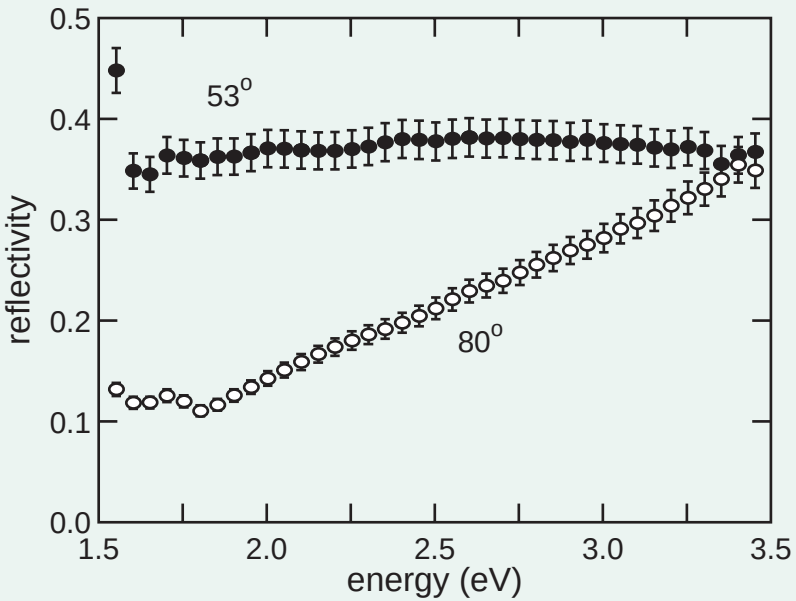
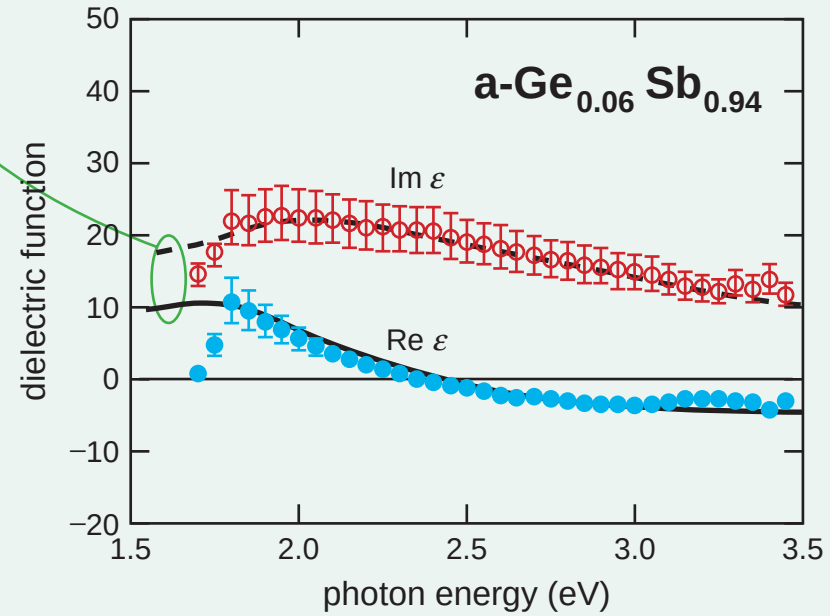
Reflectivity Spectra

cw ellipsometry data

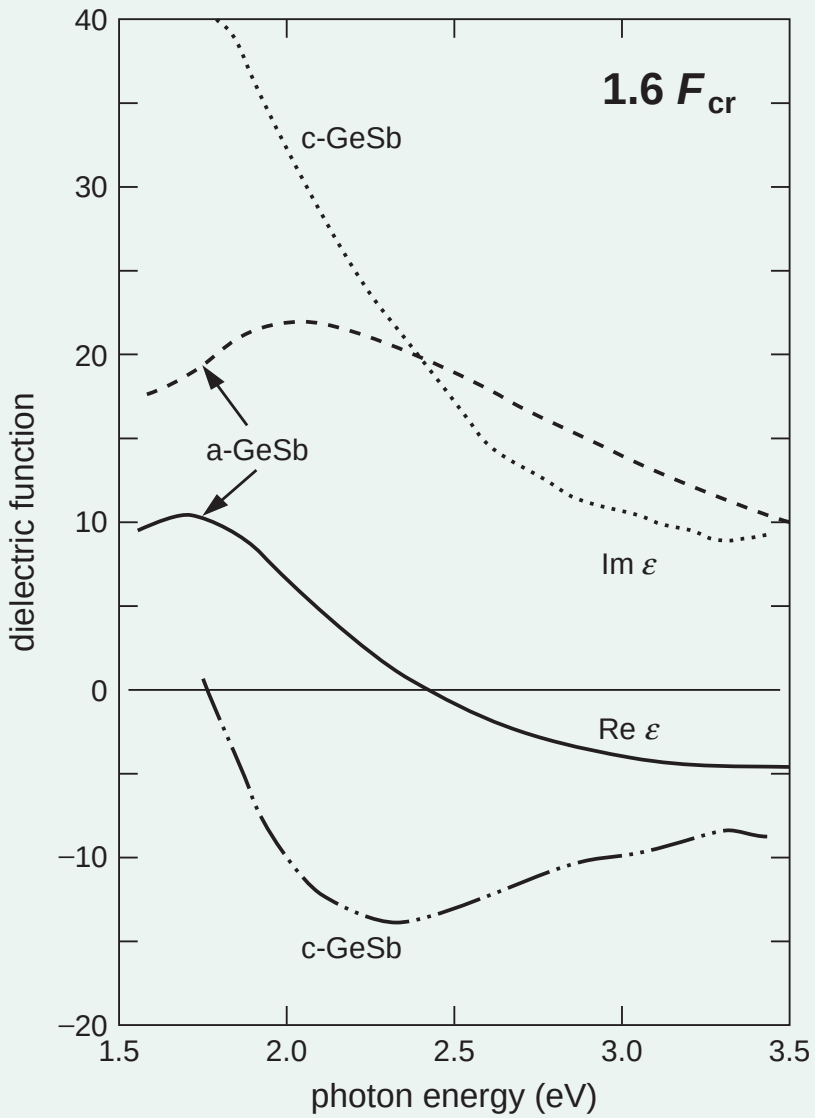
Dielectric Function





Numerically invert Fresnel formulae

Evolution of $\varepsilon(\omega)$ after excitation of $1.6F_{cr}$



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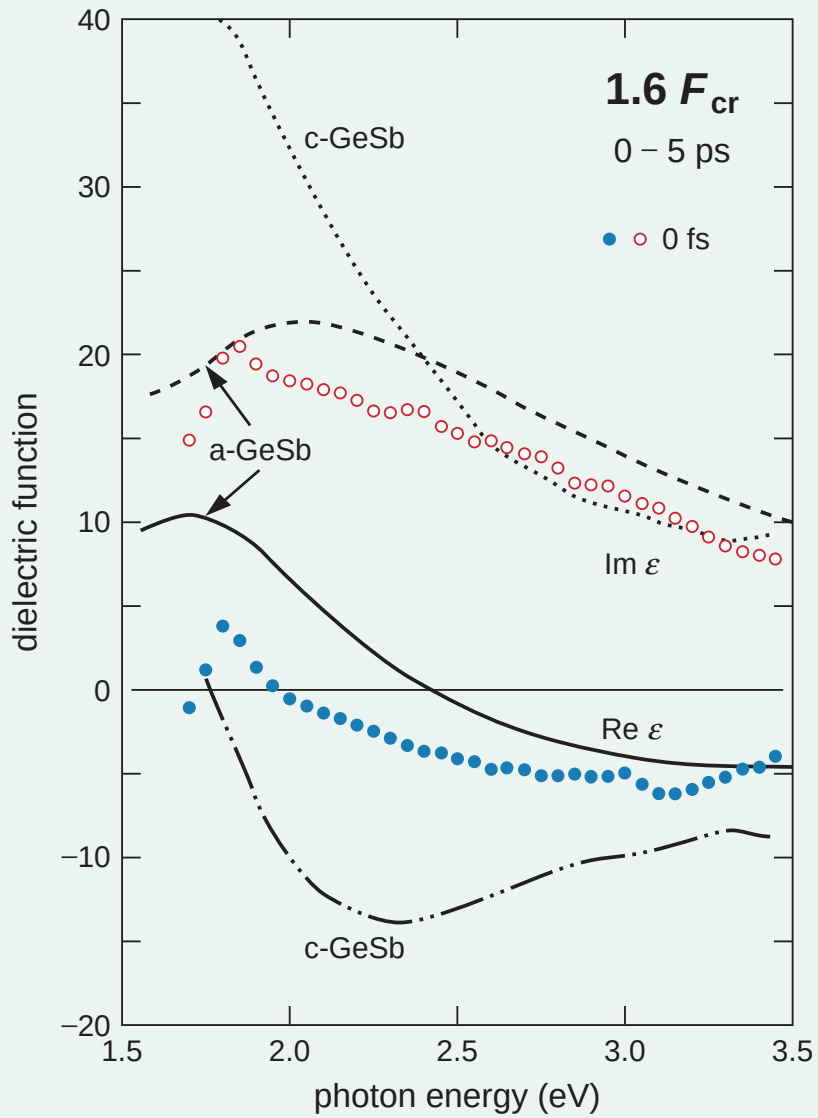

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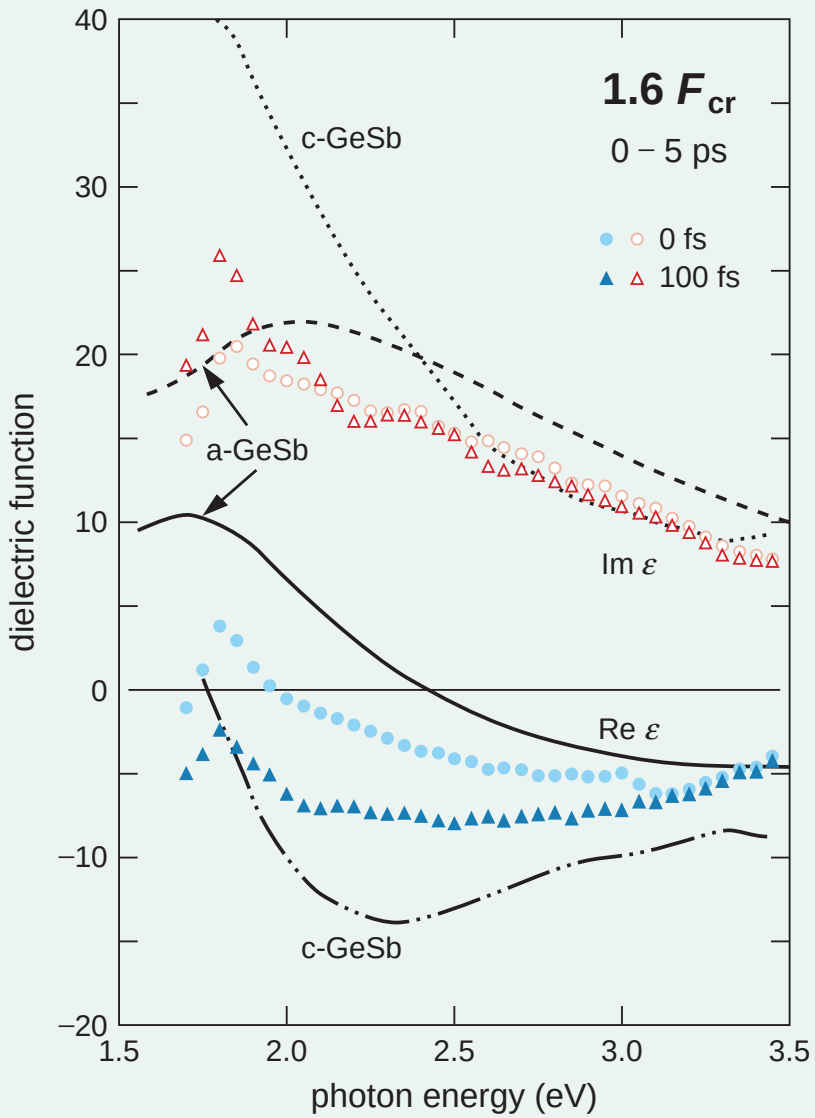

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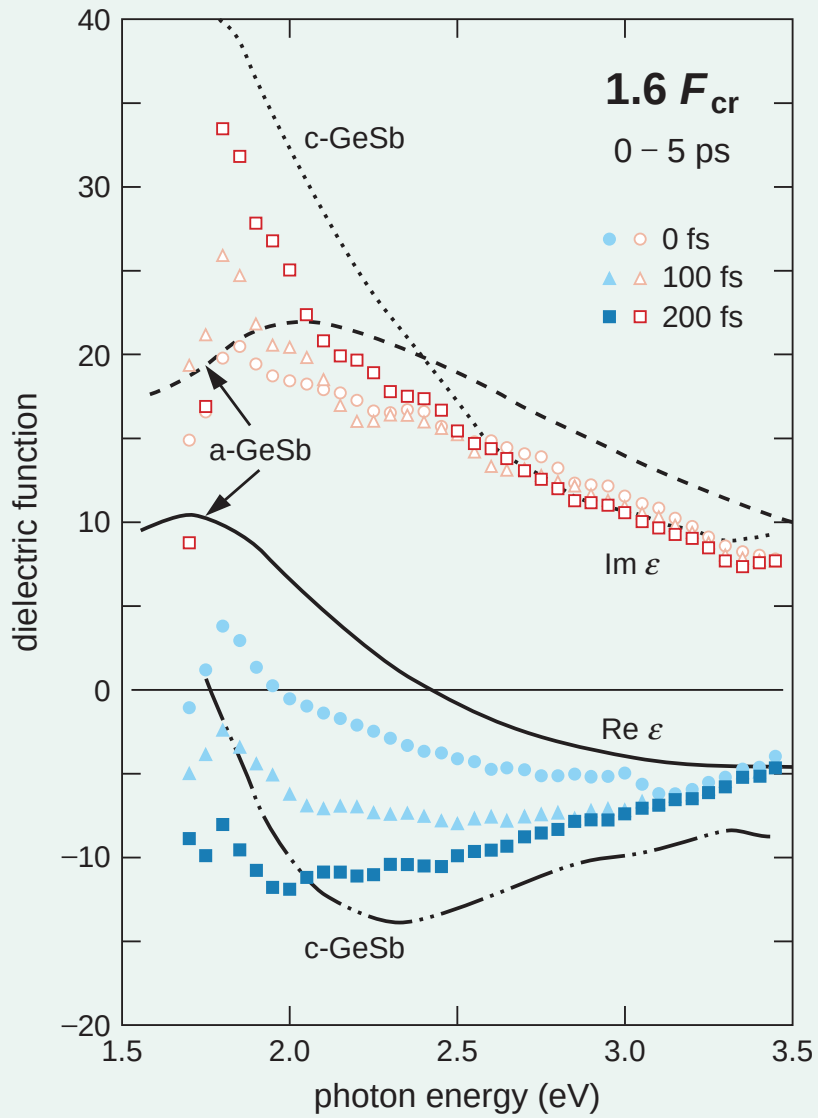

Material does not achieve crystalline phase...

