# Nonequilibrium Lattice Dynamics in photoexcited bismuth

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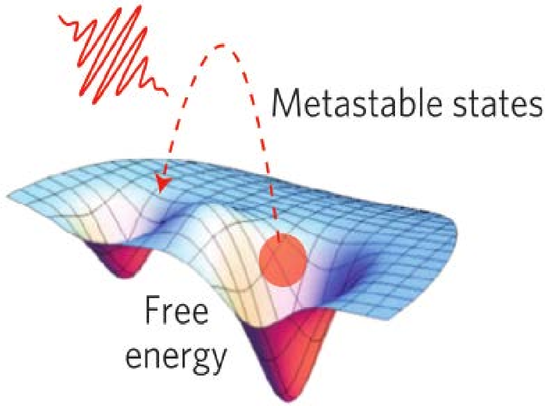
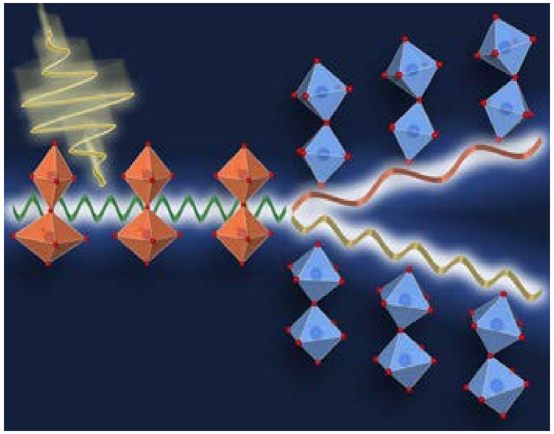
IXS2019, June 23–28, 2019, Stony Brook University

# Goal: understanding and controlling matter near and far from equilibrium




Basov (2018)



Andrea Cavalleri, MPSD

Requires new measurements,
with combined time (energy) and momentum (spatial) resolution

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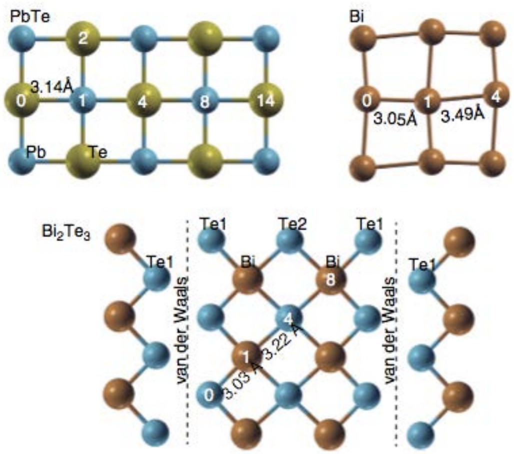
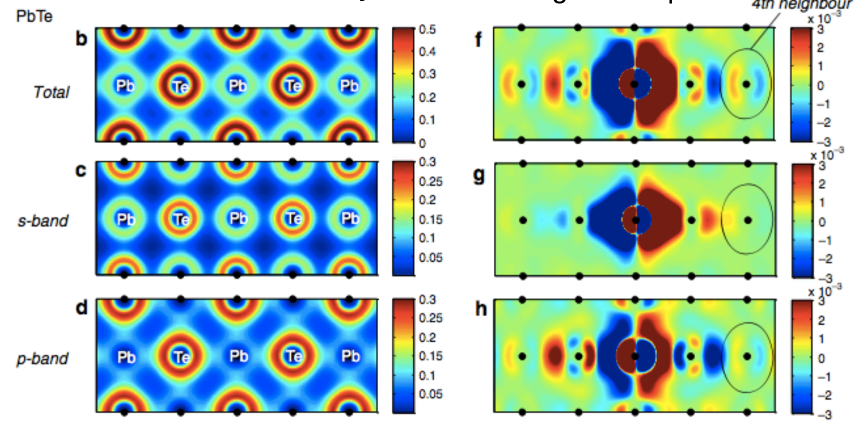
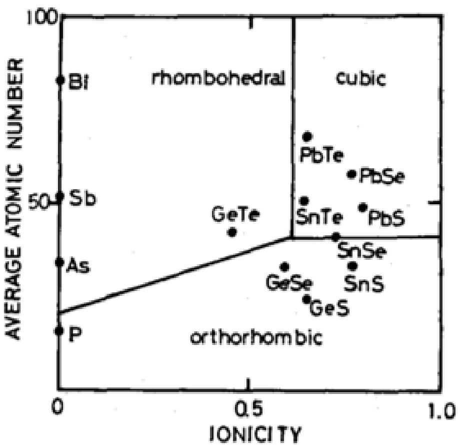
# Lattice instabilities
# of Group V and IV-VI and related materials




Ground state density
Change w/ displacement



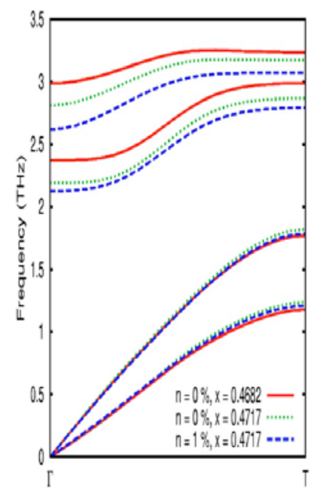

S. Lee et al., Nat. Comm. 2014




H Kawamura - Narrow Gap Semiconductors Physics and Applications, 1980

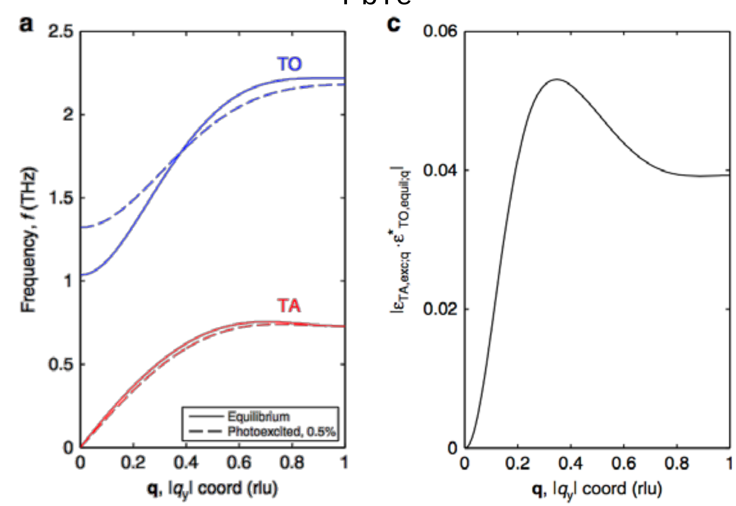
## Photo-excitation affects balance of e-ph

Bi




Murray *et al.* PRB 75 2007.

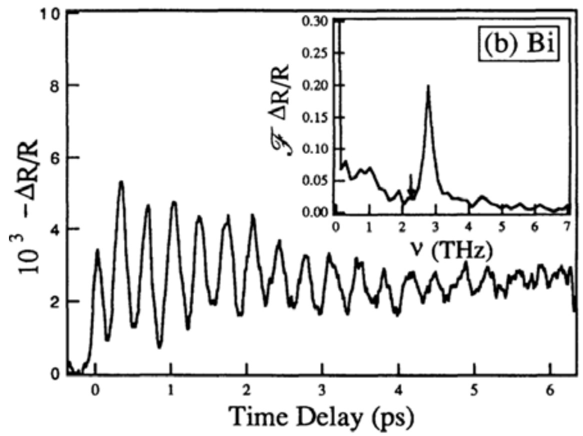
PbTe



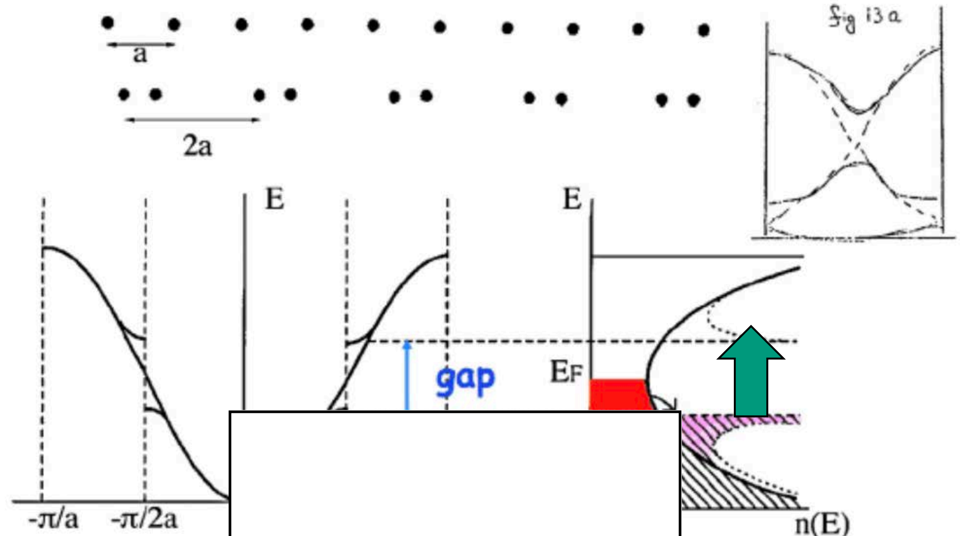

Jiang *et al.* Nat. Comm. 2016.

