Ultrafast Phase Transition Dynamics in GeSb Films

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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

\[ \text{Ge}_{0.06}\text{Sb}_{0.94} \]

amorphous phase is stabilized by Ge atoms
\[ R \approx 55\% \]
Amorphous and crystalline phases of GeSb

The amorphous phase is stabilized by Ge atoms with a stabilizing rate of $R \approx 55\%$.

The crystalline structure is identical to pure Sb, a solid solution of Ge in Sb with a rate of $R \approx 67\%$.

The critical free energy per area for crystallization is $F_{cr} = 22 \text{ mJ/cm}^2$. 

A diagram showing the atomic structures of GeSb with Ge$_{0.06}$Sb$_{0.94}$.
Amorphous and crystalline phases of GeSb

\[ F_a \sim 3F_{cr} \]

\[ Ge_{0.06}Sb_{0.94} \]

\[ F_{cr} = 22 \text{ mJ/cm}^2 \]

Amorphous phase is stabilized by Ge atoms
R \approx 55\%

Crystalline structure identical to pure Sb — solid solution of Ge in Sb
R \approx 67\%
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

**EXPERIMENTAL TECHNIQUE**

**Pump pulse:**
- 1.5 eV (800 nm)
- up to 500 µJ
- θ_{pump} < θ_1, θ_2

**Probe pulse:**
- 1.7 — 3.5 eV (350 nm — 750 nm)
- < 0.1 µJ
- θ_1 = 53°, θ_2 = 80°
Extracting the Dielectric Function

Reflectivity Spectra
Extracting the Dielectric Function

Reflectivity Spectra

Numerically invert Fresnel formulae
Extracting the Dielectric Function

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Dielectric Function

Numerically invert Fresnel formulae
Evolution of $\varepsilon(\omega)$ after excitation of $1.6F_{cr}$
Evolution of $\varepsilon(\omega)$ after excitation of $1.6F_{cr}$
RESULTS

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

![Graph showing the evolution of dielectric function after excitation.]

- $c$-GeSb
- $a$-GeSb

1.6 $F_{cr}$
- 0 – 5 ps

Dielectric function vs. photon energy (eV)

- $\text{Re} \varepsilon$
- $\text{Im} \varepsilon$

- 0 fs
- 100 fs
Evolution of $\varepsilon(\omega)$ after excitation of $1.6F_{cr}$

Material does not achieve crystalline phase...
**RESULTS**

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

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 $\varepsilon(\omega)$. 

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig.png}
\caption{Evolution of $\varepsilon(\omega)$ after excitation of $1.6F_{cr}$}
\end{figure}
Evolution of $\varepsilon(\omega)$ after excitation of $1.6 F_{cr}$

[Graph showing the evolution of the dielectric function $\varepsilon(\omega)$ for crystalline ($c$) and amorphous ($a$) GeSb after excitation with $1.6 F_{cr}$, with data points at 5 and 475 ps.]
Evolution of $\varepsilon(\omega)$ after excitation of $1.6F_{cr}$

Results

- $c$-GeSb
- $a$-GeSb

Photon energy (eV) vs. dielectric function

- Real part ($Re \, \varepsilon$)
- Imaginary part ($Im \, \varepsilon$)

5 - 475 ps
Evolution of $\varepsilon(\omega)$ after excitation of $1.6F_{cr}$

![Graph showing the evolution of $\varepsilon(\omega)$ after excitation of $1.6F_{cr}$ with different photon energies and times.](image)
Evolution of $\varepsilon(\omega)$ after excitation of $1.6F_{cr}$

Optical properties constant to $\sim 0.5$ns.
Evolution of $\varepsilon(\omega)$ after excitation of 4.0$F_{cr}$
Evolution of $\varepsilon(\omega)$ after excitation of $4.0F_{cr}$
Evolution of $\varepsilon(\omega)$ after excitation of $4.0F_{cr}$
RESULTS

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

Dielectric function

Photon energy (eV)

0 fs

100 fs
Evolution of $\varepsilon(\omega)$ after excitation of $4.0 F_{cr}$ vs $1.6 F_{cr}$ for 0 - 5 ps

Dielectric Function

- $c$-GeSb
- $a$-GeSb

Photon energy (eV)

0 fs
- 100 fs
- 200 fs
Evolution of $\varepsilon(\omega)$ after excitation of $4.0 F_{cr}$

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

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

Subsequent dynamics due to strong excitation
Evolution of $\varepsilon(\omega)$ after excitation of 4.0$F_{cr}$

- Subsequent dynamics due to strong excitation
- Signs of recrystallization
Evolution of $\varepsilon(\omega)$ after excitation of $0.6F_{cr}$
Evolution of $\varepsilon(\omega)$ after excitation of $0.6F_{\text{cr}}$
Evolution of $\varepsilon(\omega)$ after excitation of $0.6F_{cr}$
Evolution of $\varepsilon(\omega)$ after excitation of $0.6F_{cr}$

![Graph showing the evolution of dielectric function for different photon energies and times for c-GeSb and a-GeSb.](image)

- **0.6 $F_{cr}$**:
  - 0 - 5 ps
  - c-GeSb
  - a-GeSb

- **1.6 $F_{cr}$**:
  - 0 - 5 ps
  - c-GeSb
  - a-GeSb
Evolution of $\varepsilon(\omega)$ after excitation of $0.6 F_{cr}$

0.6 $F_{cr}$
- c-GeSb
- 0 – 5 ps
- 0 fs
- 100 fs
- 200 fs

1.6 $F_{cr}$
- c-GeSb
- 0 – 5 ps
- 0 fs
- 100 fs
- 200 fs
Evolution of $\varepsilon(\omega)$ after excitation of $0.6F_{cr}$

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

**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 ε(ω)
Comparison with previous results

Time-resolved $\varepsilon(\omega)$ \xrightarrow{\text{Fresnel Formulae}}
Comparison with previous results

Time-resolved $\varepsilon(\omega)$

Fresnel Formulae

Reflectivity

Time delay (ps)

2.01 eV, 0°

Crystalline

Amorphous

4.0 $F_{cr}$

1.6 $F_{cr}$

0.6 $F_{cr}$
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.

Sokolowski-Tinten et al. PRL, 81, 3670 (1998)
New non-thermal phase of Sb-rich GeSb films
CONCLUSION

New non-thermal phase of Sb-rich GeSb films

No ultrafast disorder-to-order transition in GeSb
CONCLUSION

New non-thermal phase of Sb-rich GeSb films

No ultrafast disorder-to-order transition in GeSb

Femtosecond time-resolved ellipsometry is powerful tool for probing ultrafast phase changes
Dr. K. Sokolowski-Tinten
Dr. Craig Arnold

This work can be found in
J. P. Callan et al., PRL, 86, 3650 (2001)

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