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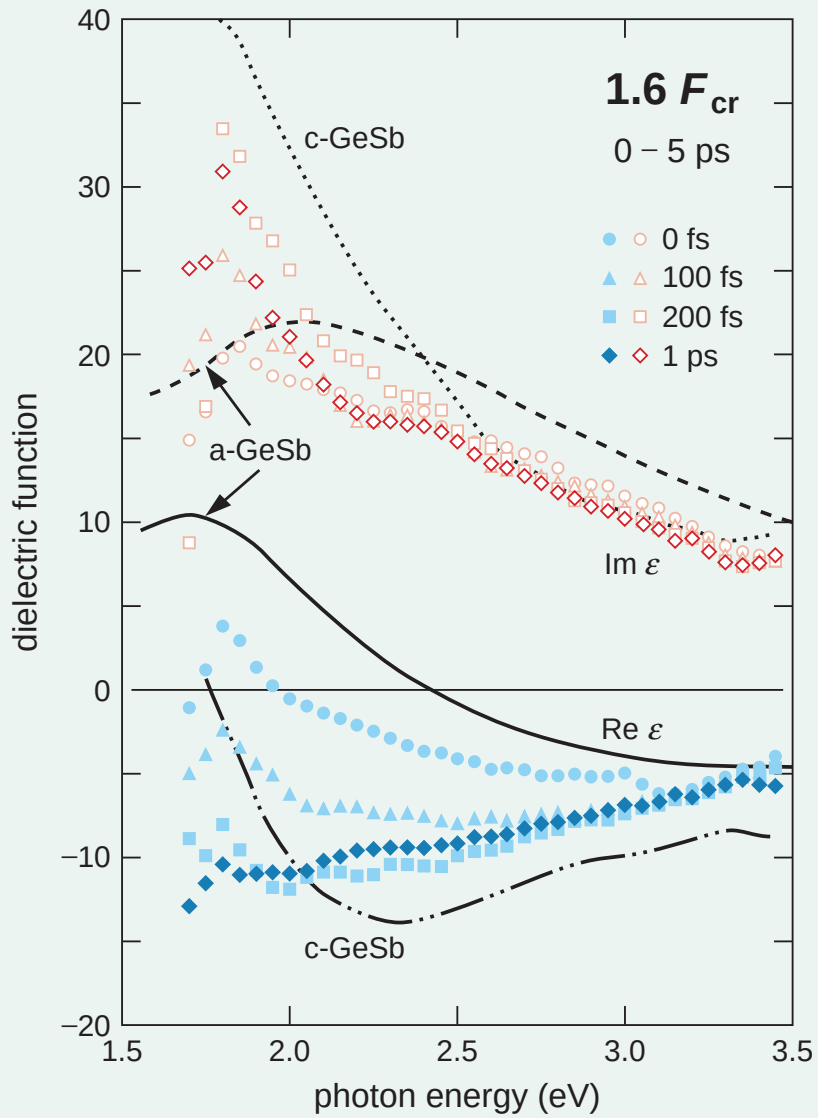
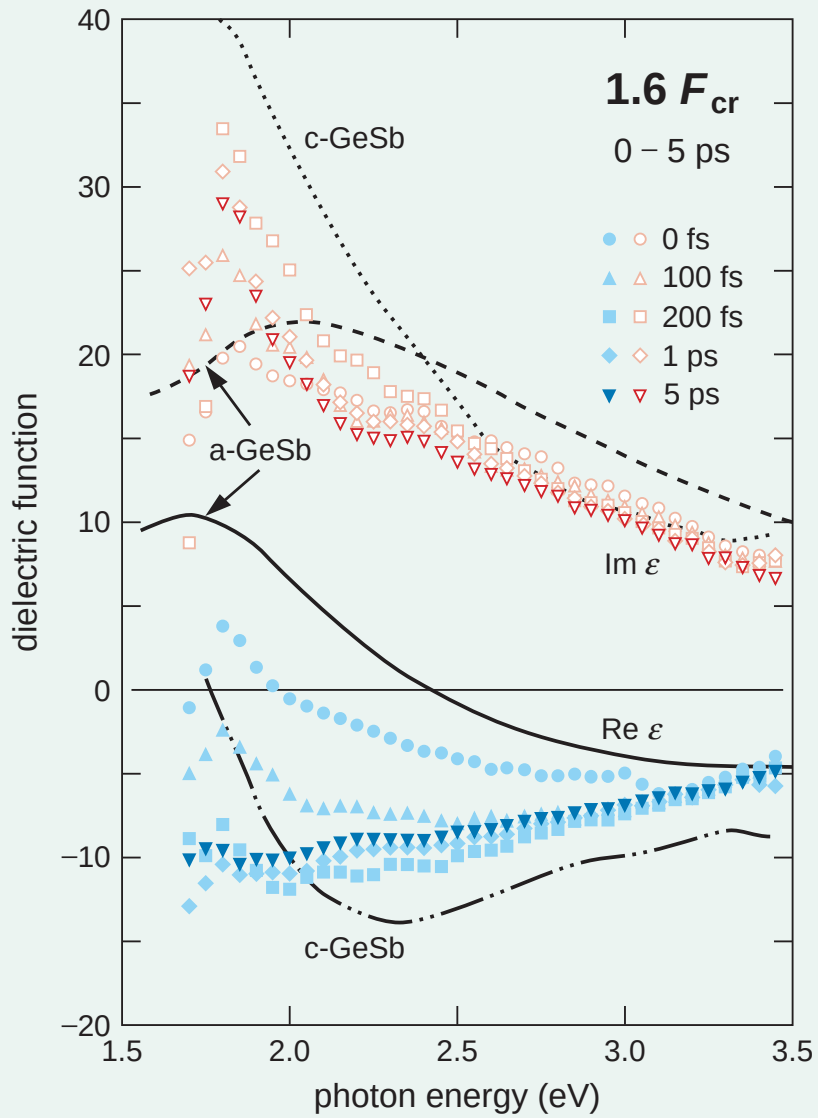


Material does not achieve crystalline phase...

Dynamics stop after 200fs.

Evolution of $\varepsilon(\omega)$ after excitation of $1.6F_{cr}$

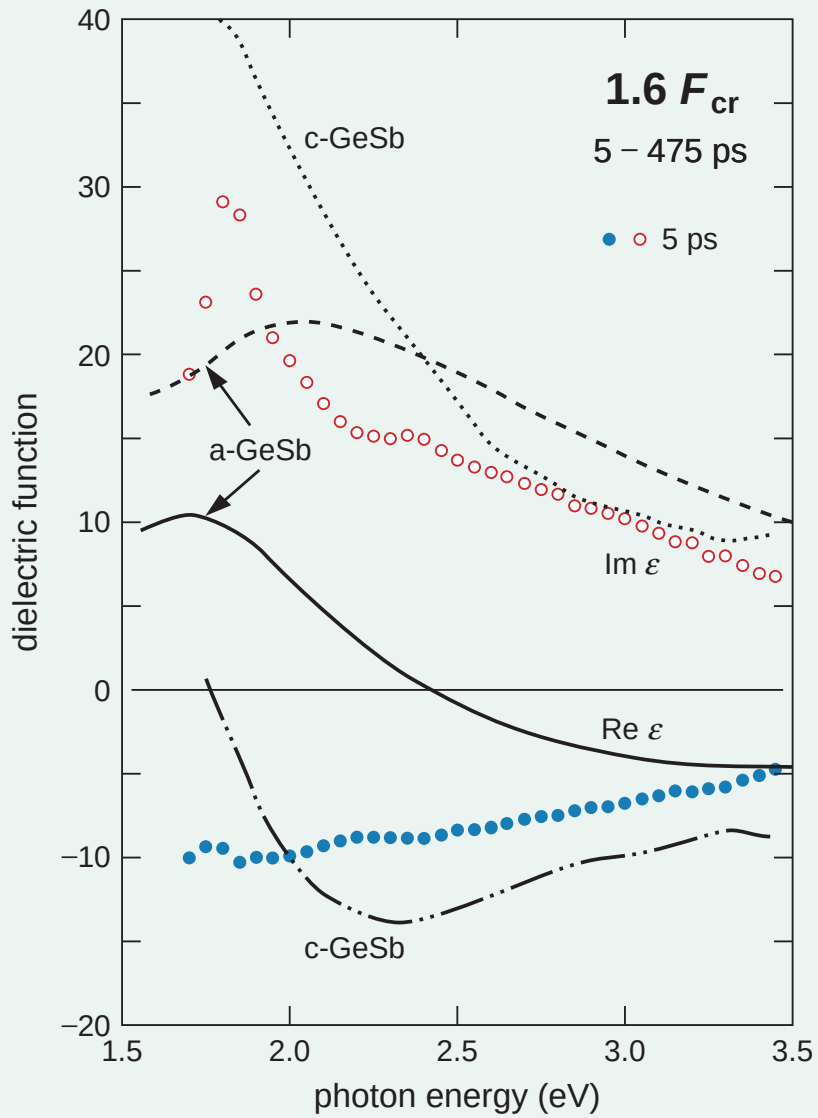


Material does not achieve crystalline phase...

Dynamics stop after 200fs.

Electrons and lattice reach thermal equilibrium:
little change in $\varepsilon(\omega)$.

Evolution of $\varepsilon(\omega)$ after excitation of $1.6F_{cr}$



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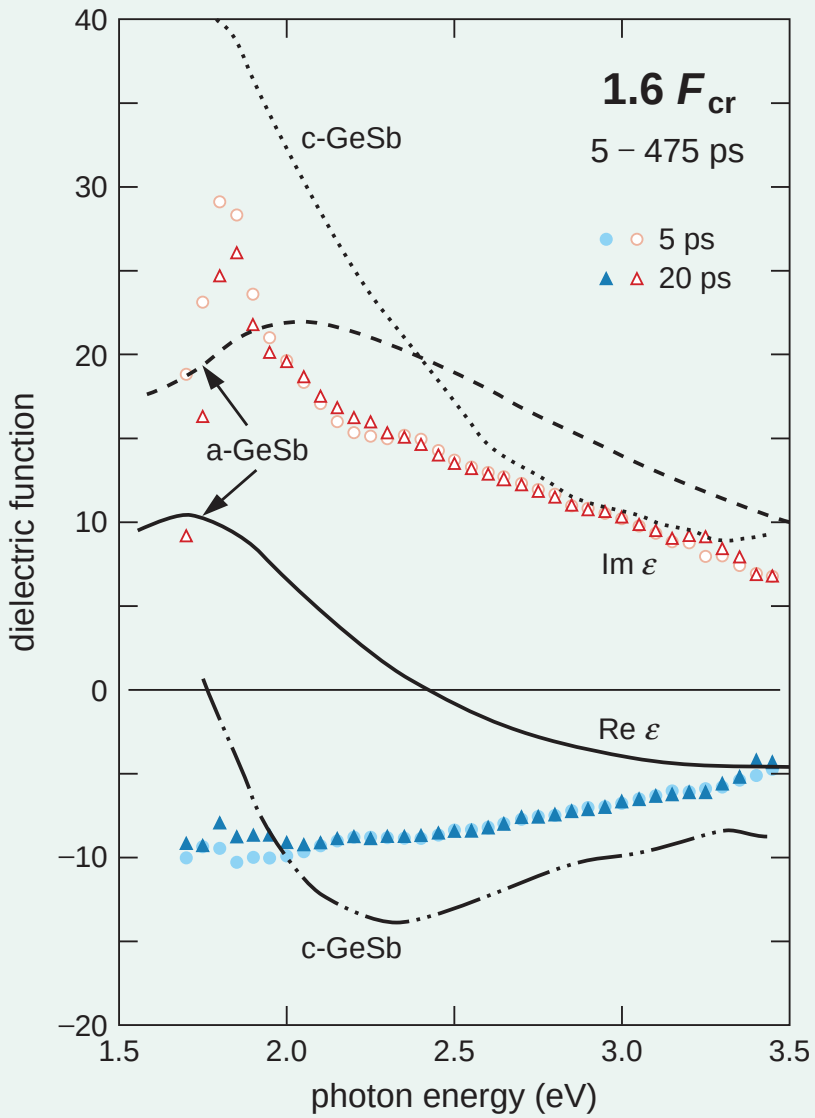

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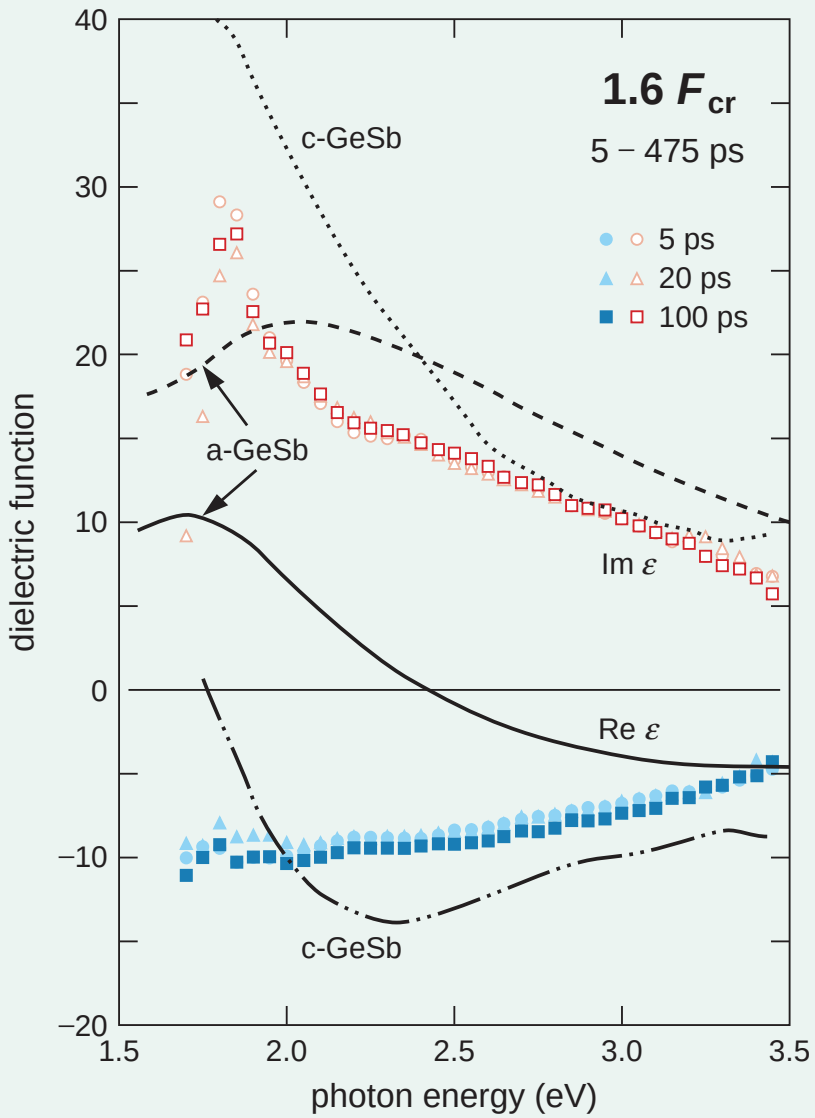

