Ultrafast Lattice-Bonding Dynamics in Tellurium

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Structural transitions driven by fs pulses
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order-disorder
(a.k.a. non-thermal melting)
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order-order
Structural transitions driven by fs pulses

order-disorder
(a.k.a. non-thermal melting)

order-order

associated electronic transition
(i.e. metal-insulator)
INTRODUCTION

Cheng et al., APL 59, 1923 (1991)

$10^3 \Delta R/R$

Delay (ps)

$-2$ $-1$ $0$ $0.5$ $1.0$ $1.5$ $2.0$ $2.5$ $3.0$

$-2$ $-1$ $0$ $0.5$ $1.0$ $1.5$ $2.0$ $2.5$ $3.0$

Te

$x \times 10^{-3}$

Excitation was performed using the output of a mode-locked laser (producing ~100 MHz transform-limited pulses with a repetition rate of ~10 Hz) in a standard optical configuration. Changes induced by a pump-probe experiment (5 ps pump pulse duration, 25 ps probe pulse duration) were monitored by systematically varying the probe delay.
Te is close to band-crossing transition

P. Tangney (Princeton) and S. Fahy (Cork), *private communication*
Experimental technique
   time-resolved dielectric function

Results
   effects of coherent phonons on $\epsilon(\omega)$

Discussion
   two-atom model
   DFT calculations
EXPERIMENTAL TECHNIQUE

Time-resolved ellipsometry

White-light pump-probe setup

Ti:sapphire
800 nm, 35 fs

CaF$_2$
1.7–3.5 eV
1 µJ

0.5 mJ

spectral filter

spectrometer

delay
Time-resolved ellipsometry
EXPERIMENTAL TECHNIQUE

Time-resolved ellipsometry

pump
35 fs, 800 nm

50°

sample
Time-resolved ellipsometry

probes 350 μm - 750 μm

sample
Time-resolved ellipsometry

probes
350 - 750 μm

R₁
R₂

Fresnel equations
EXPERIMENTAL TECHNIQUE

Time-resolved ellipsometry

- Probes: 350 - 750 μm
- Fresnel equations

\[
\begin{align*}
\text{Re}[\psi(w)] & \\
\text{Im}[\psi(w)] &
\end{align*}
\]
RESULTS

-500 fs

dielectric function

energy (eV)

Im[

Re[
RESULTS

Dielectric function

-500 fs

energy (eV)

dielectric function

Im[\[\text{ord}\]]

Re[\[\text{ext}\]]

Re[\[\text{ord}\]]

Im[\[\text{ext}\]]

Re[\[\text{ord}\]]

Re[\[\text{ext}\]]
RESULTS

-500 fs

Energy (eV)

Dielectric function

Re[$\varepsilon_{\text{ext}}$]

Im[$\varepsilon_{\text{ord}}$]

Im[$\varepsilon_{\text{ext}}$]
RESULTS

-500 fs
"Two-atom" model
"Two-atom" model

Bonding-antibonding splitting
"Two-atom" model

Lorentz oscillator model
"Two-atom" model

photon promotes an electron...
"Two-atom" model

... weakening the bond...
“Two-atom” model

... establishing new equilibrium positions
"Two-atom" model

ions move to new equilibrium positions...
"Two-atom" model

... decreasing the splitting...
"Two-atom" model

... and redshifting the dielectric function
"Two-atom" model

ions overshoot equilibrium positions...
"Two-atom" model

... reversing travel and overshooting again
"Two-atom" model

Oscillation around displaced equilibrium position
Tellurium lattice

helical radius $x = 0.26d$
Tellurium lattice

$A_1$ mode modulates $x$
Band structure is sensitive to $x$

**Diagram:**

- **Energy (eV):**
  - $0.3$ eV

- **K-space points:**
  - A
  - H
  - K

- **Labels:**
  - Te

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P. Tangney (Princeton) and S. Fahy (Cork), *private communication*
Bands cross when x changes by 6%
DISCUSSION

Track zero of real part

Dielectric function

Energy (eV)

-500 fs

-20

-10

0

10

20

30

40

50

60

Energy (eV)
Track zero of real part

![Graph showing dielectric function vs. energy (eV) at 220 fs.](image)
Track zero of real part

![Graph showing the redshift of zero of Re(\text{E}) as a function of time delay (ps). The graph displays oscillations with a peak at 0.0 and a label indicating 120 J/m².](image-url)
Higher fluence: larger amplitude phonons
Frequency less than 3.6 THz

![Graph showing redshift of zero of Re[e] vs. time delay (ps) for different fluences (90 J/m² and 120 J/m²) at 3.3 THz.](image)
Phonon mode softens

![Graph showing redshift of zero of Re][1](time delay (ps))
Compare shift to band gap

DISCUSSION

0.3 eV

3.5 THz

3.3 THz

90 J/m²

120 J/m²

time delay (ps)

redshift of zero of \Re[\hat{I}]

-0.1

0.0

0.1

0.2

0.3

0.4

0.5

-0.5

0

0.5

1.0

1.5

2.0
Compare shift to band gap

- 0.3 eV
- 3.5 THz
- 3.3 THz
- 90 J/m²
- 120 J/m²
- 220 fs
$\varepsilon(\omega)$ is not metallic

![Graph](image-url)
Semiconducting because of 0.3-eV gap
After bands cross...
... material can become metallic...
... provided phonons scatter electrons
If $\square_{\text{scatter}} > T_{\text{phonon}}$, 'frustrated' metal
Coherent phonons modulate dielectric function
Coherent phonons modulate dielectric function

Evidence for transient band-crossing
Coherent phonons modulate dielectric function

Evidence for transient band-crossing

... but no metal
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For a copy of this talk and additional information, see:

http://mazur-www.harvard.edu