# “Displacive Excitation of Coherent Phonons”

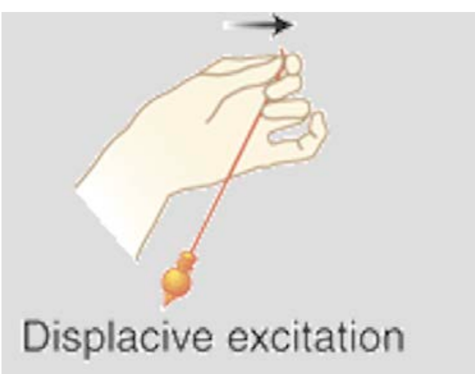
$A_{1g}$–mode optical generation/detection




Zeiger et al., PRB 1992.



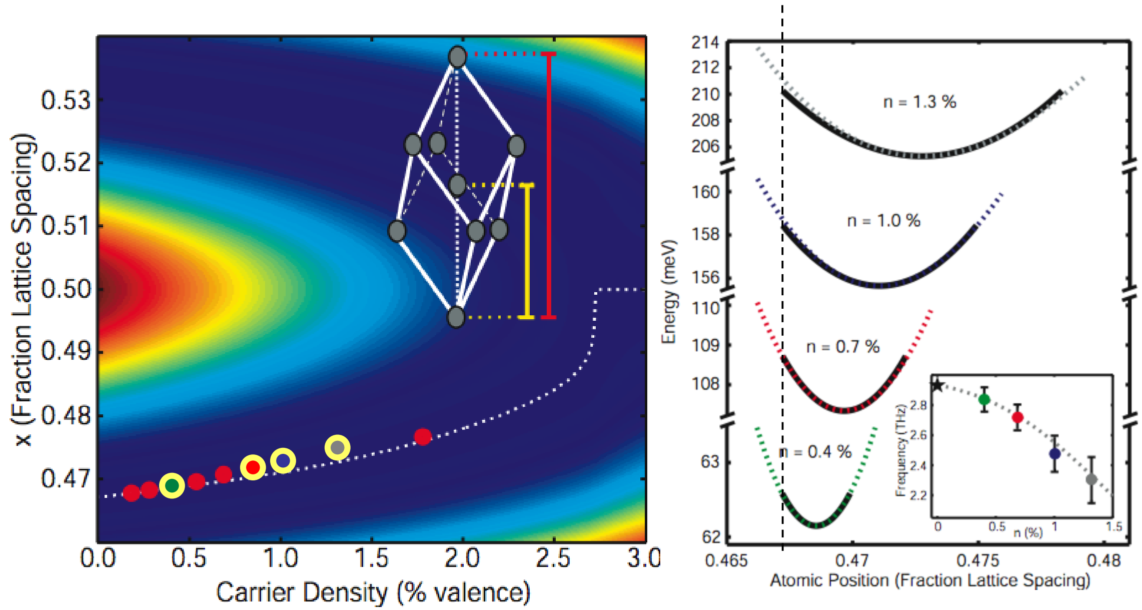

Mod. F (2016)



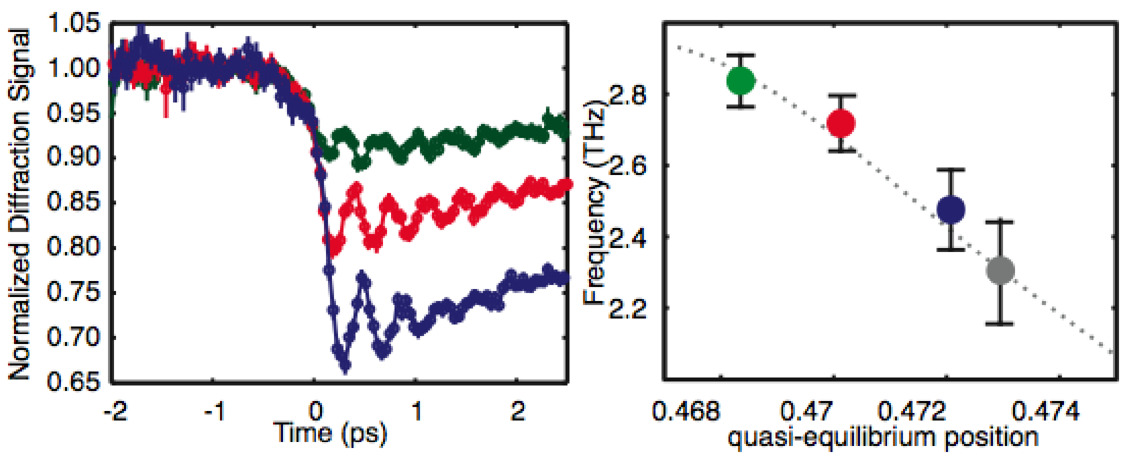

Modified from P. HUEY/*SCIENCE*

Oscillations about new equilibrium eierls distortion

# Femtosecond diffraction ca. 2007

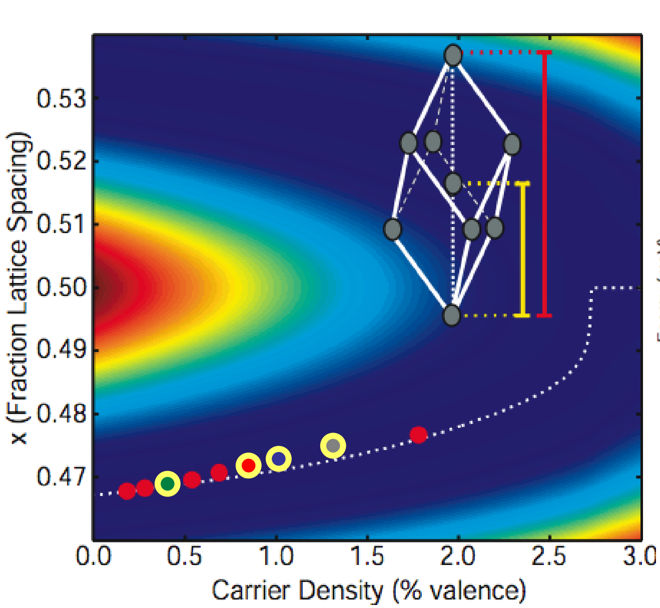


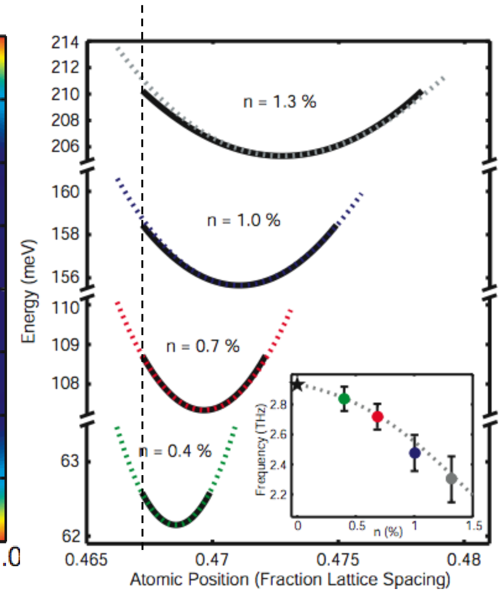

D. M. Fritz et al. Science 315, 2007.




(SPPS)

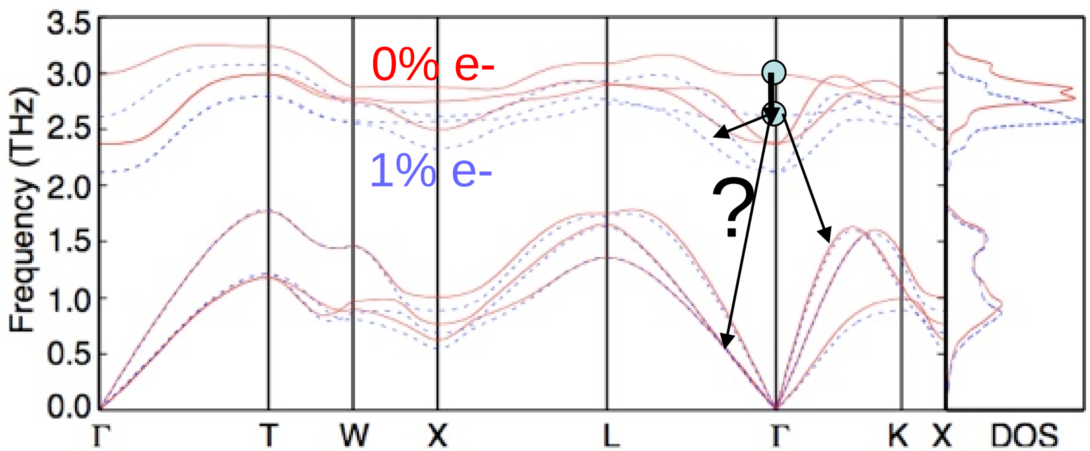
# Femtosecond diffraction ca. 2007




D. M. Fritz et al. Science 315, 2007.

DFPT calculation




Murray *et al.* PRB 75 2007.