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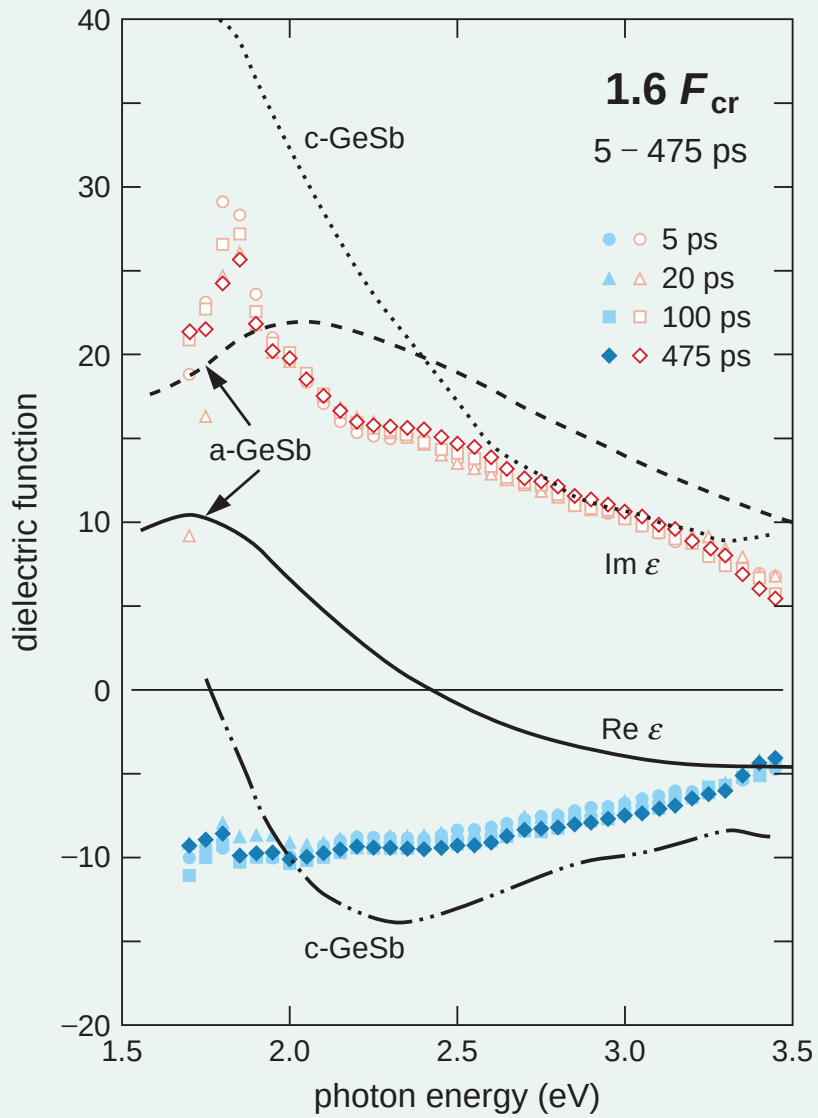
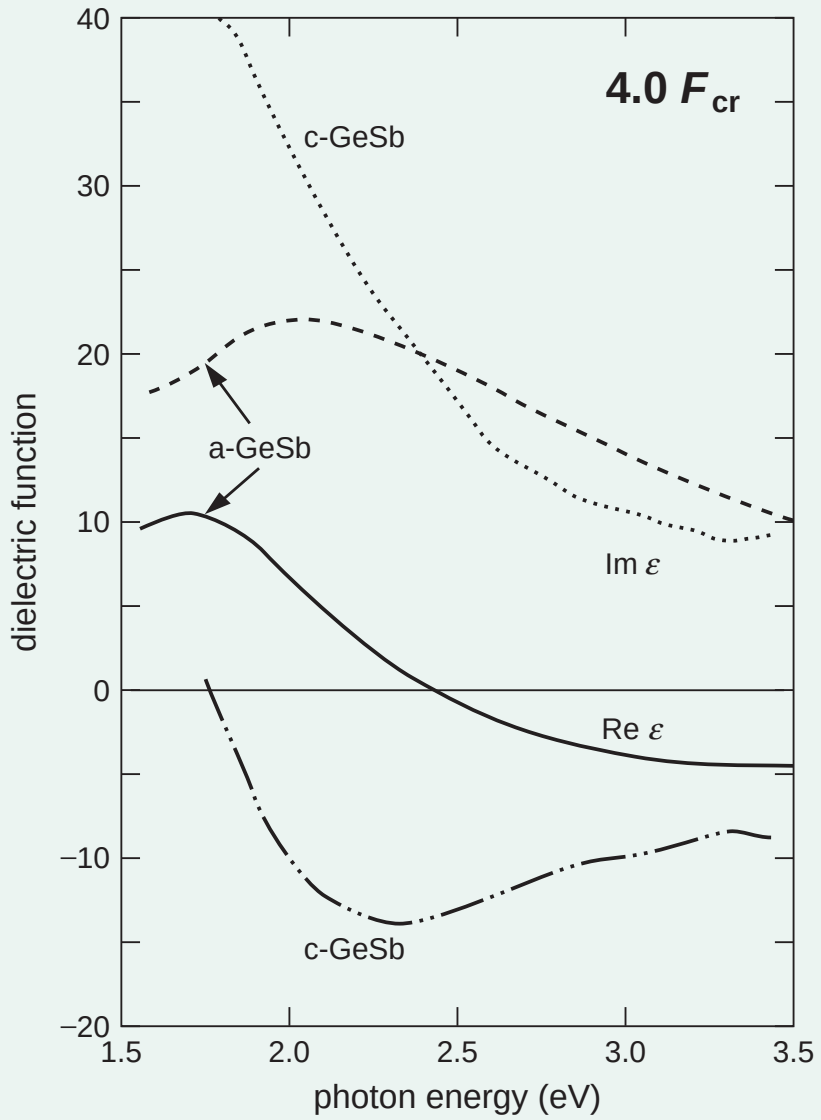
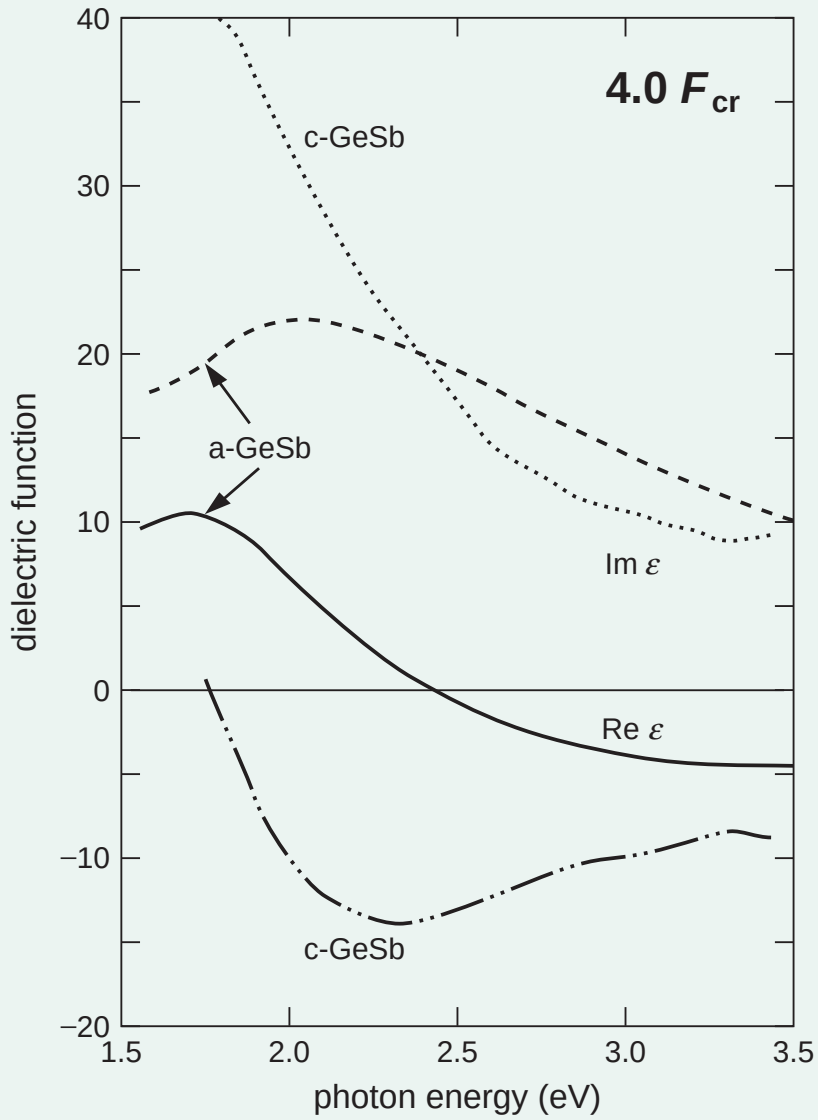


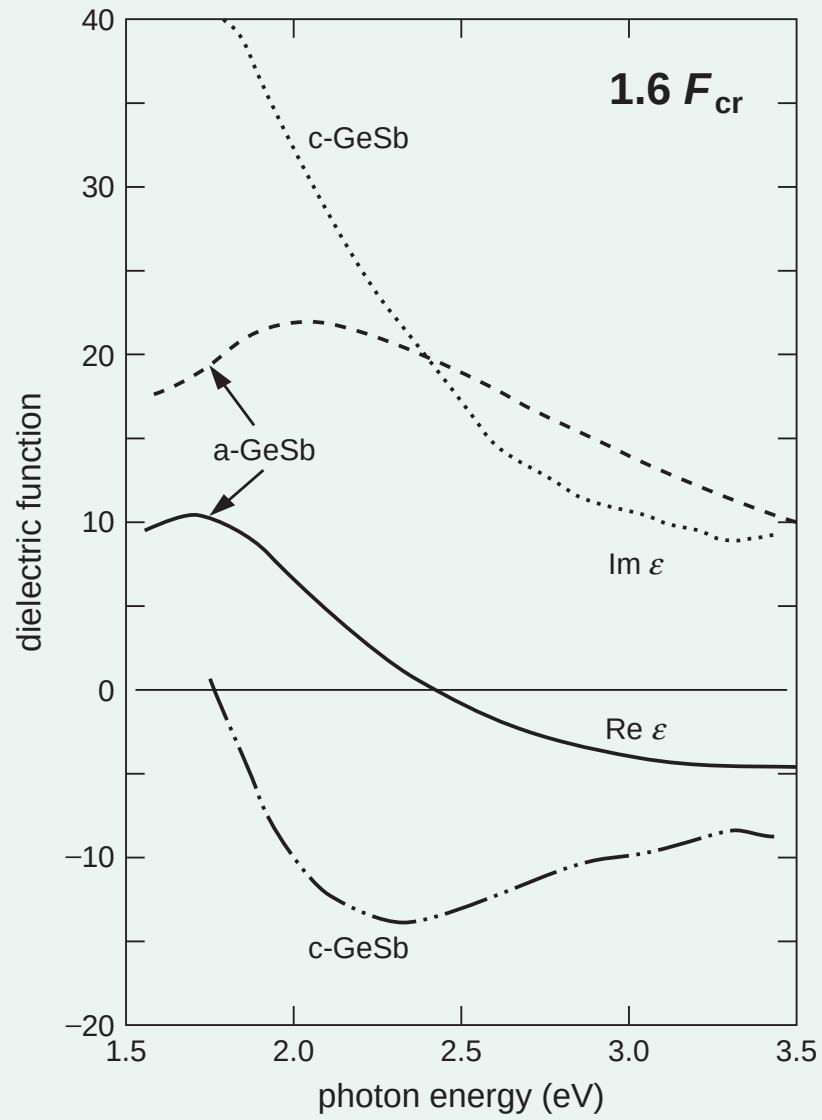
Optical properties constant to ~ 0.5ns.

Evolution of $\varepsilon(\omega)$ after excitation of $4.0F_{cr}$

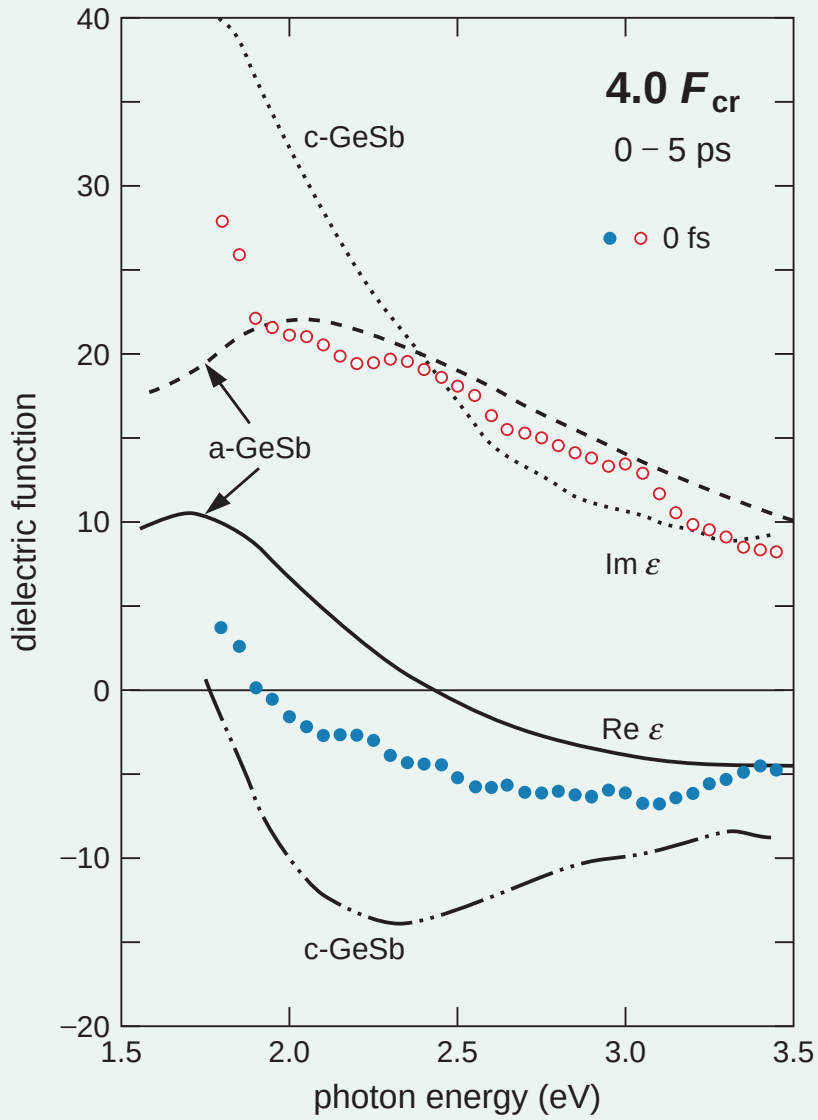


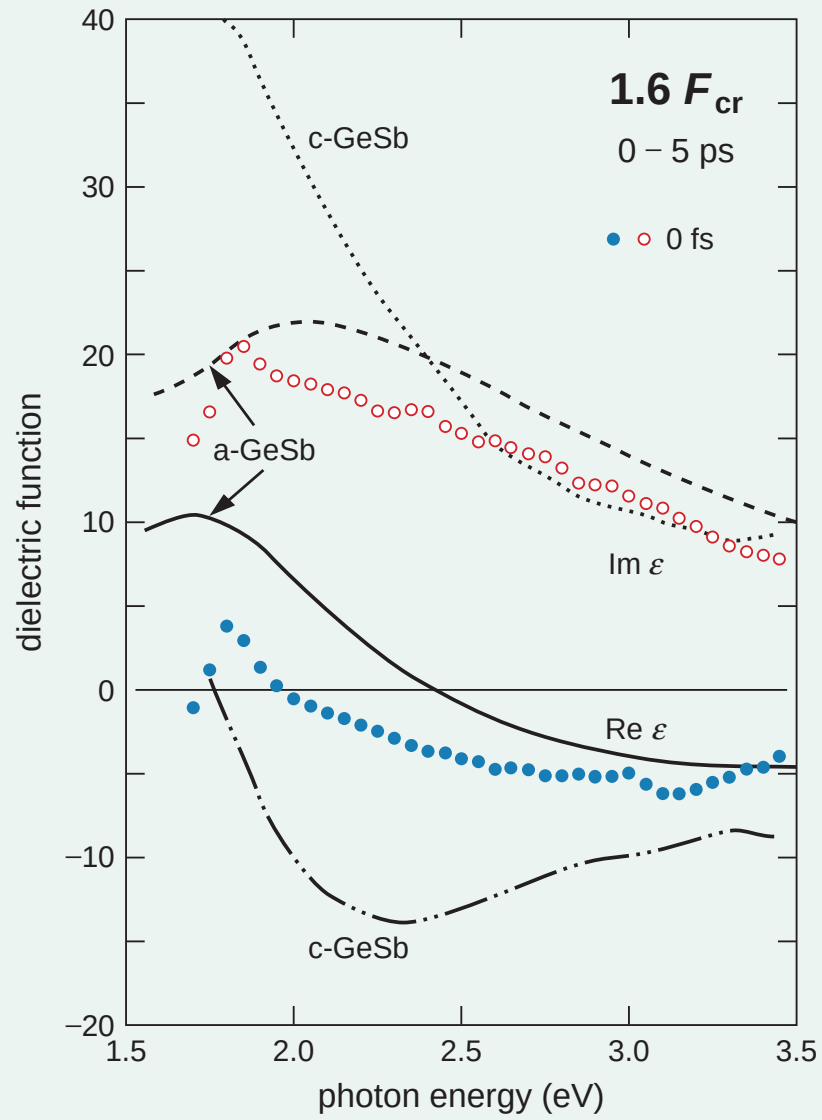
Evolution of $\varepsilon(\omega)$ after excitation of $4.0F_{cr}$





