Nonequilibrium Lattice Dynamics in photoexcited bismuth

David A. Reis
Stanford PULSE Institute
Departments of Applied Physics and Photon Science

dreis@stanford.edu

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Goal: understanding and controlling matter near and far from equilibrium

Requires new measurements, with combined time (energy) and momentum (spatial) resolution
Lattice instabilities of Group V and IV-VI and related materials

S. Lee et al., Nat. Comm. 2014

Photo-excitation affects balance of e-ph

H Kawamura - Narrow Gap Semiconductors Physics and Applications, 1980

Murray et al. PRB 75 2007.

“Displacive Excitation of Coherent Phonons”

$A_{1g}$-mode optical generation/detection


Displacive excitation

Modifications from P. HUEY/SCIENCE

Oscillations about new equilibrium due to reduction in Peierls distortion
Femtosecond diffraction ca. 2007

Femtosecond diffraction ca. 2007


DFPT calculation

Murray et al. PRB 75 2007.
• Introduction to time-resolved (non-resonant) x-ray scattering (TRXS, FT-IXS)

• Measurement of Anharmonic decay constants (3rd order forces)/channels in photo-excited bismuth

• Measurement of Nonequilibrium Dispersion/Interatomic forces in photo-excited bismuth.
Energy-resolved, time-integrated x-ray scattering (IXS)

\[ I_s(Q, \omega) \propto S(Q, \omega) = \frac{1}{2\pi} \int S(Q, \Delta t)e^{-i\omega \Delta t} d\Delta t \]

\[ \propto \sum_j \left\langle (Q \cdot u_{jq})^2 \right\rangle \delta(\omega \pm \omega_{jq}) \]

van Hove, Squires, …

Time-resolved, energy-integrated x-ray scattering (TRXS, FT-IXS)

\[ I_s(Q, t) \propto S(Q; t) \propto \sum_{jj'} \left\langle Q \cdot u_{jq}(t)u_{qj'}^*(t) \cdot Q \right\rangle \]

\[ q = G - Q \]

Trigo & DAR
Fourier-Transform IXS

\[ I_s(Q, t) \sim \sum_{jj'} \langle Q \cdot u_{jq}(t) u_{j'q}^*(t) \cdot Q \rangle \]

Trigo et al. Nature Physics. 9, 790, 2013
The Linear Coherent Light Source (LCLS) at SLAC
The XPP instrument at LCLS

Photo credit: Daniele Fanelli
Example, GaAs ~10fs pump/probe

2M pixel/120 Hz readout

Area detector

Zhu, Henighan, Trigo... unpublished
Example, GaAs ~10fs pump/probe

Zhu, Henighan, Trigo... unpublished
• Non-equilibrium measurement
• Efficient use of photons, broad band, no analyzer ($\tau_p < 1/\omega$)
• Resolution limited by maximum delay

Zhu, Henighan, Trigo... unpublished
Mechanism?

\[ H' \propto I(t)u \]

1st order Raman, coherent state

\[ \langle u \rangle \propto \cos\Omega t \]
\[ \langle u^2 \rangle \propto \cos2\Omega t \]

Henighan et al., 94, 020302(R) (2016)

\[ H' \propto I(t)u^2 \]

2nd order Raman, squeezed state

\[ \langle u \rangle = 0 \]
\[ \langle u^2 \rangle \propto \cos2\Omega t \]
Mechanism?

\[ H' \propto I(t)u \]

1st order Raman  coherent state

\[ \nu = (2n + 1) \frac{1}{2\Delta t} \]

\[ H' \propto I(t)u^2 \]

2nd order Raman  squeezed state

\[ \nu = (2n + 1) \frac{1}{4\Delta t} \]

Henighan et al., 94, 020302(R) (2016)
For perfect crystals, squeezing...
Stabilization of paraelectric phase in PbTe with photo-excitation

Transverse two-phonon Spectra along bonding direction ($\Gamma-X$)

overtone ($2\omega_{TA}$) & combination ($\omega_{TO} \pm \omega_{TA}$)

Consistent with reduction of long-range interactions and Peierls-like electron-lattice instability

TR-diffuse scattering from photo-ex. Bi
FT-IXS spectra for Bi

Teitlebaum, Trigo, Henighan et al.
PRL 121, 125901 (2018).
Predictions for $A_{1g}$ decay in Bi

Anharmonic decay of coherent phonon (parametric resonance)

\[ H = \frac{1}{2} (P_0^2 + \Omega^2 u_0^2) + \frac{1}{2N} \sum_q (P_q^2 + \omega_q^2 (1 + 2g_q u_0) u_q^2) \]

Single phonon

Continuum of anharmonic phonons

Coherent phonon:
\[ \langle u_0 \rangle = A(1 - \cos(\Omega t)e^{-i\gamma_0 t}) \rightarrow \tilde{\omega}_q(u_0) = \tilde{\omega}_q(t) \]

Squeezed phonons:
\[ \langle u_q^2 \rangle \sim g_q A \omega_q / \gamma_q \left( e^{-\gamma_0 t/2} - e^{-\gamma_q t} \right) \sin \Omega t \]

(on resonance)

Image of downcoverted phonons

- Measured FT-IXS @ $\omega_{A1g}$
- Prediction with uniform coupling

Teitlebaum, Trigo, Henighan et al.
PRL 121, 125901 (2018).
Measurement of $q$-dependent coupling constants

$g_q = -1.0$

(DFPT -8.4)

Teitlebaum, Trigo, Henighan et al.
PRL 121, 125901 (2018).
Fluence dependence of excited-state dispersion bismuth

Preliminary, S.Teitelbaum et al. 2019
Extraction of excited-state forces: determine frequencies from FT-IXS

Dominant frequency

TDS intensity

Time (ps)
...Fit modified dispersion to IF-model
Excited-state interatomic forces of Bi

A: 1nn

B: 2nn

G: 9nn

Theory (Ha/bohr)

<table>
<thead>
<tr>
<th>Pair</th>
<th>NN</th>
<th>$n=0%$</th>
<th>$n=1%$</th>
<th>Distance</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>First</td>
<td>-33.23</td>
<td>-27.74</td>
<td>5.766</td>
</tr>
<tr>
<td>B</td>
<td>Second</td>
<td>-0.88</td>
<td>-0.52</td>
<td>6.604</td>
</tr>
<tr>
<td>C</td>
<td>Third</td>
<td>-0.67</td>
<td>-0.86</td>
<td>8.544</td>
</tr>
<tr>
<td>D</td>
<td>Fourth</td>
<td>-0.30</td>
<td>-0.50</td>
<td>8.873</td>
</tr>
<tr>
<td>E</td>
<td>Sixth</td>
<td>-0.26</td>
<td>-0.16</td>
<td>10.360</td>
</tr>
<tr>
<td>F</td>
<td>Eighth</td>
<td>-0.43</td>
<td>-0.26</td>
<td>11.766</td>
</tr>
<tr>
<td>G</td>
<td>Ninth</td>
<td>-4.16</td>
<td>-4.40</td>
<td>12.318</td>
</tr>
</tbody>
</table>
Comparison with DFT calculations

Experimental (Reconstruction)

DPFT Calculations*

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LCLS: D. Zhu, M. Chollet, T. Sato

S. Fahy, S. O’Mahony, I. Savic (Cork), E. Murray (Imperial), Olivier Delaire (Duke) C. Uher, T.P. Bailey (Michigan)