# Outline

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- Introduction to time-resolved (non-resonant) x-ray scattering (TRXS, FT-IXS)
- Measurement of Anharmonic decay constants (3$^{rd}$ 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)

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

$$\propto \sum_j \left\langle \left(\boldsymbol{Q}\cdot\boldsymbol{u}_{\boldsymbol{jq}}\right)^2 \right\rangle \boldsymbol{\delta}(\boldsymbol{\omega} \pm \boldsymbol{\omega}_{\boldsymbol{jq}})$$

van Hove, Squires, …

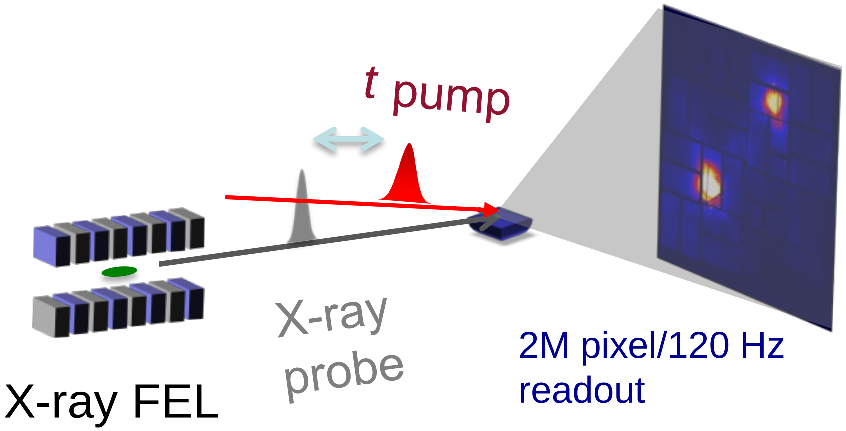
# Time-resolved, energy-integrated x-ray scattering (TRXS, FT-IXS)

$$I_s(\boldsymbol{Q},t) \propto S(\boldsymbol{Q};t) \qquad \propto \sum_{jj'} \left\langle \boldsymbol{Q}\cdot\boldsymbol{u}_{jq}(t)\boldsymbol{u}^*_{\bar{\boldsymbol{q}}\,j'}(t)\cdot\boldsymbol{Q}\right\rangle$$

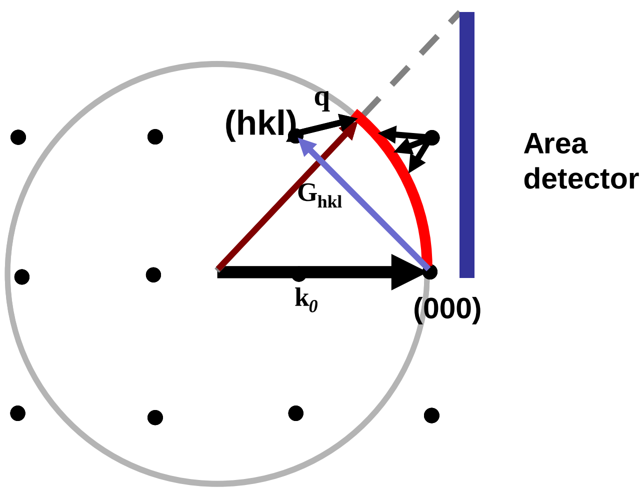
$$\boldsymbol{q} = \boldsymbol{G} - \boldsymbol{Q}$$

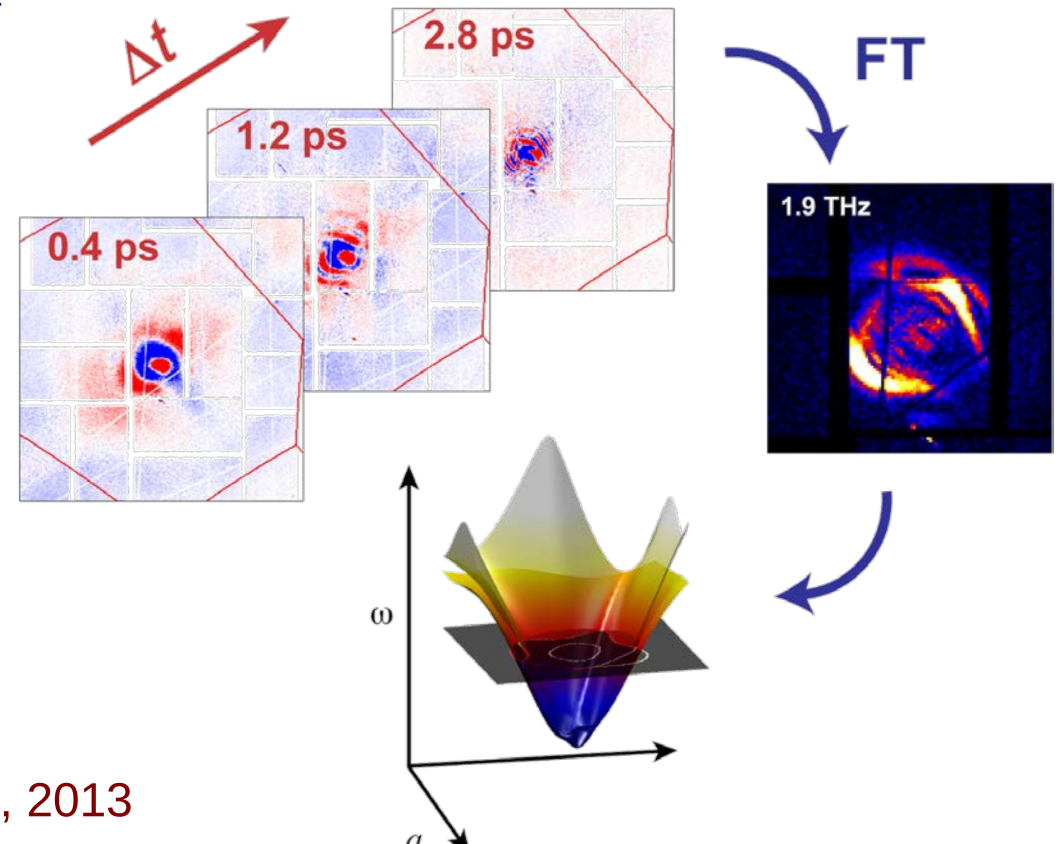
Trigo & DAR

# Fourier-Transform IXS




$$I_s(\boldsymbol{Q},t) \sim \sum_{jj'} \left\langle \boldsymbol{Q}\cdot \boldsymbol{u}_{jq}(t)\,\boldsymbol{u}^*_{j'\bar{q}}(t)\cdot \boldsymbol{Q}\right\rangle$$