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





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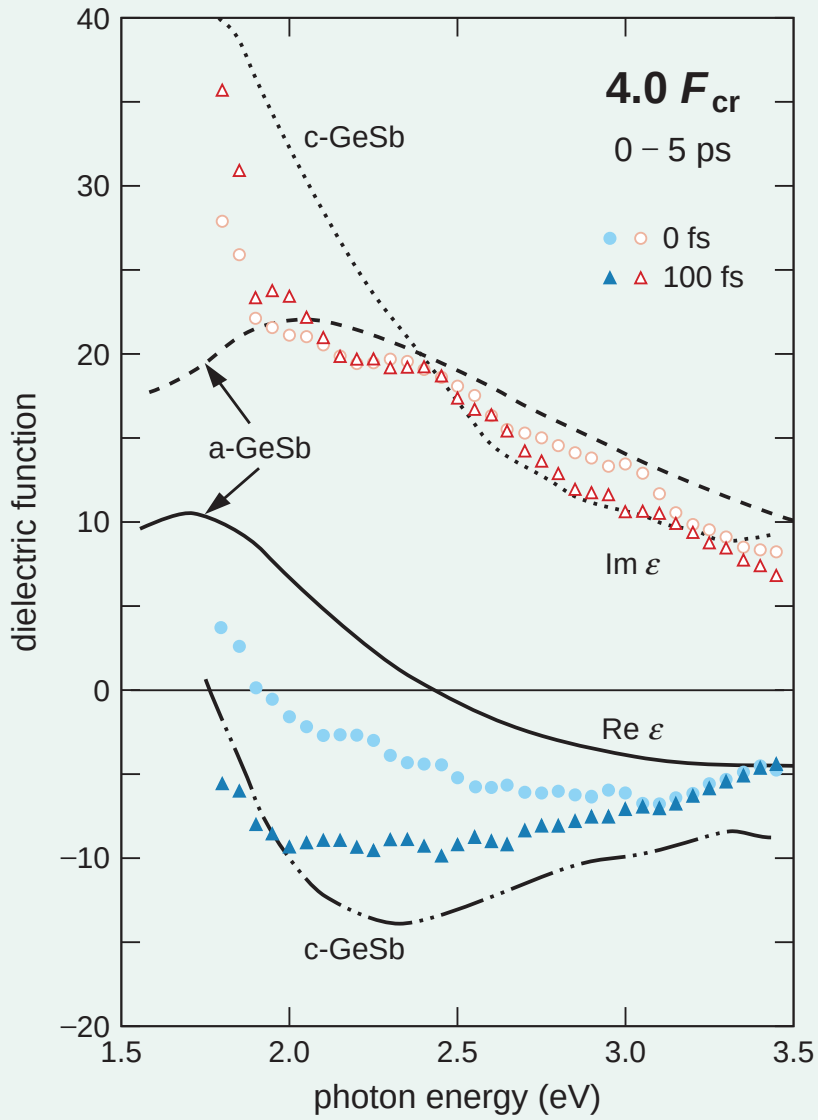

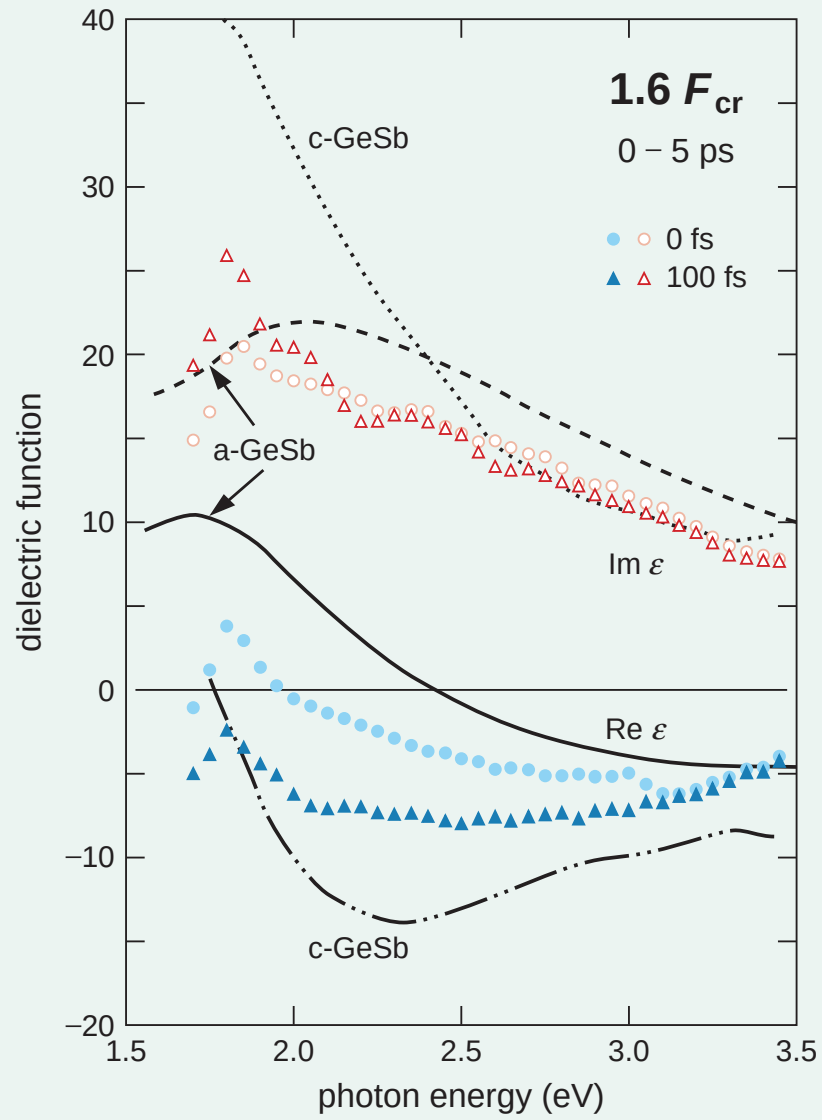

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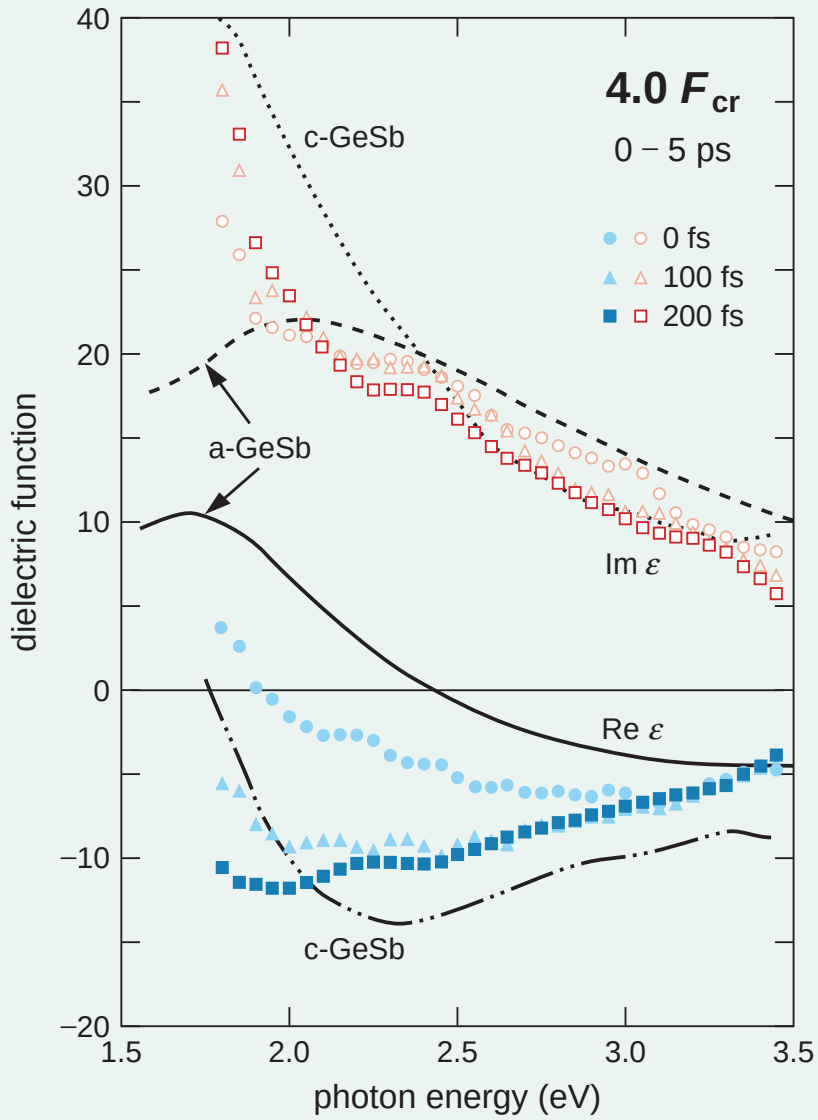