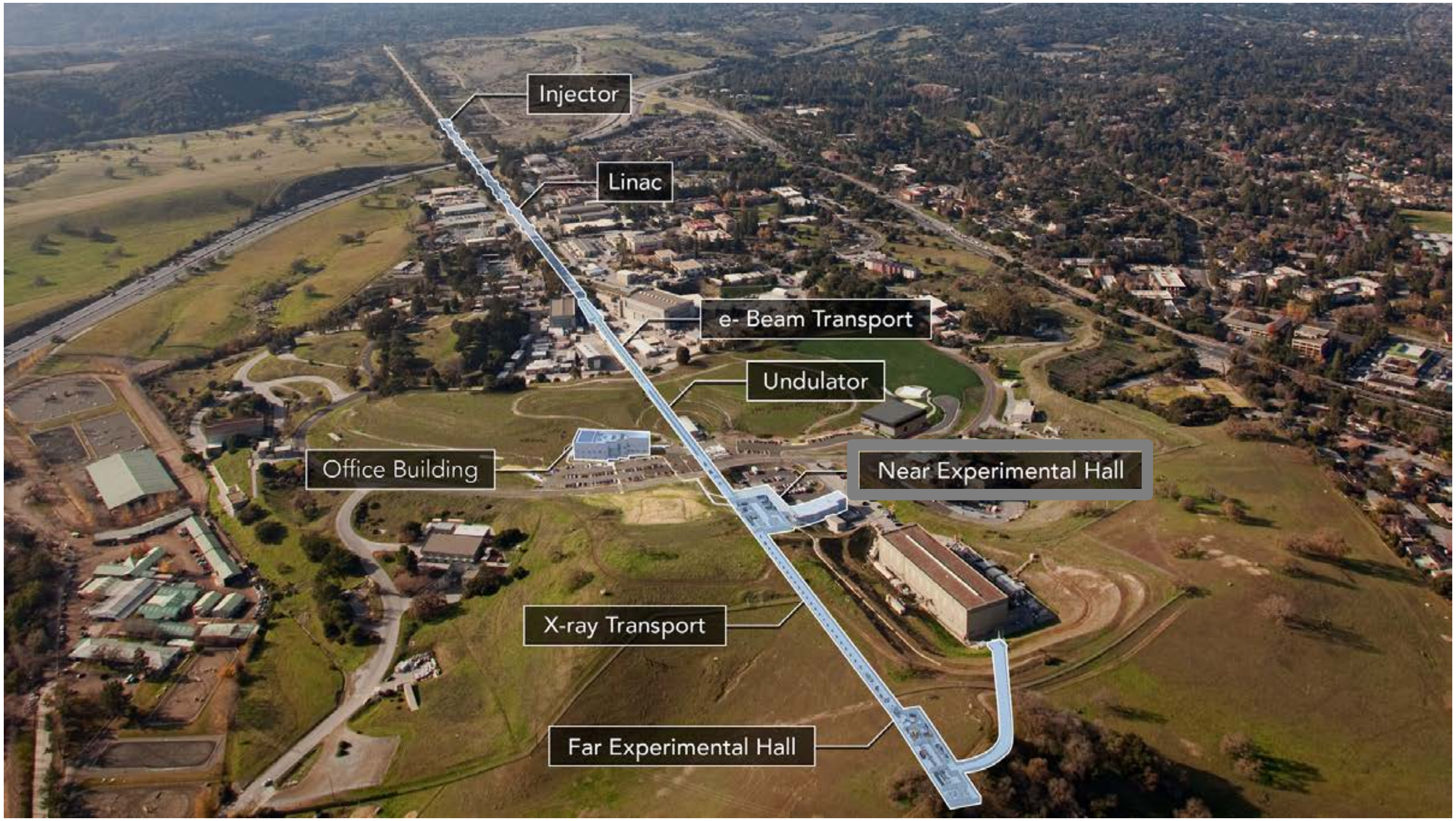



Trigo et al. Nature Physics. 9, 790, 2013

Zhu et al., Phys. Rev. B, 92, 054303, 2015

# The Linear Coherent Light Source (LCLS) at SLAC



# The XPP instrument at LCLS

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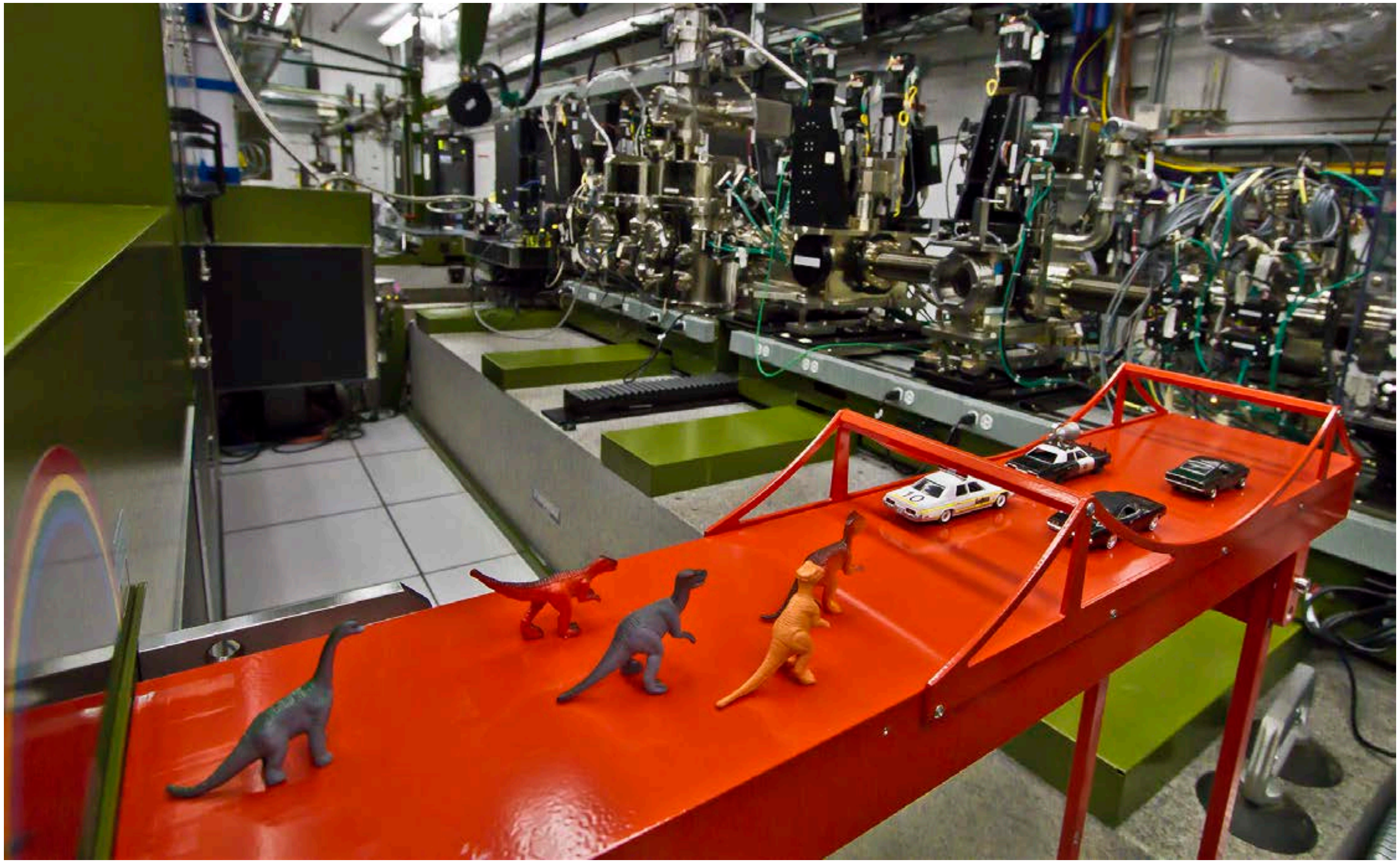
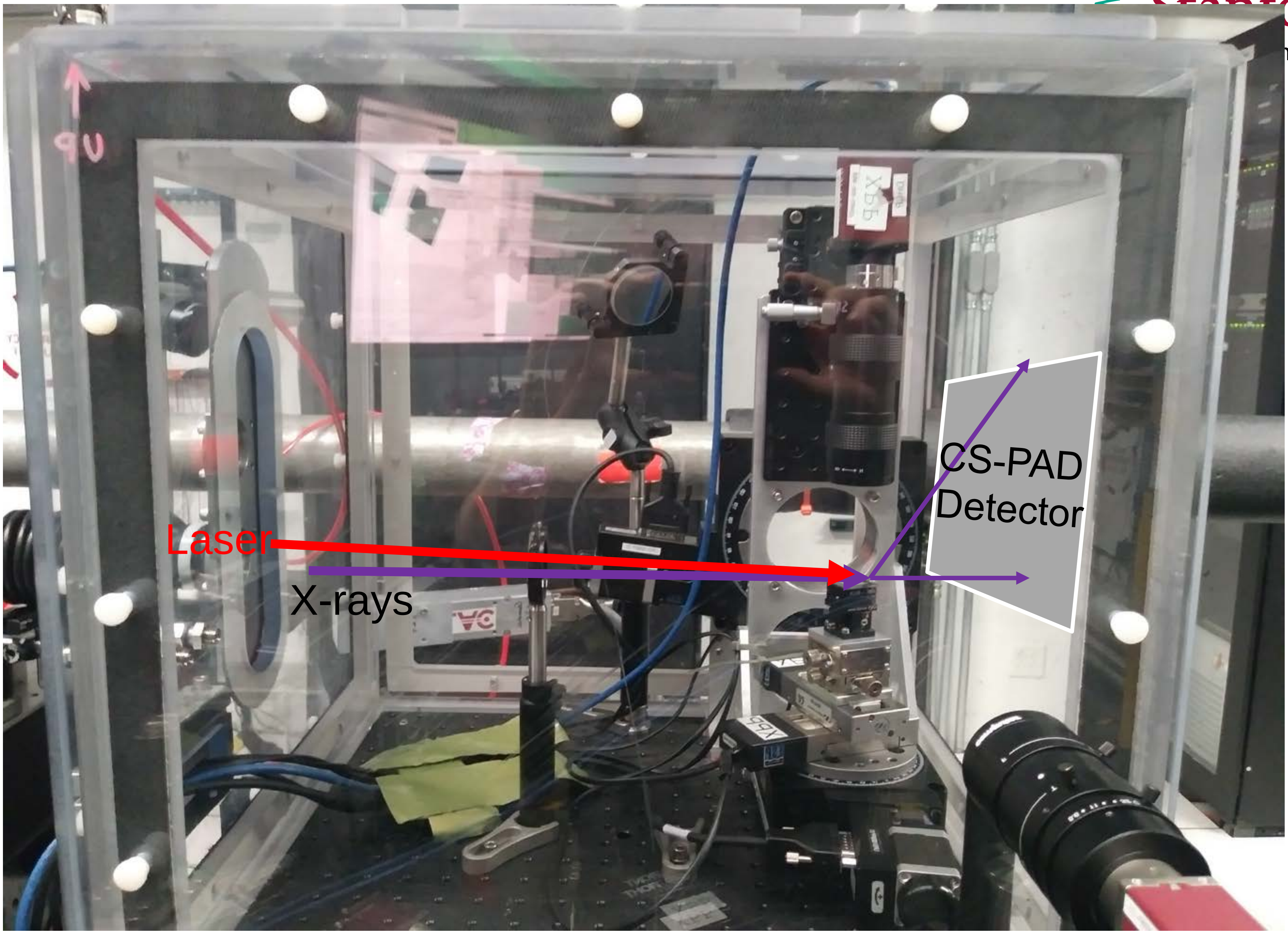
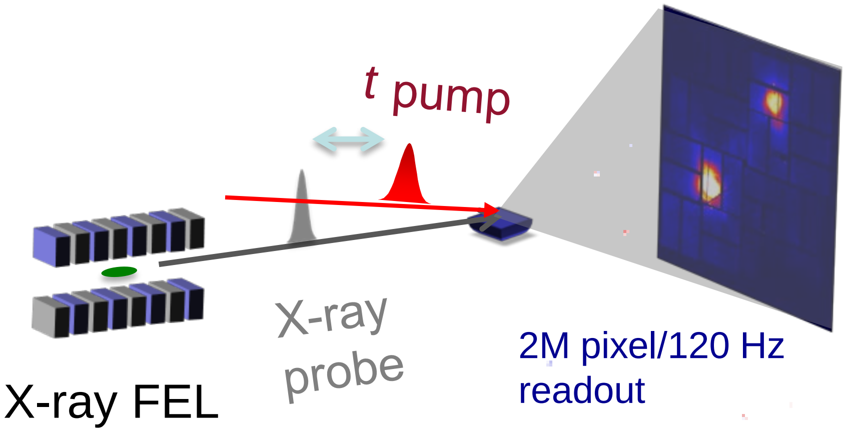
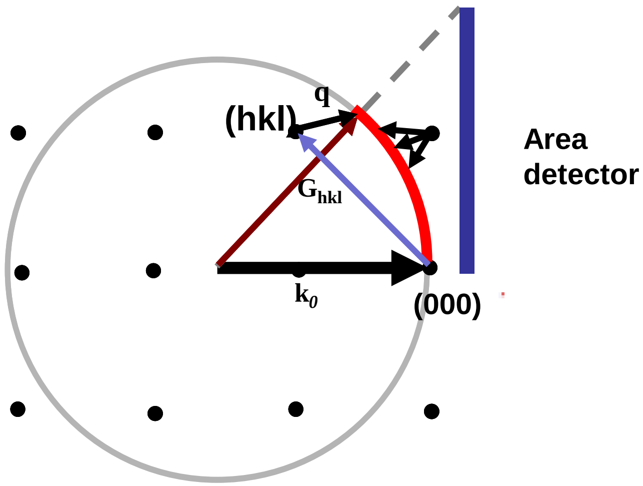