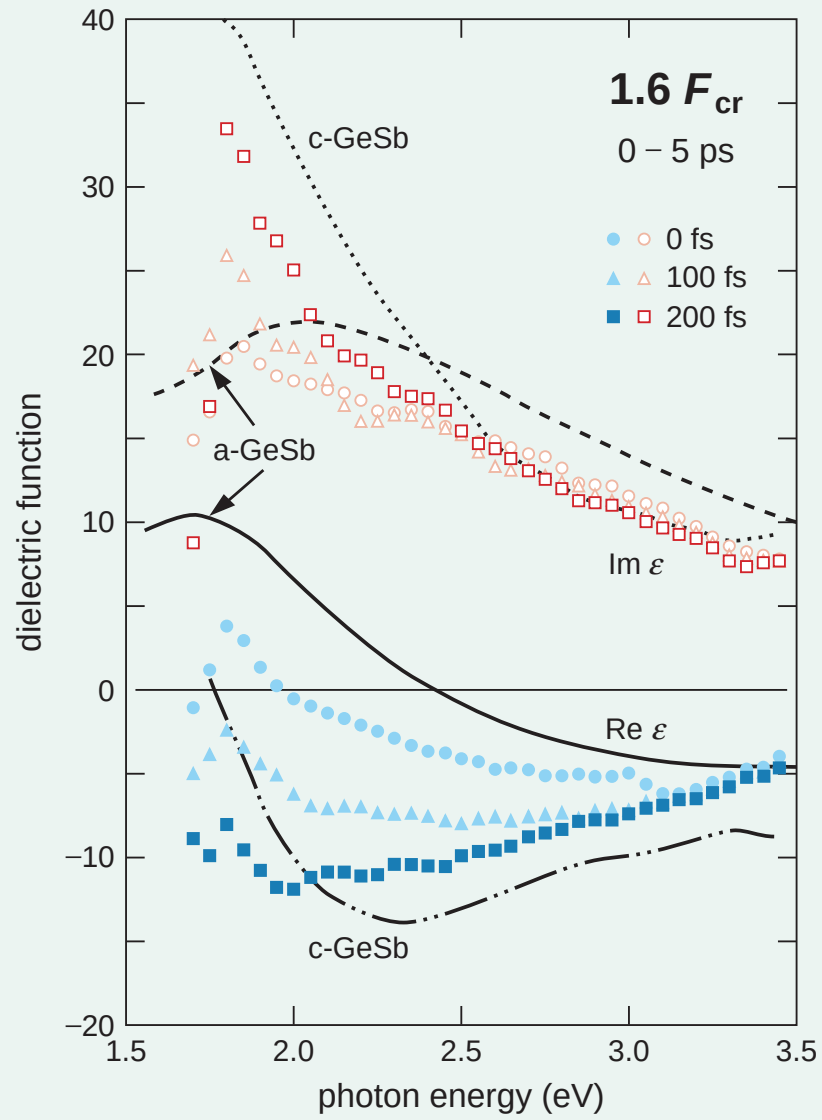

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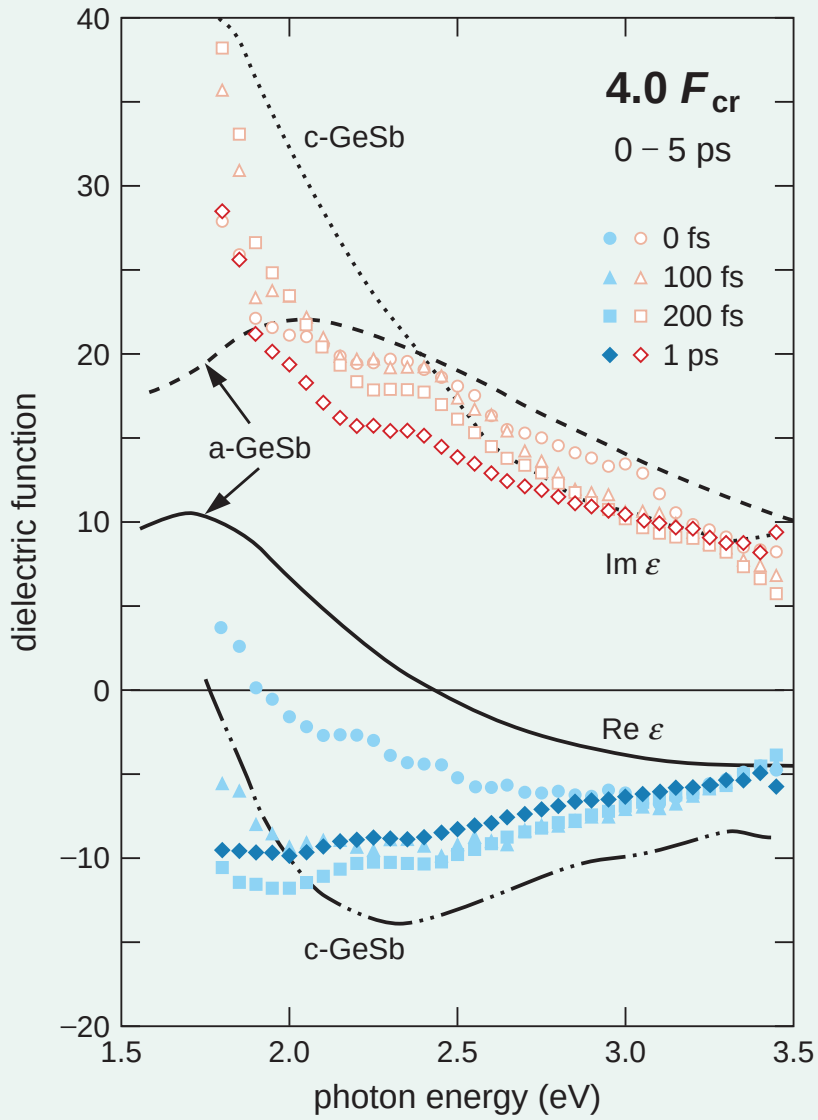
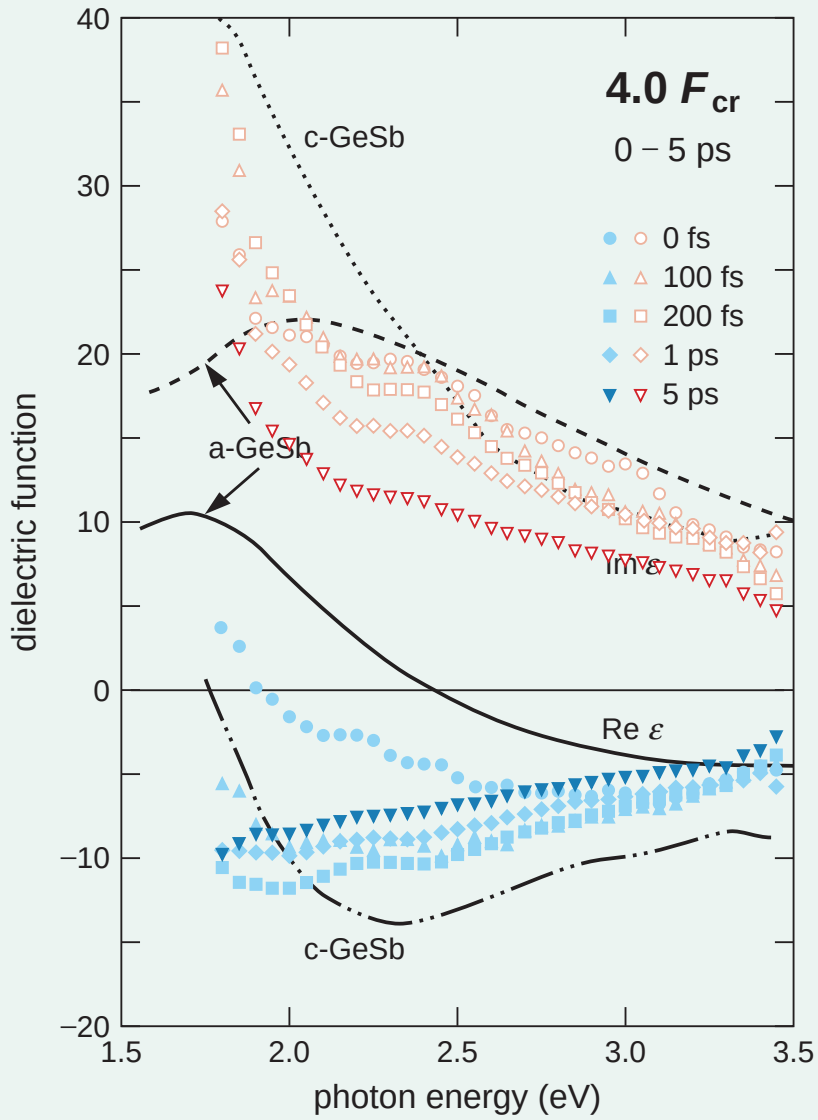


Evidence of new non-thermal phase

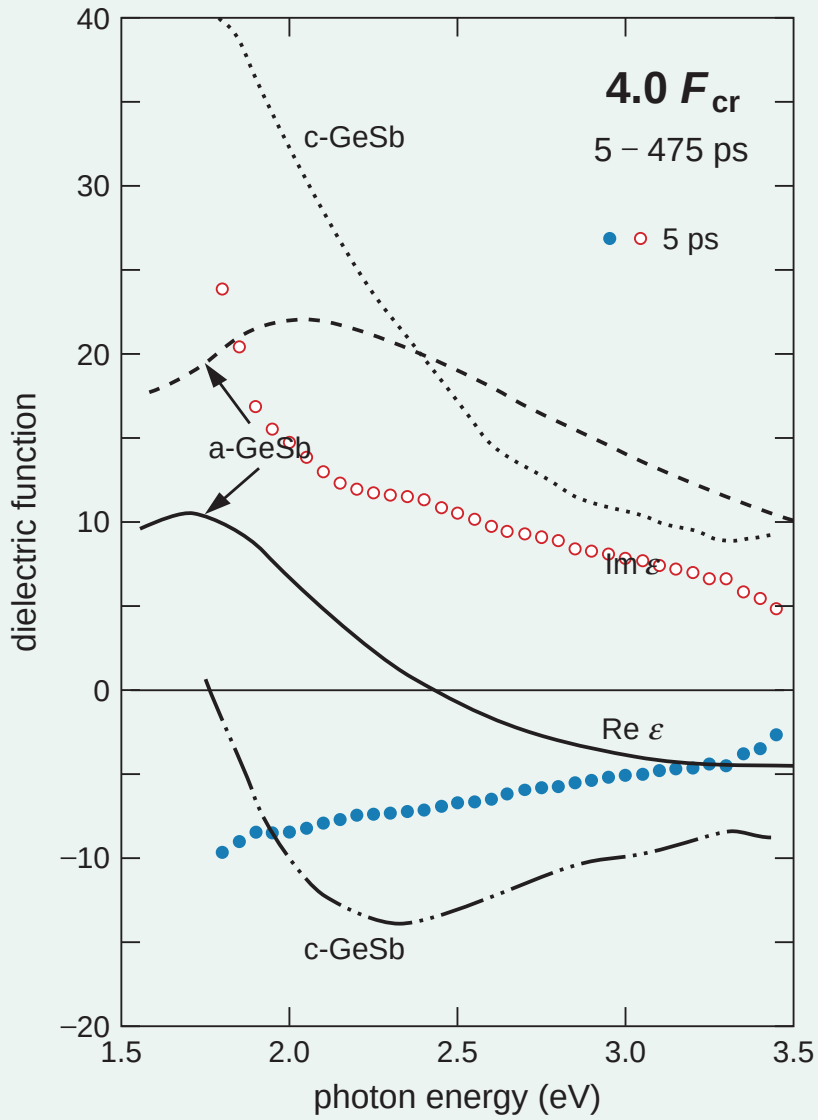
Evolution of $\varepsilon(\omega)$ after excitation of $4.0F_{cr}$



Evidence of new non-thermal phase

Subsequent dynamics due to strong excitation

Evolution of $\varepsilon(\omega)$ after excitation of $4.0F_{cr}$



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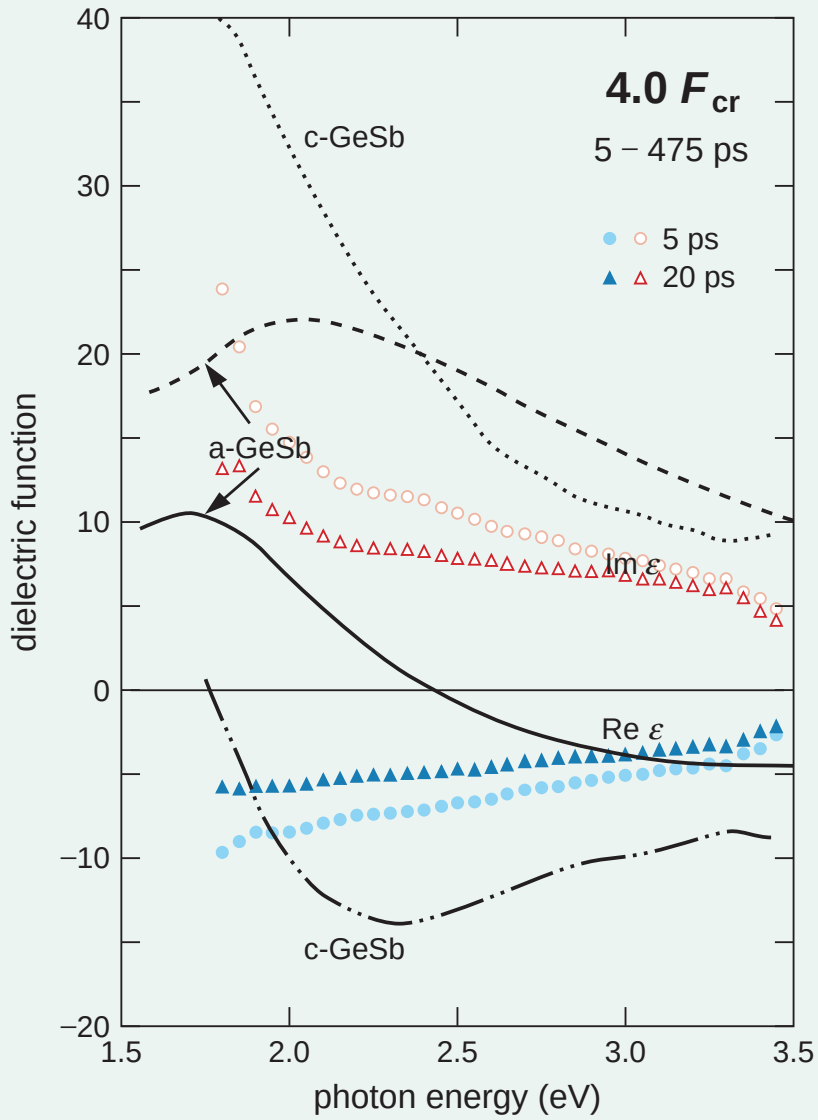

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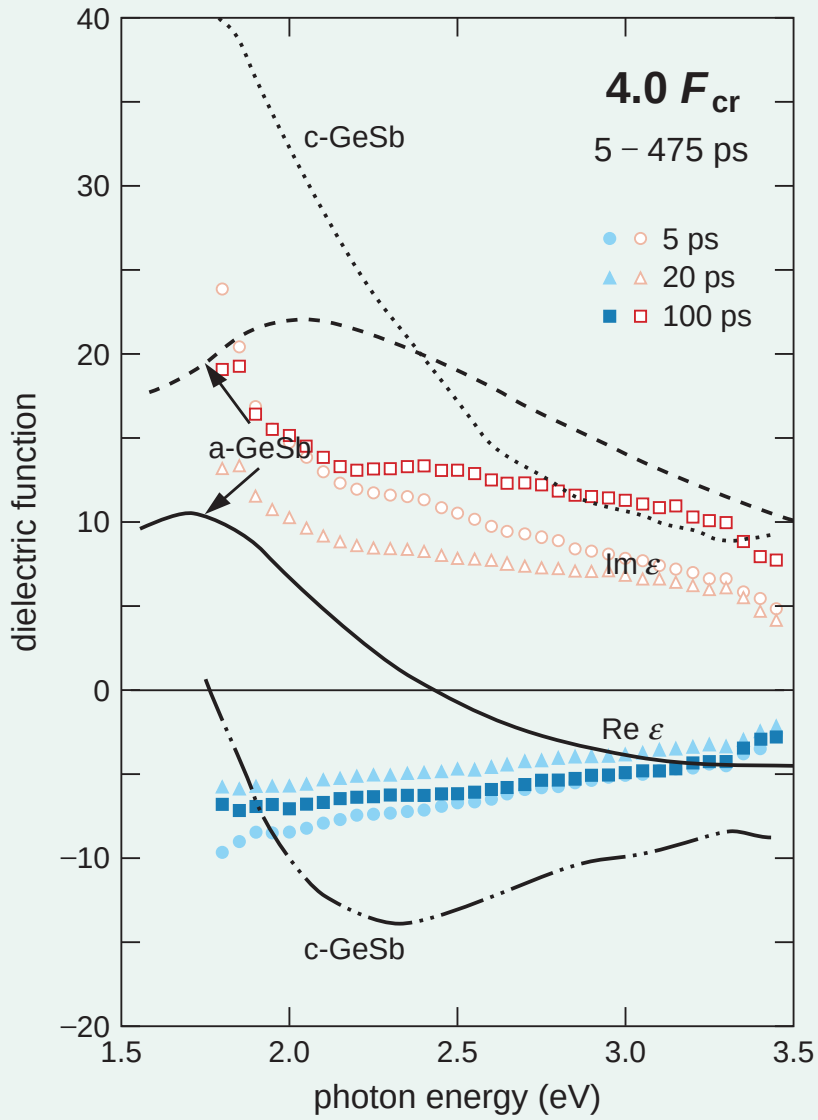

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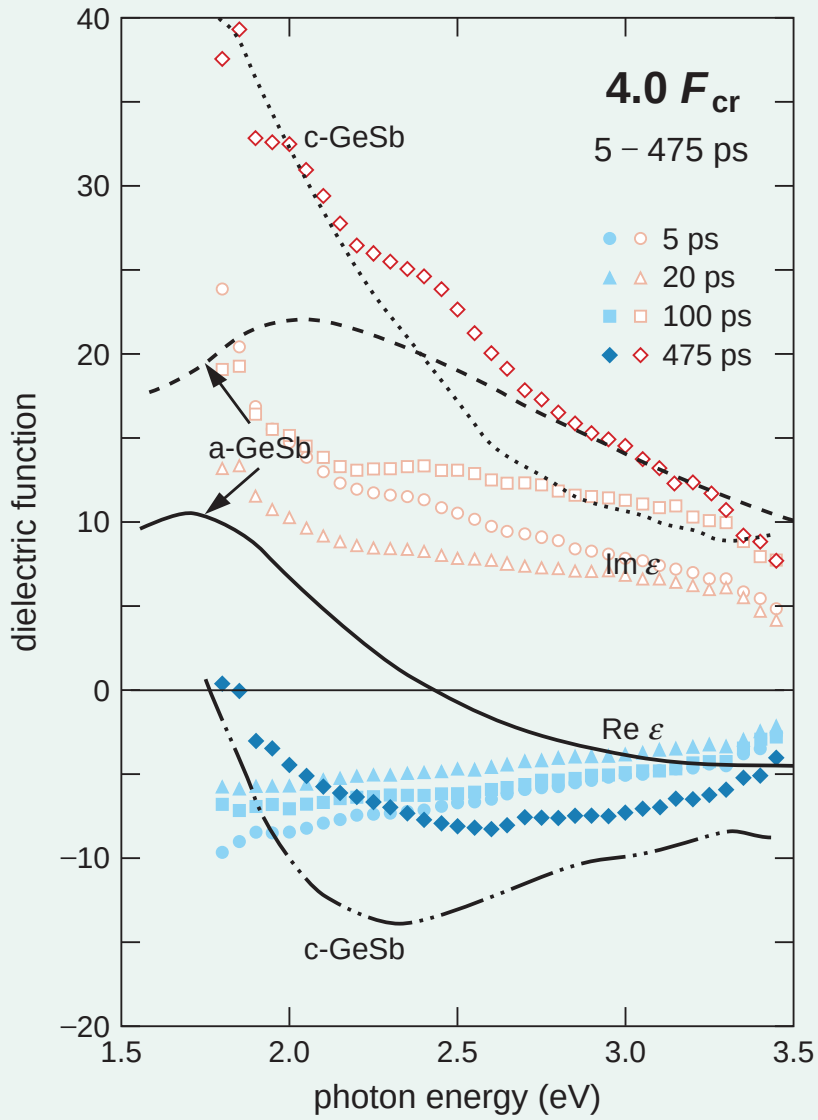
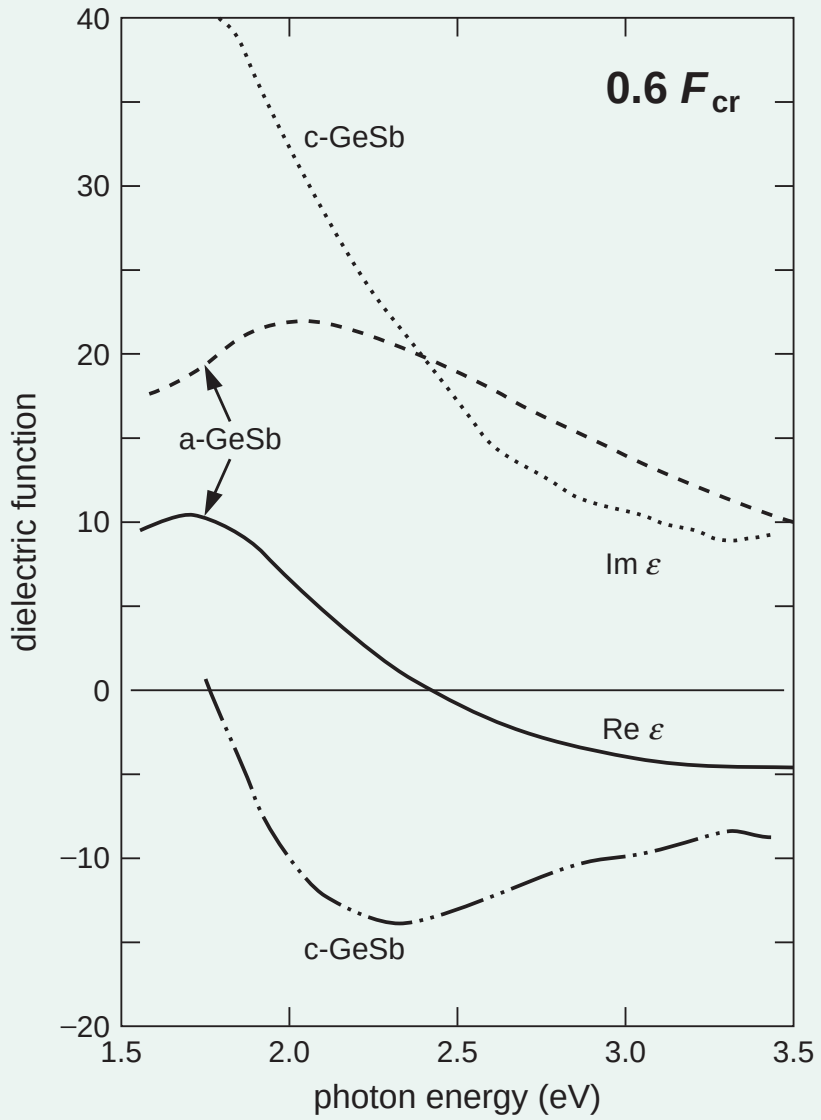


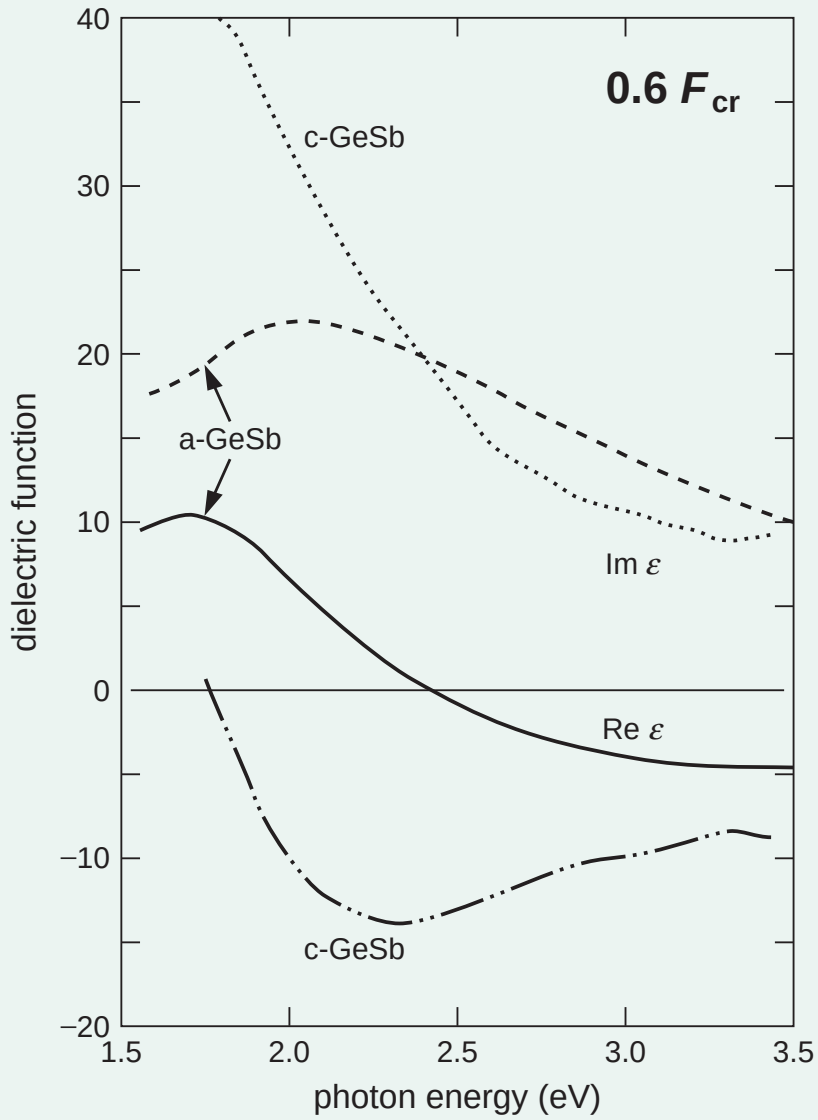
Subsequent dynamics due to strong excitation

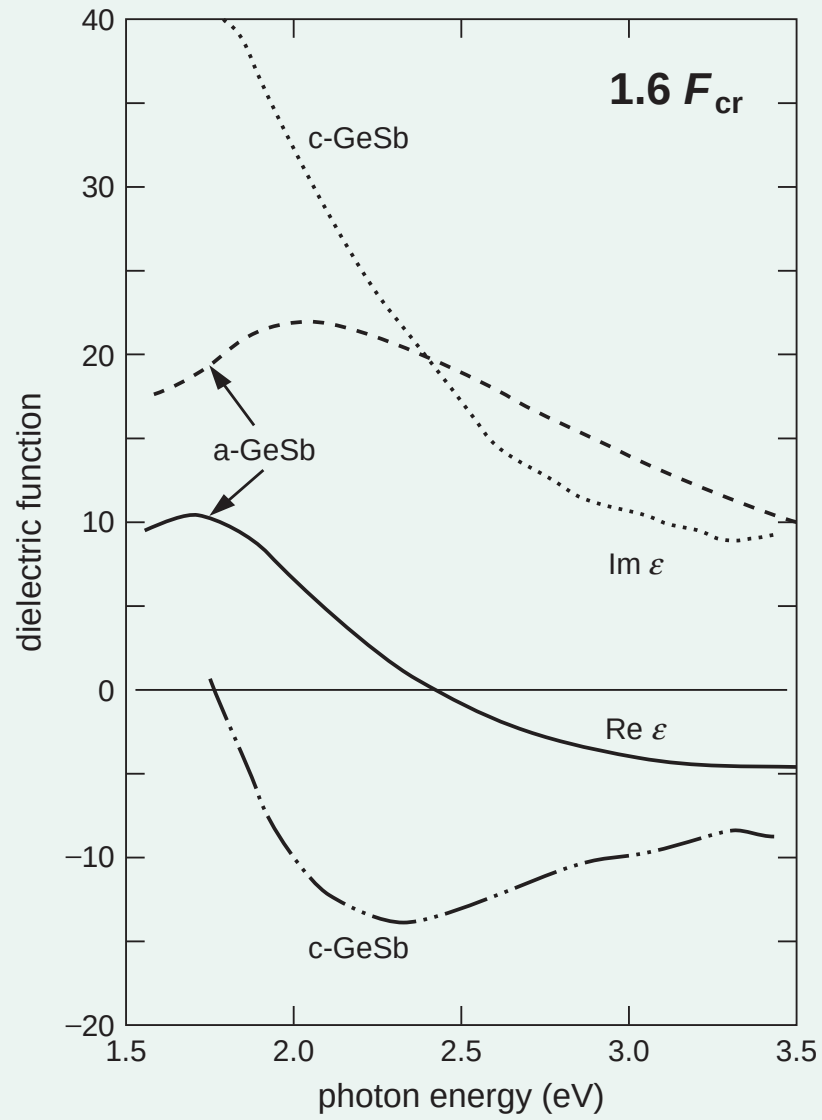
Signs of recrystallization

Evolution of $\varepsilon(\omega)$ after excitation of $0.6F_{cr}$



Evolution of ε(ω) after excitation of $0.6F_{cr}$





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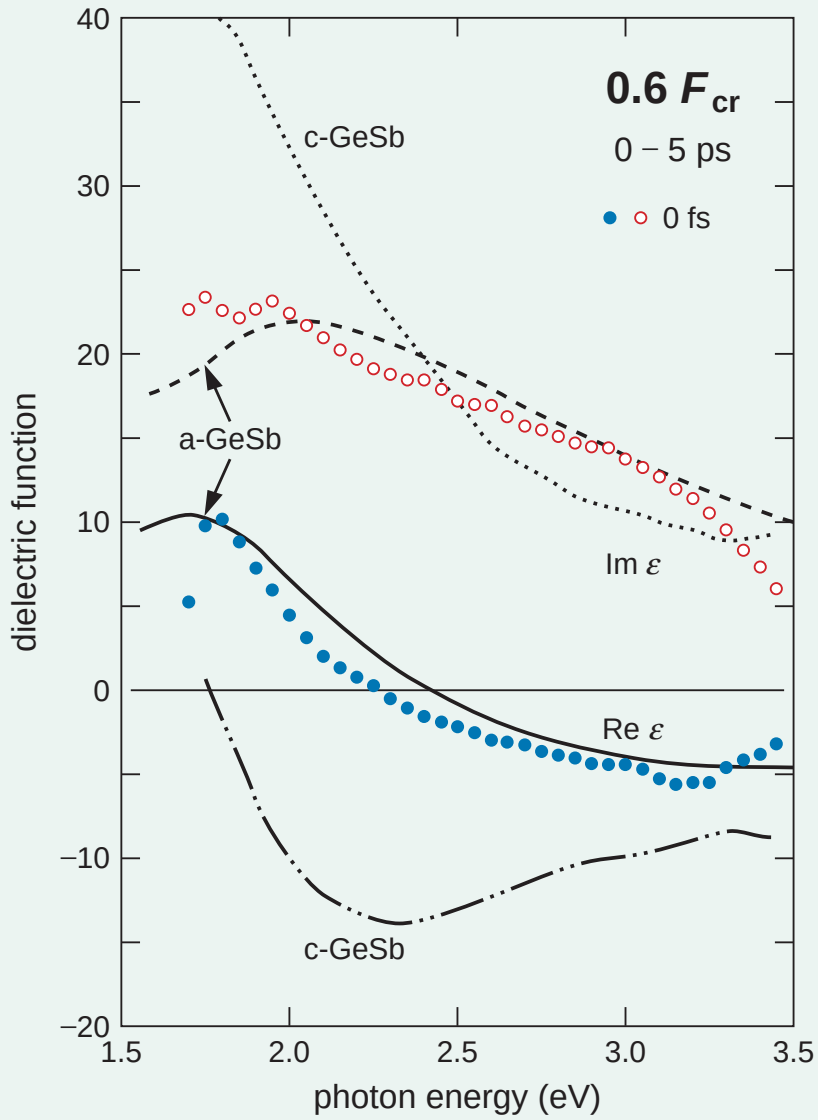