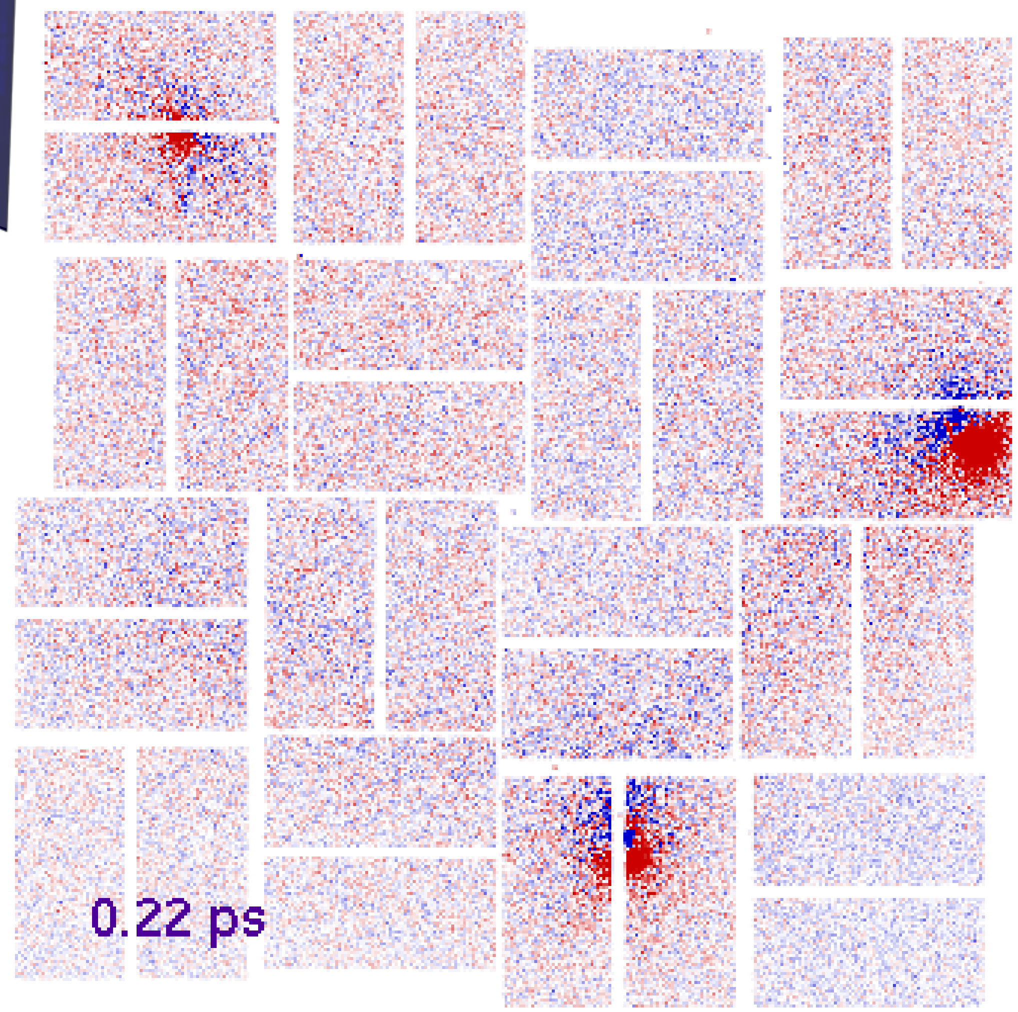
Photo credit: Daniele Fanelli


CS-PAD
Detector
Laser
X-rays

# Example, GaAs ~10fs pump/probe

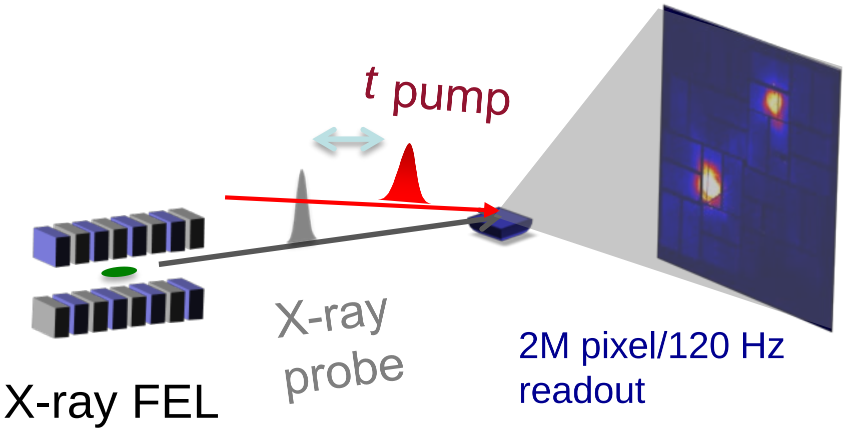


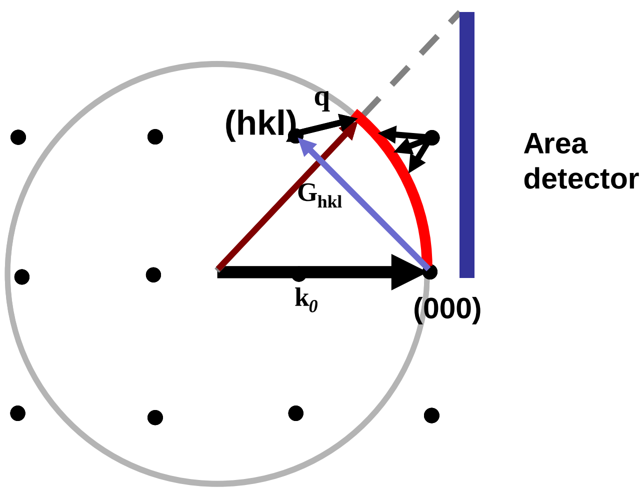



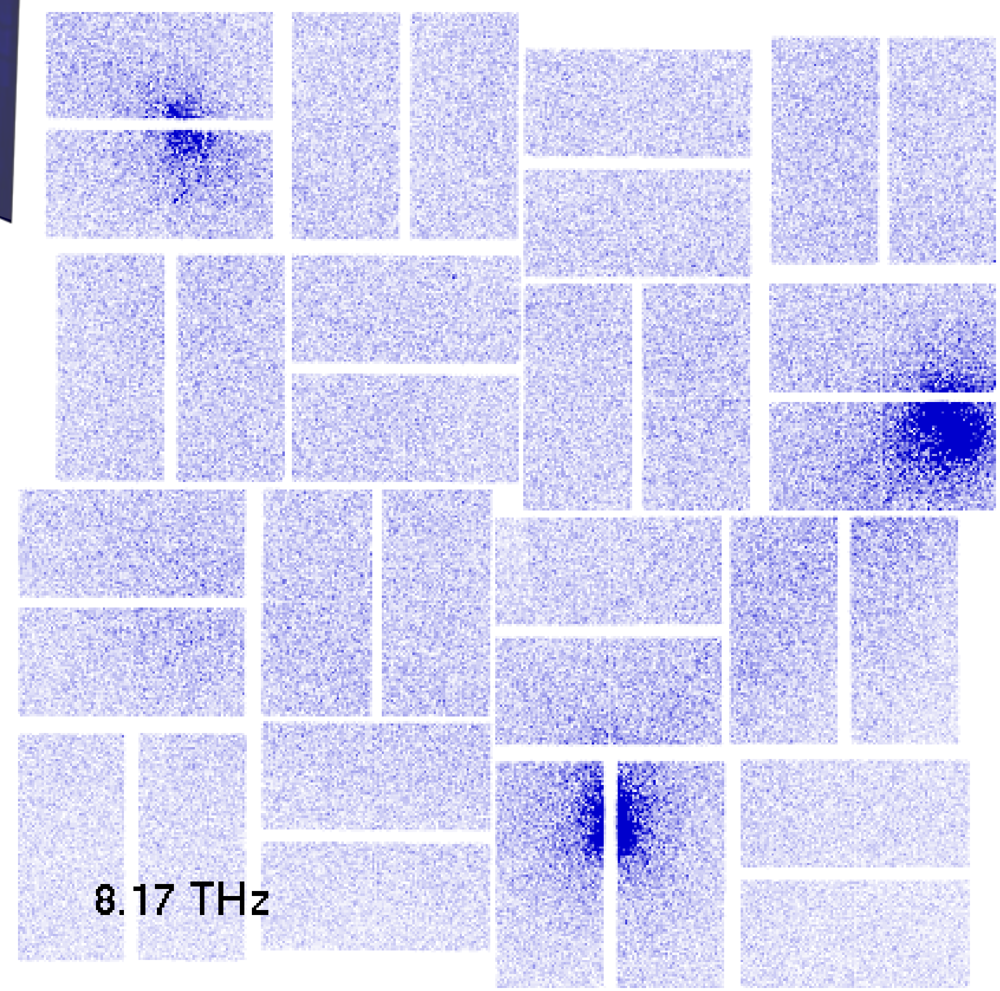




Zhu, Henighan, Trigo… unpublished

# Example, GaAs ~10fs pump/probe

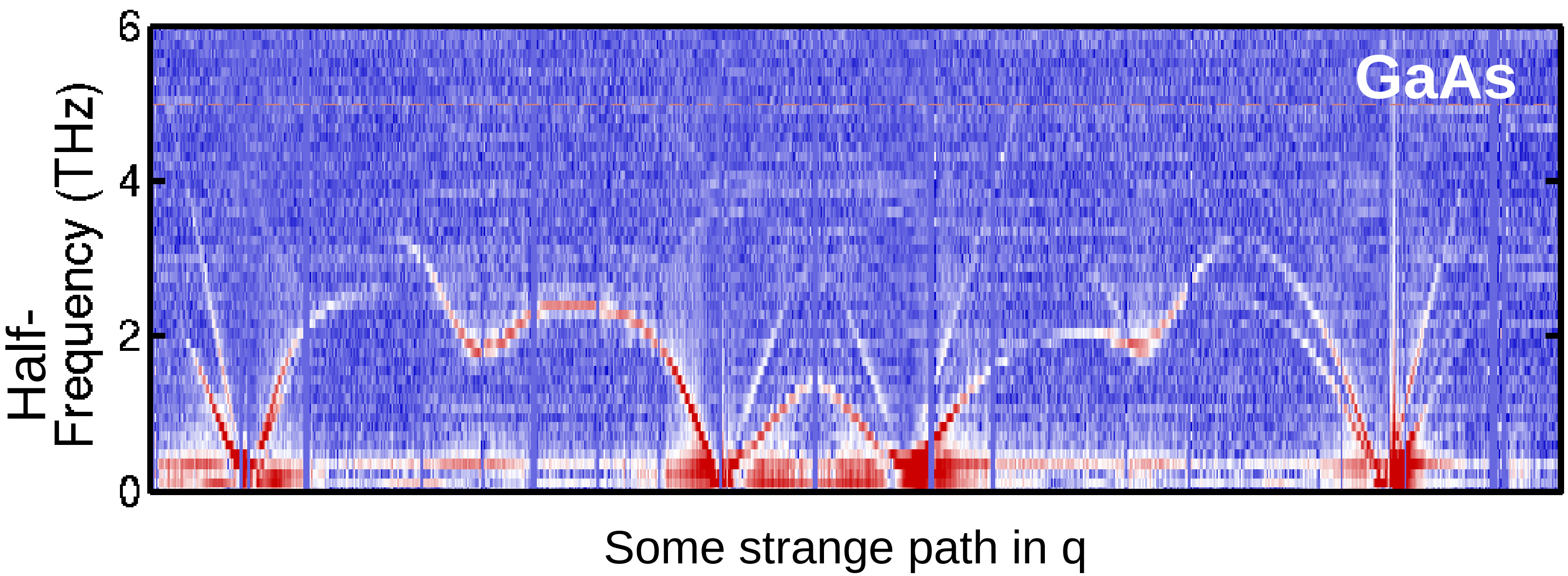



Zhu, Henighan, Trigo…
unpublished

# FT-IXS




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