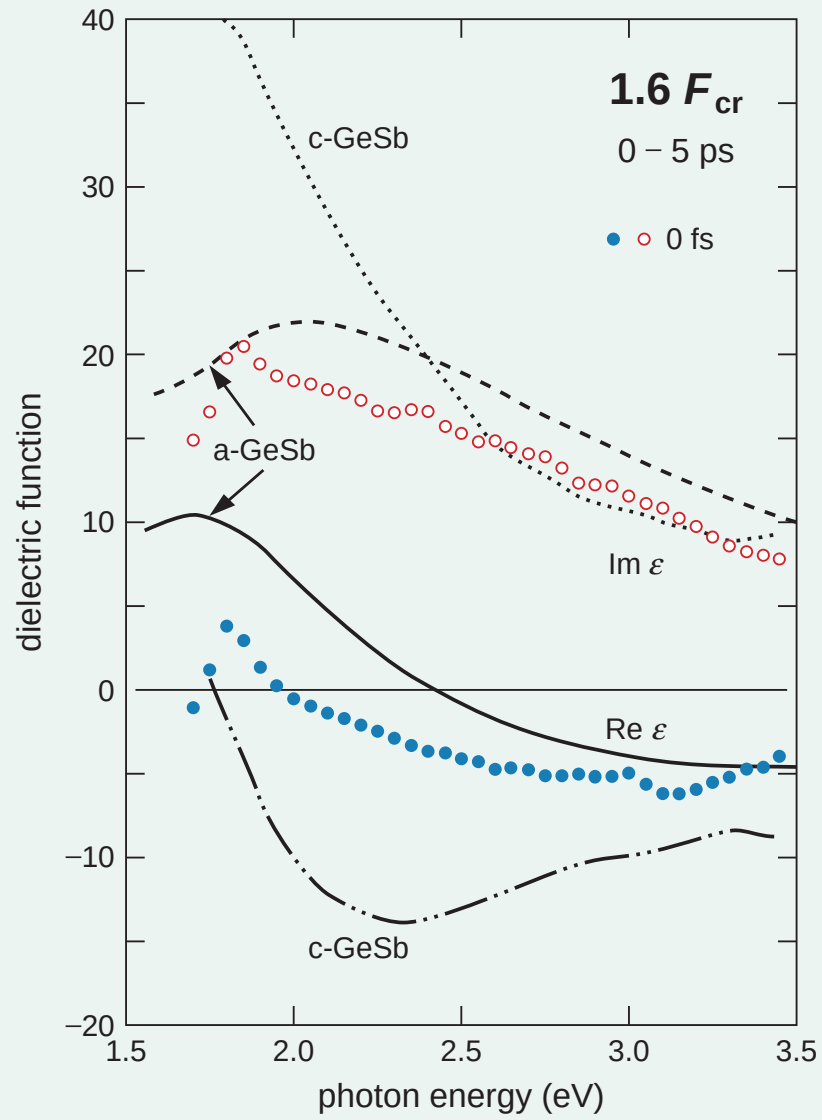

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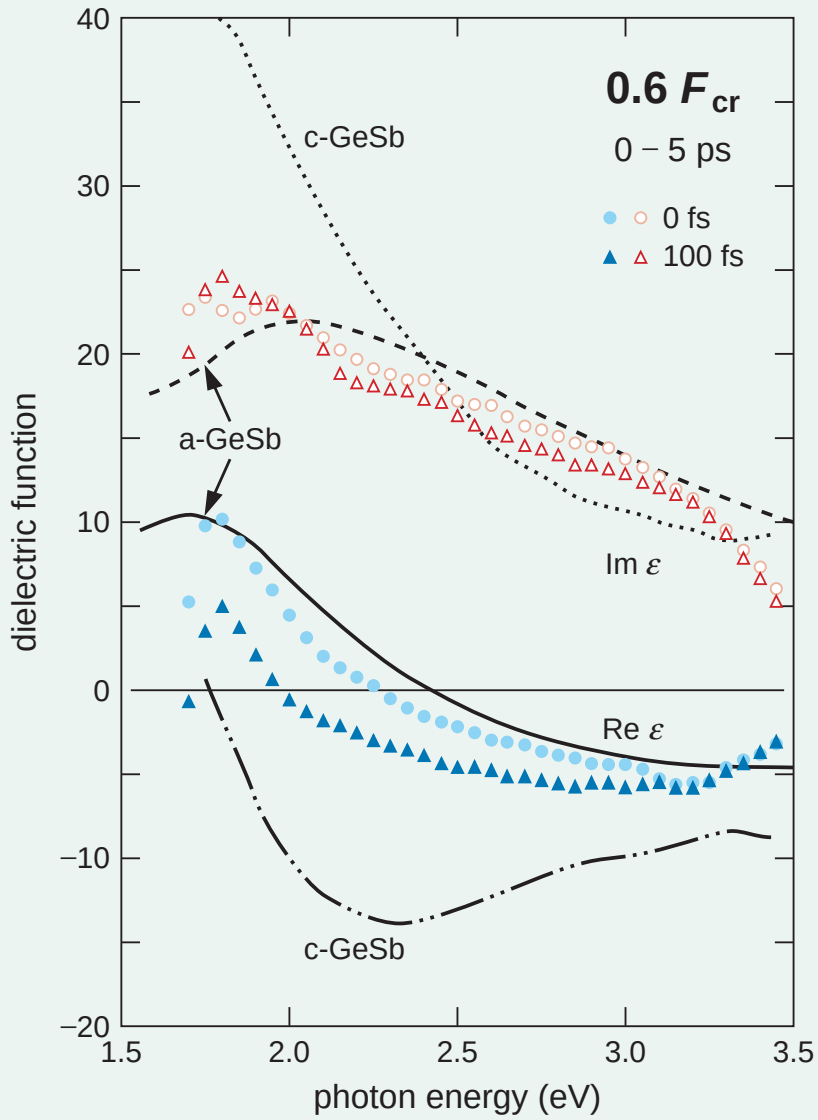

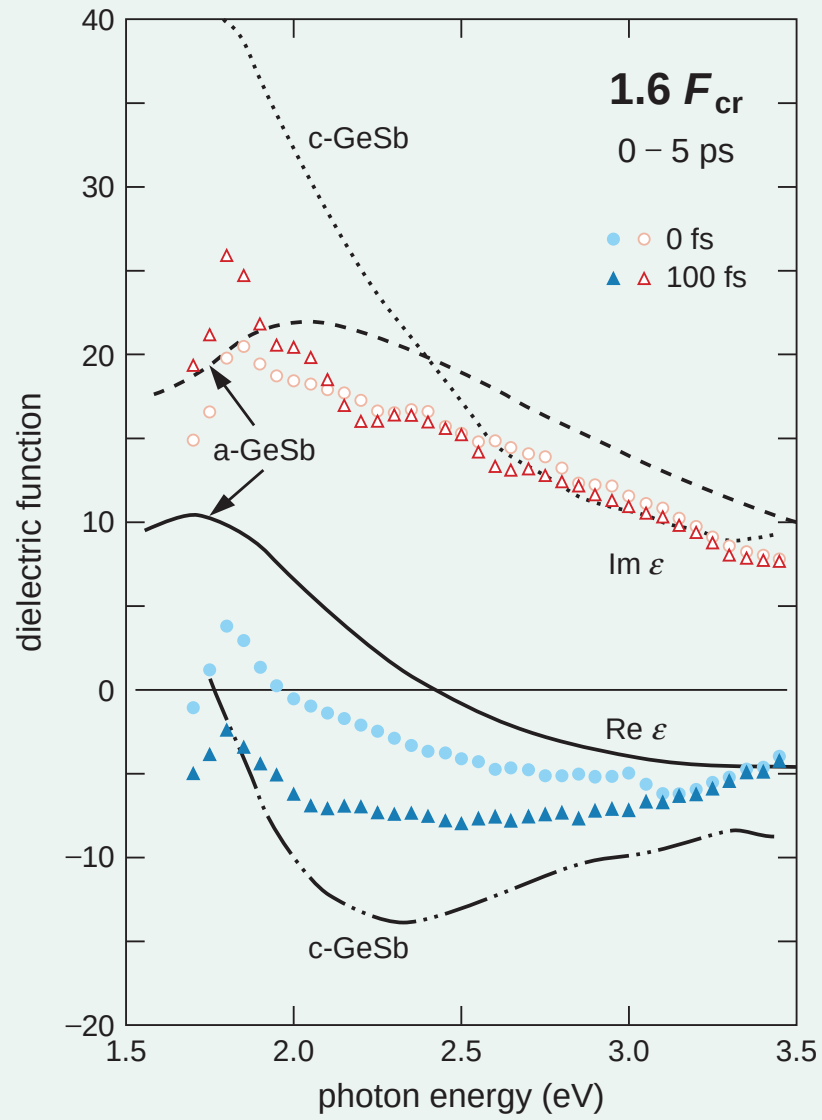

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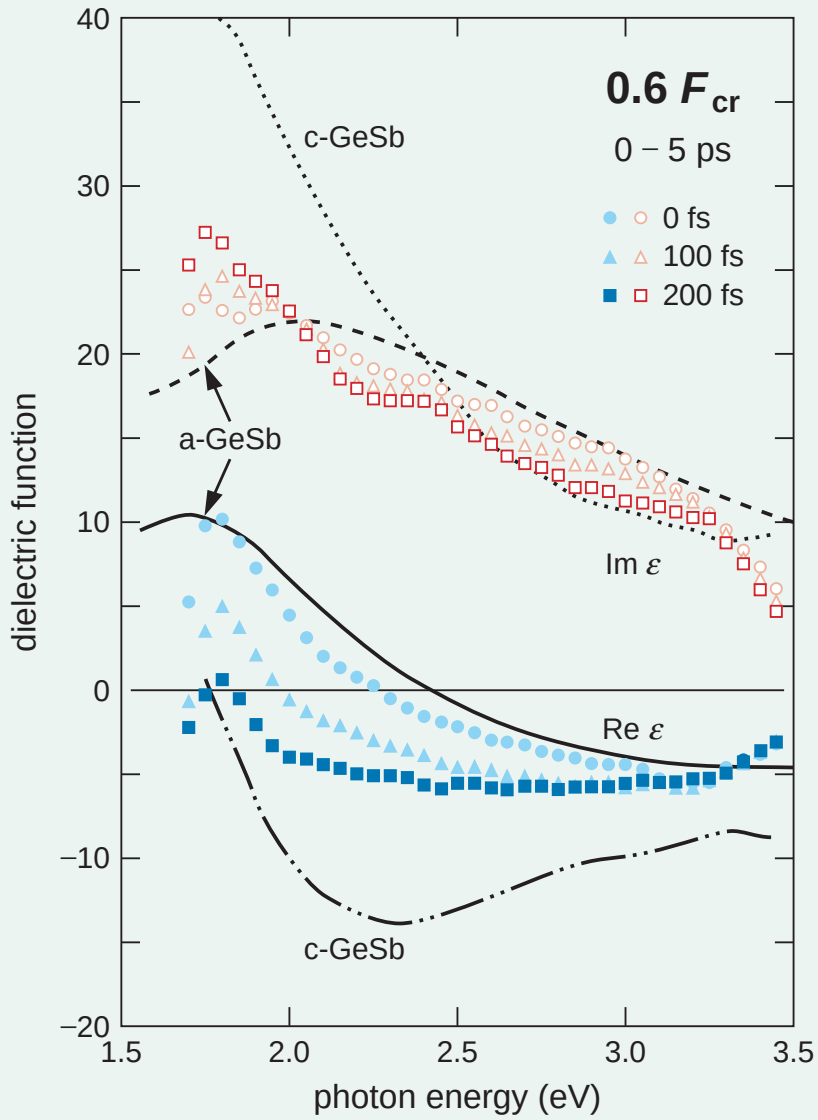