Zhu, Henighan, Trigo…
unpublished

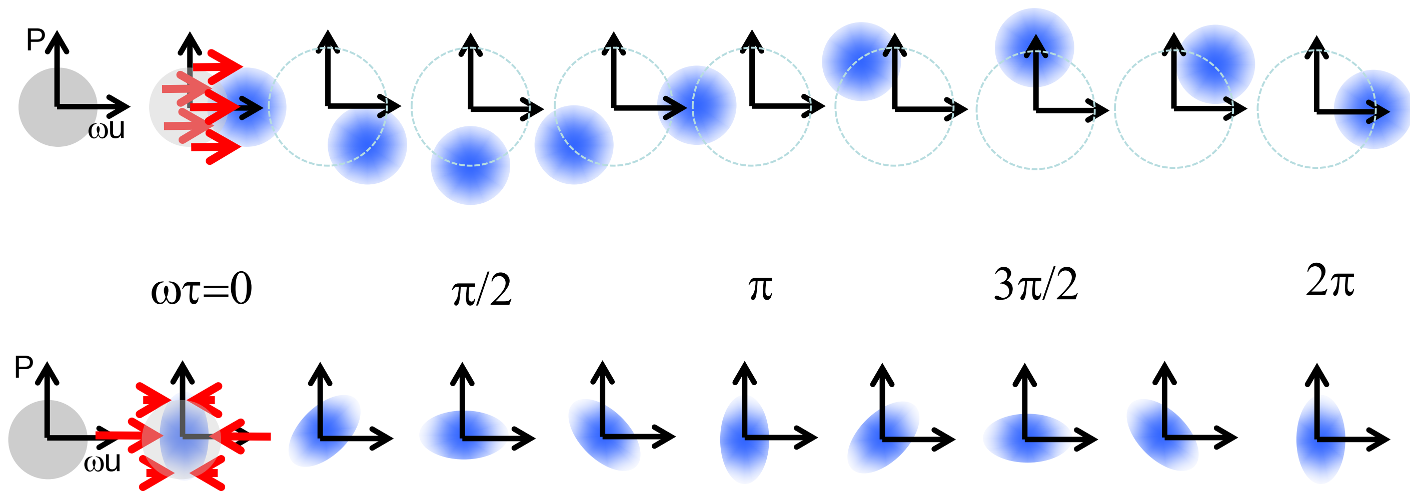
# Mechanism?

$$\langle u \rangle \propto cos\Omega t$$

$$\boxed{\langle u \rangle^2 \propto cos2\Omega t}$$

$$H' \propto I(t)u$$

1st order Raman     coherent state




$$\boxed{\langle u \rangle = 0}$$

$$H' \propto I(t)u^2$$

$$\boxed{\langle u^2 \rangle \propto cos2\Omega t}$$

2nd order Raman     squeezed state

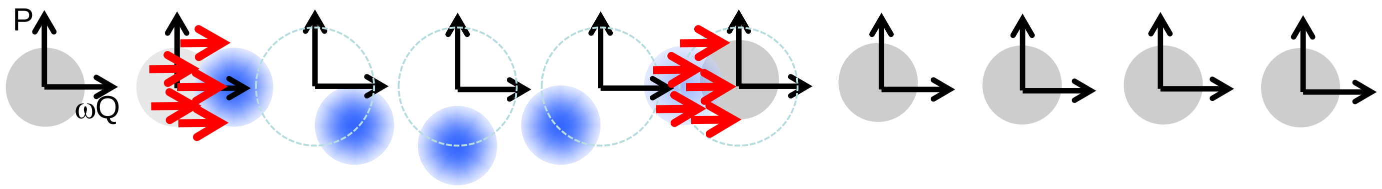
Henighan et al., 94, 020302(R) (2016)

# Mechanism?

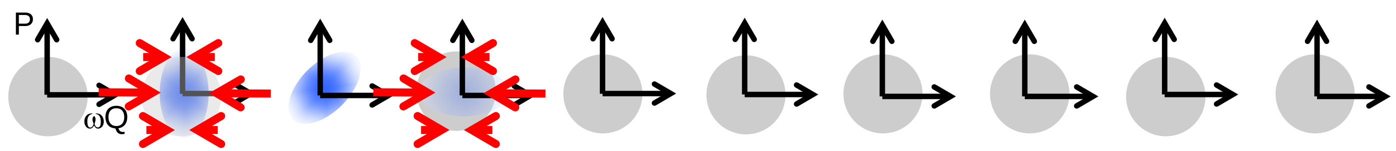
$$H' \propto I(t)u$$

1st order Raman coherent state

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




ωτ=0 π/2 π 3π/2 2π



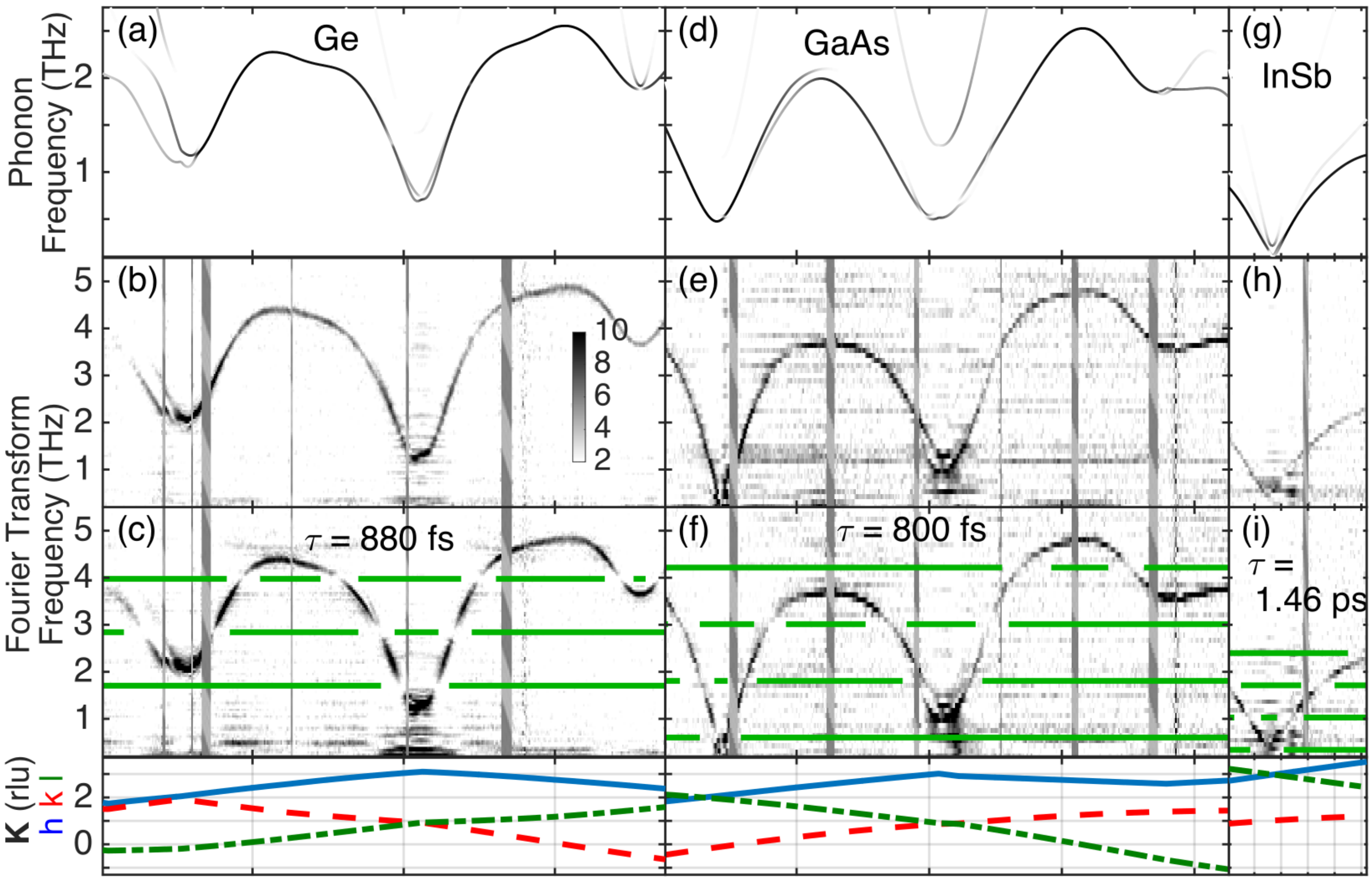

$$H' \propto I(t)u^2$$

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

2nd order Raman squeezed state

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

# For perfect crystals, squeezing…




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

# Stabilization of paraelectric phase in PbTe with photo-excitation

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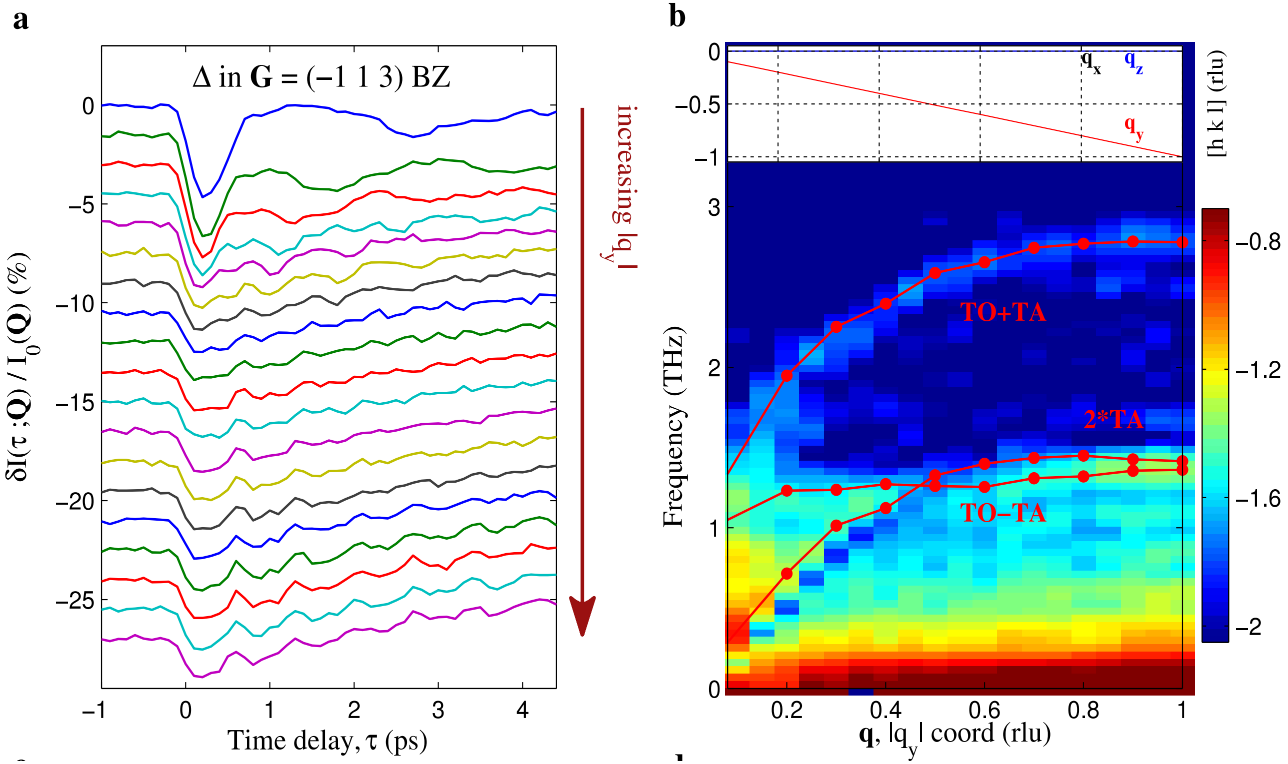
Transverse two-phonon Spectra along bonding direction (Γ–X )

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

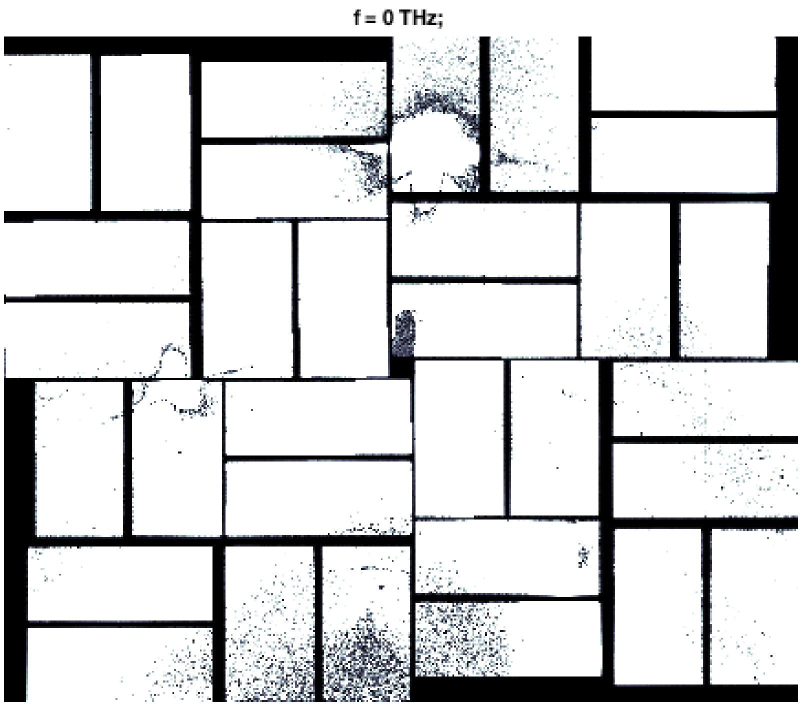


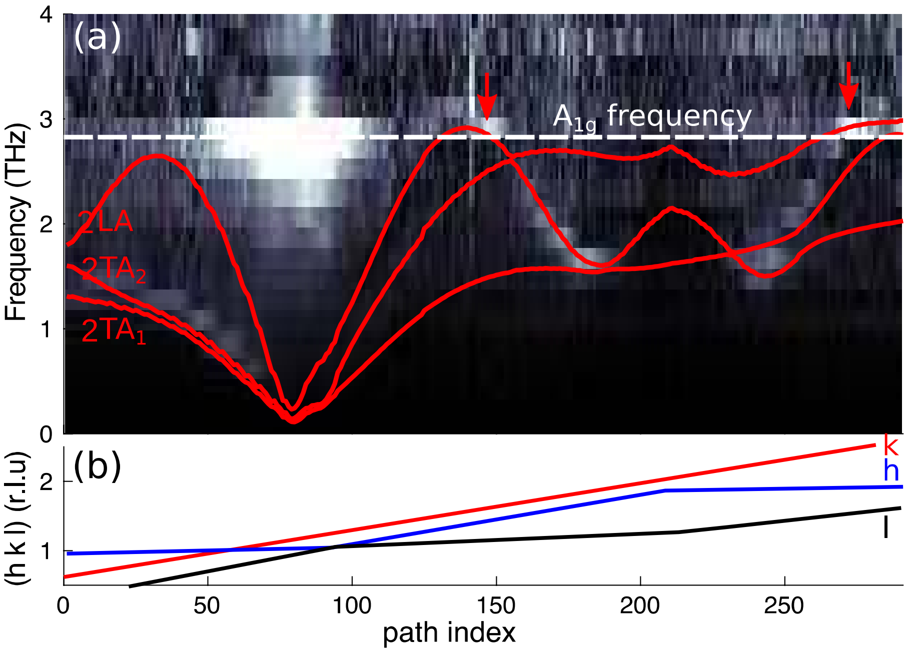

# Consitent with reduction of long-range interactions and Peierls-like electron-lattice instability

M.P. Jiang et al., Nat Commun, 7:12291, 2016.