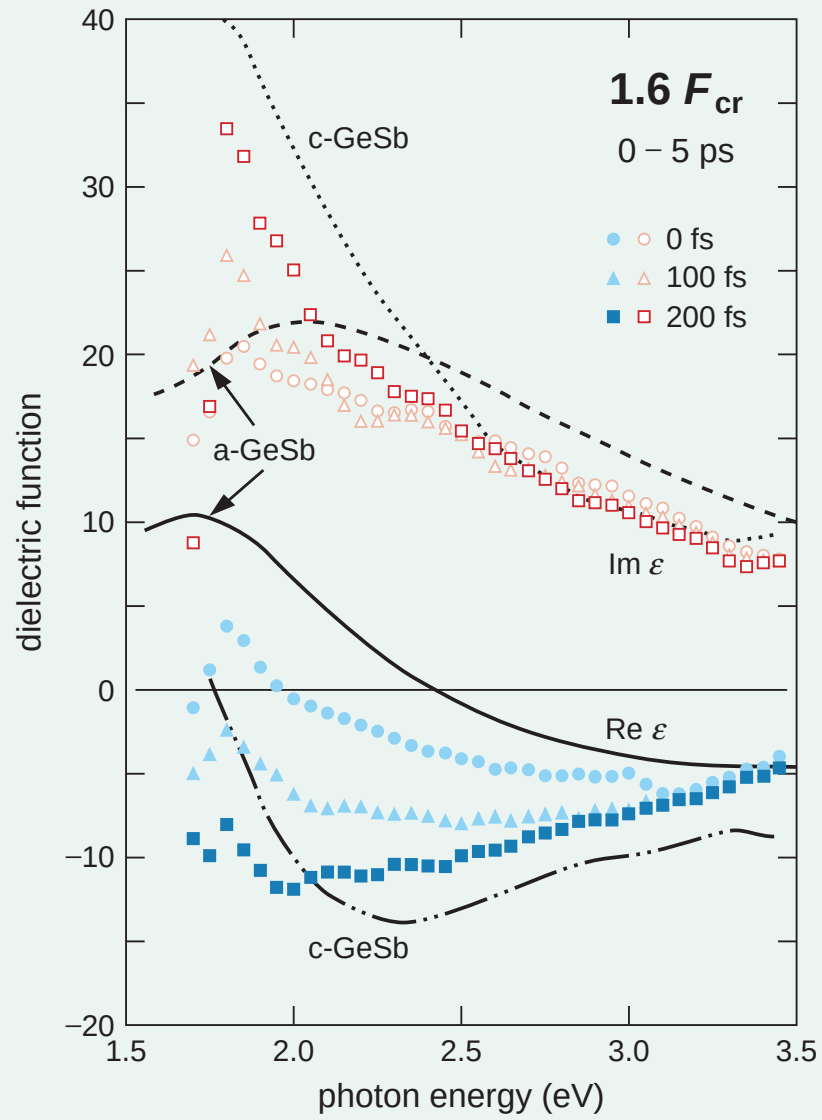

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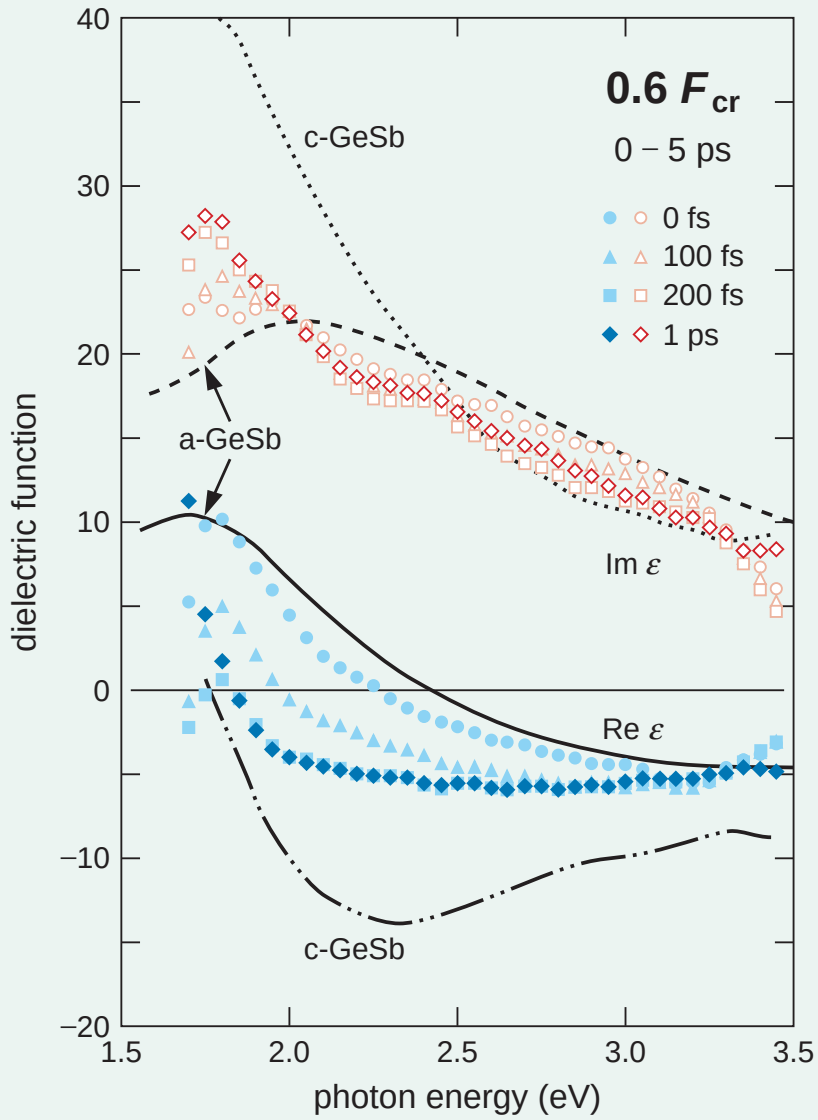
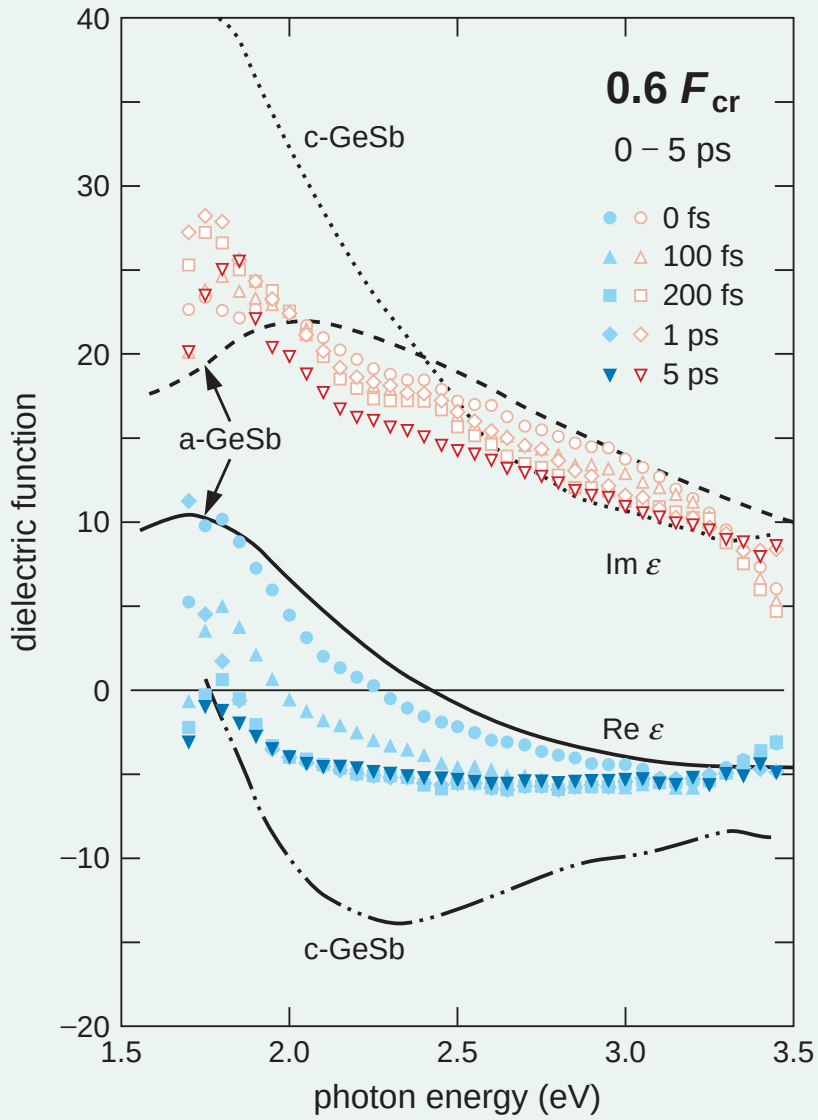


Material does not reach new phase for $F < F_{cr}$

Evolution of $\varepsilon(\omega)$ after excitation of $0.6F_{cr}$



Material does not reach new phase for $F < F_{cr}$

Evidence for transition in optically thin layer

Comparison with previous results

Comparison with previous results

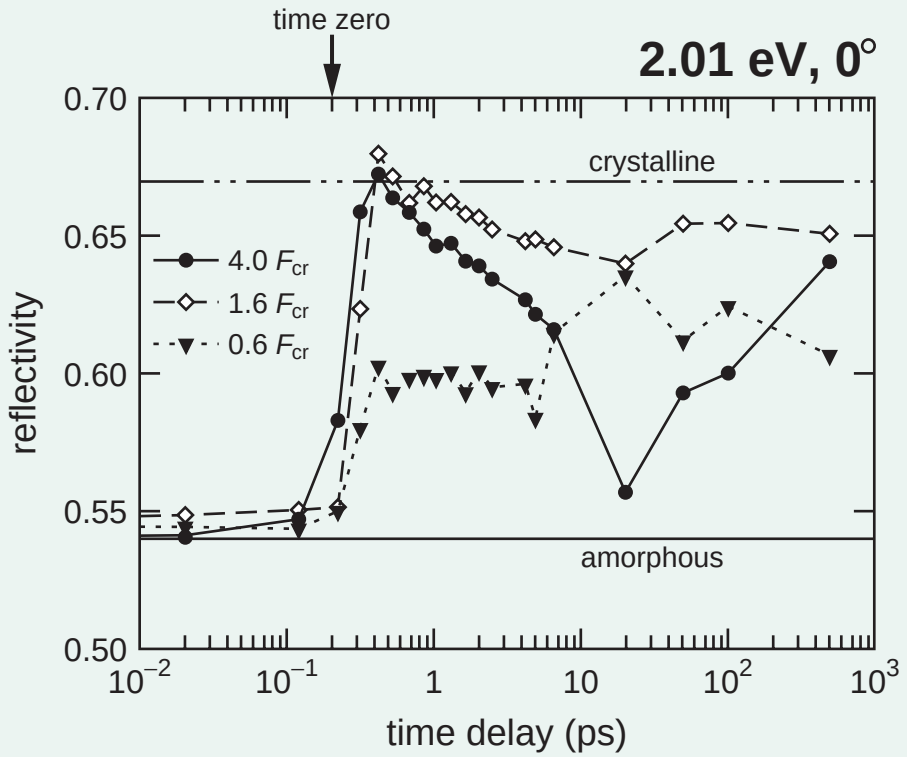
Time-resolved
$\varepsilon(\omega)$

Comparison with previous results

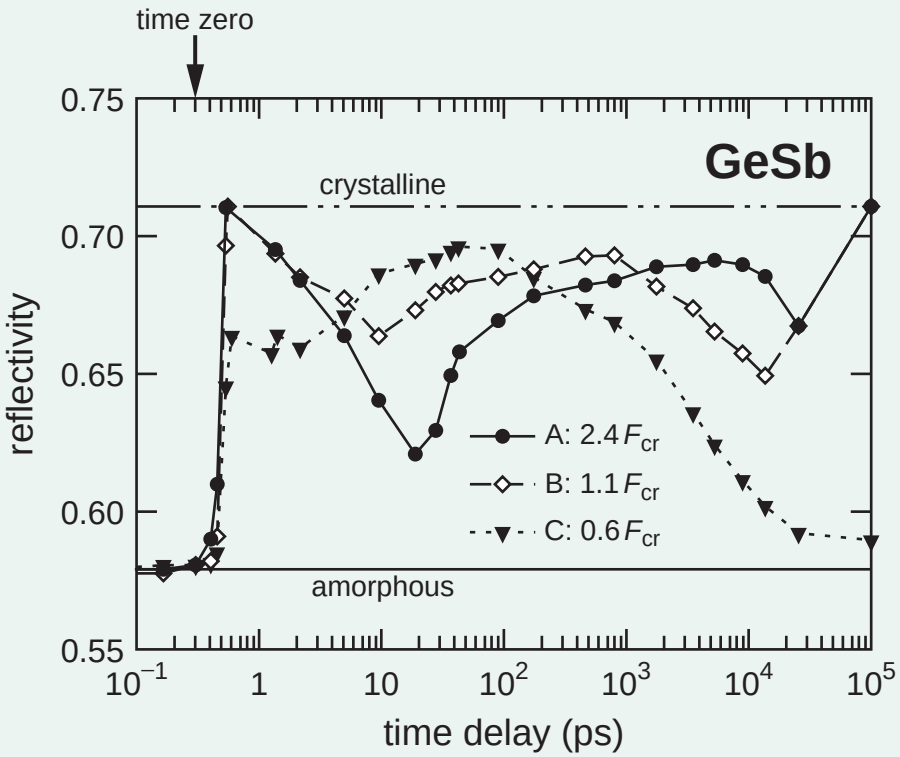
Time-resolved $\varepsilon(\omega)$ $\xrightarrow[\text{Formulae}]{\text{Fresnel}}$

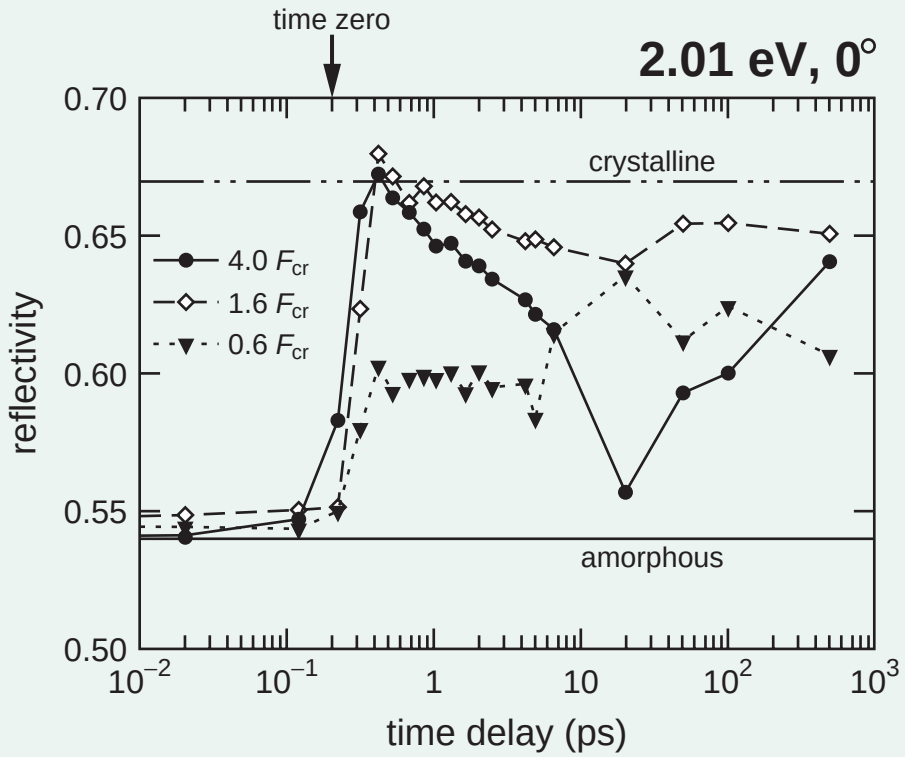
Comparison with previous results

Time-resolved $\varepsilon(\omega)$ $\xrightarrow[\text{Formulae}]{\text{Fresnel}}$



Comparison with previous results

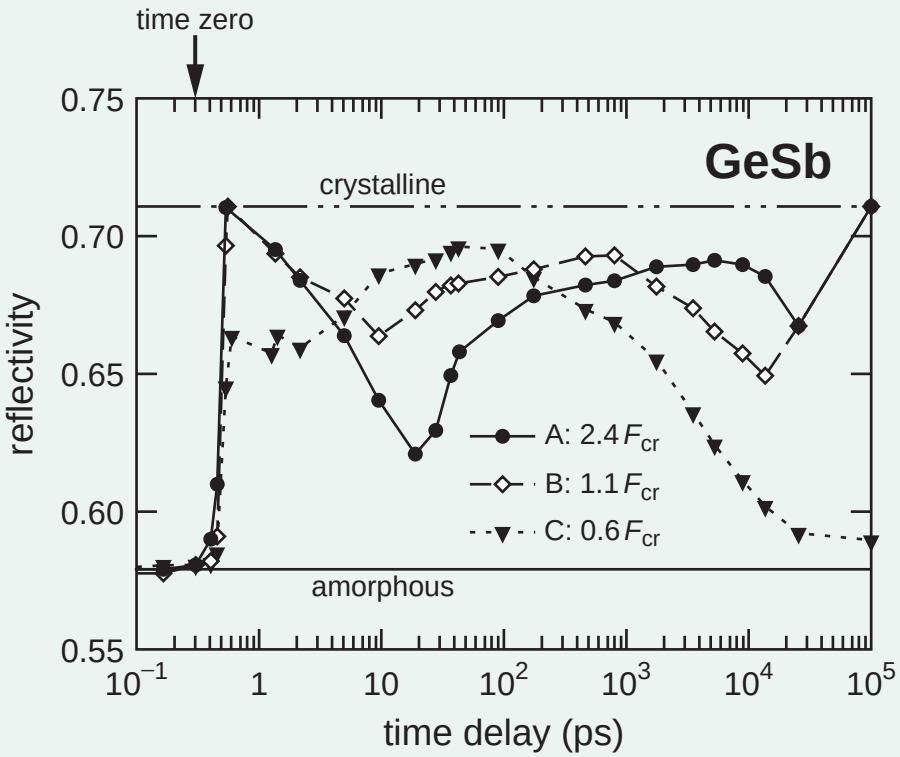


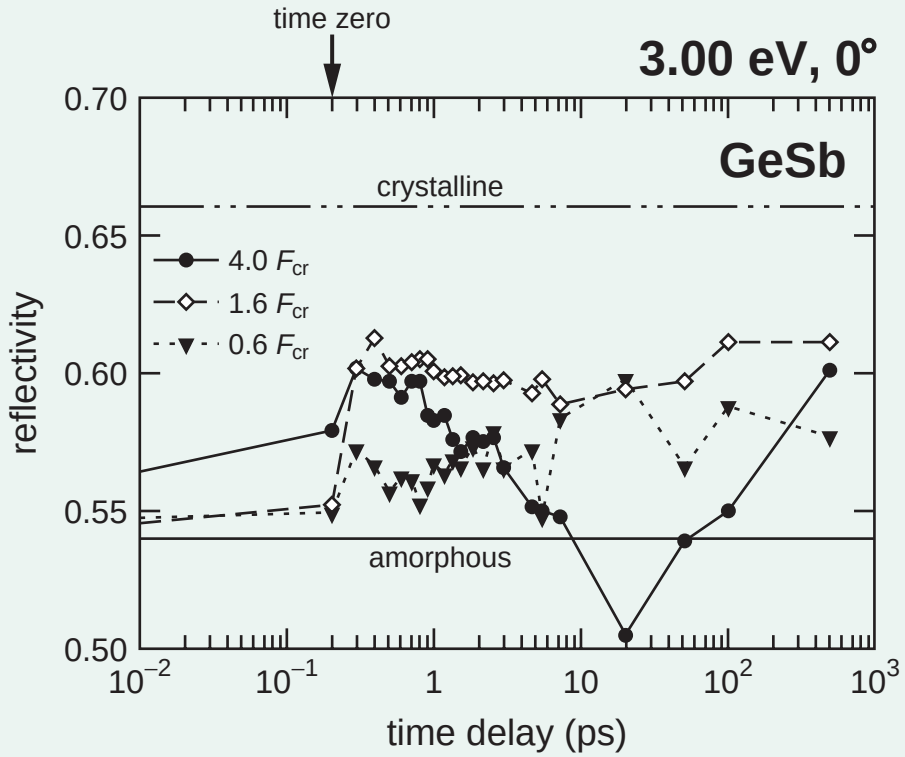


Excellent agreement at 2.01 eV and 0° angle of incidence.

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

Comparison with previous results





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

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CONCLUSION

New non-thermal phase of Sb-rich GeSb films

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

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This work can be found in

J. P. Callan *et al*., PRL, **86**, 3650 (2001)

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