# TR-diffuse scattering from photo-ex. Bi

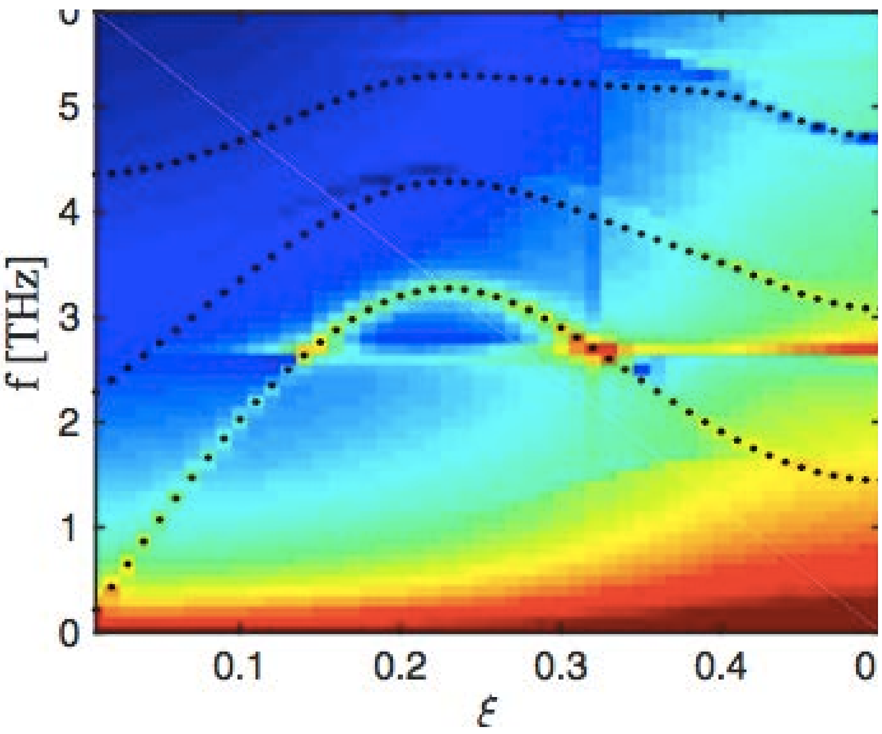
# FT-IXS spectra for Bi

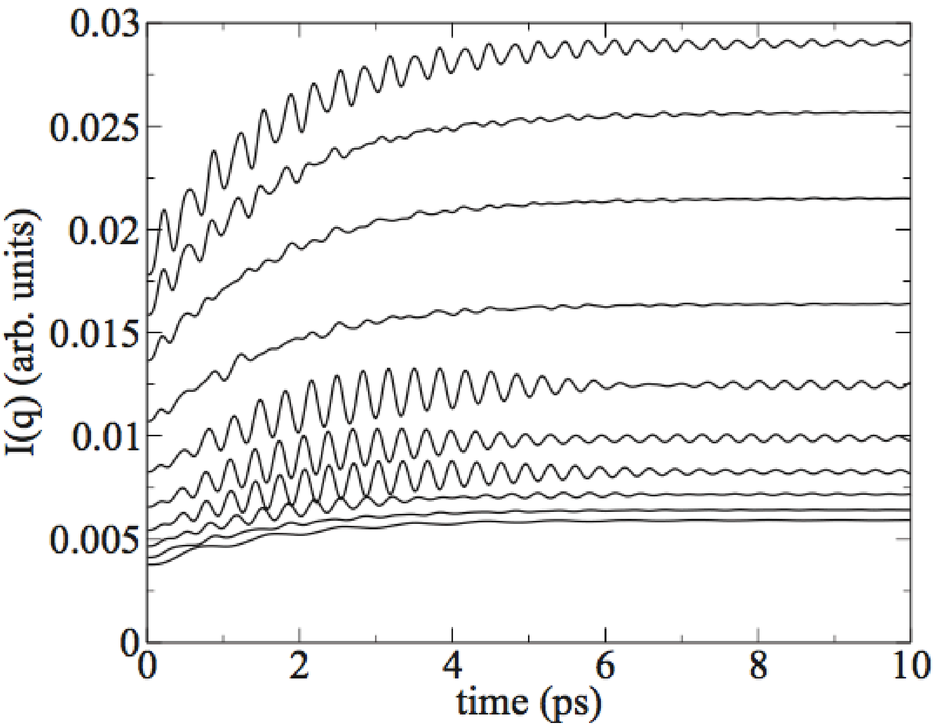



Teitlebaum, Trigo, Henighan et al.
PRL 121, 125901 (2018).

# Predictions for $A_{1g}$ decay in Bi




S. Fahy, E. D. Murray, and DAR, PRB 93:134308, 2016.

# Anharmonic decay of coherent phonon (parametric resonance)

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$$H = \underbrace{\frac{1}{2}\left( P_0^2 + \Omega^2 u_0^2 \right)}_{\text{Single phonon}} + \frac{1}{2N}\sum_q \underbrace{\left(P_q^2 + \overbrace{\omega_q^2\left(1 + 2g_q u_0\right)}^{\widetilde{\omega}_q^2(u_0)} u_q^2\right.}_{\text{Continuum of anharmonic phonons}}$$

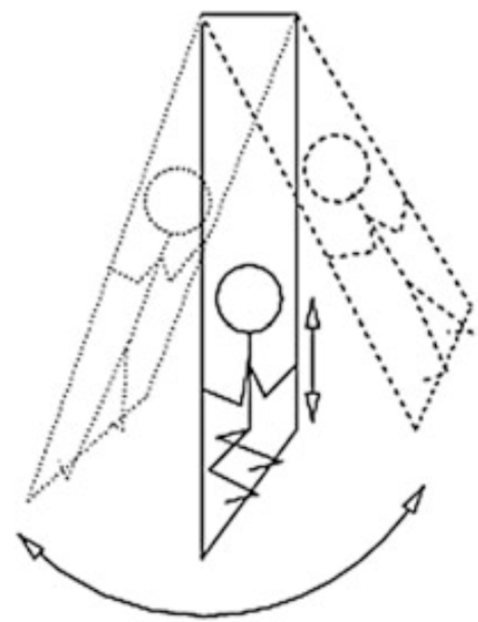
Coherent phonon:

$$\langle u_0 \rangle = A\left(1 - \cos(\Omega t) e^{-i\gamma_0 t}\right) \rightarrow \widetilde{\omega}_q(u_0) = \widetilde{\omega}_q(t)$$

Squeezed phonons:

$$\langle u_q^2 \rangle \sim g_{\boldsymbol{q}} A\, \omega_q / \gamma_q \left(e^{-\frac{\gamma_0 t}{2}} - e^{-\gamma_q t}\right) sin\Omega t$$

(on resonance)

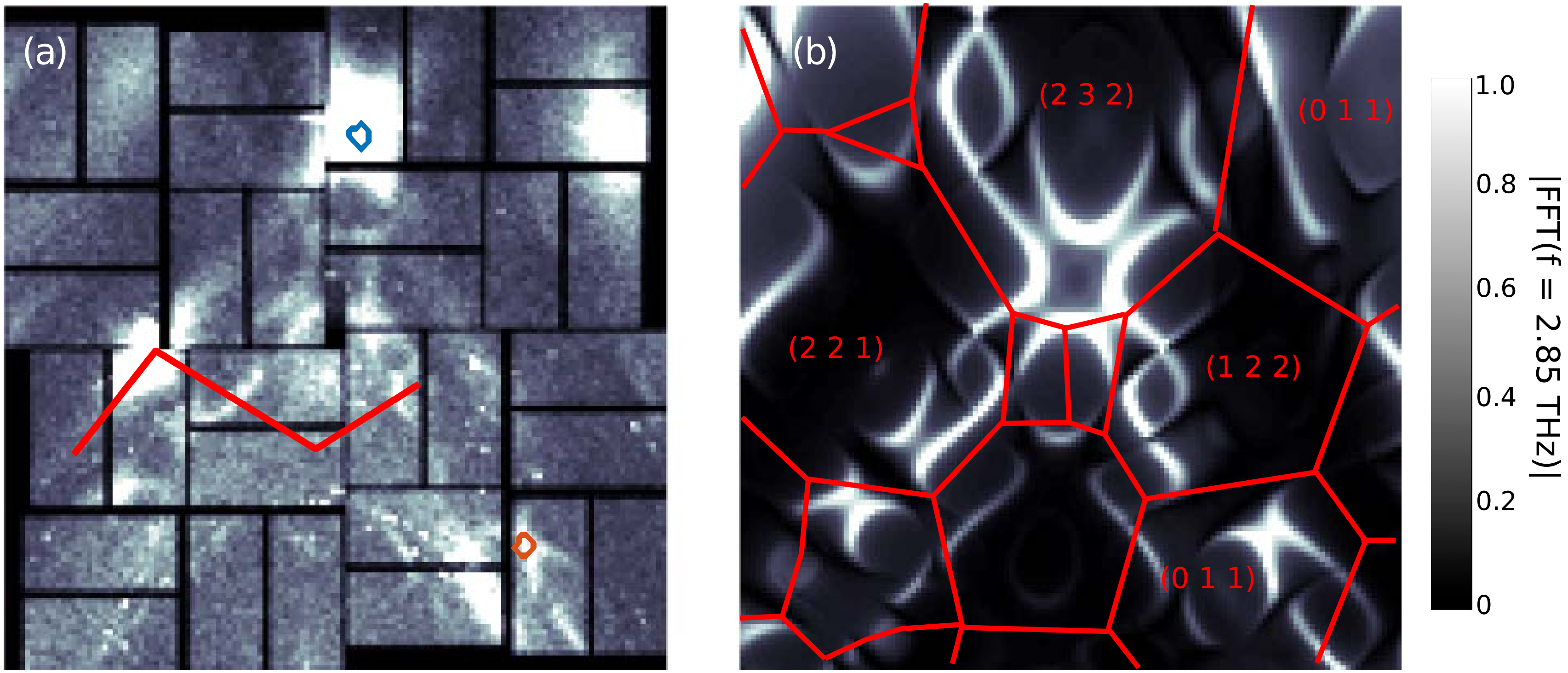


A. Champneys, Encyclopedia of Complexity and Systems Science

S. Fahy, E. D. Murray, and DAR, PRB 93:134308, 2016.

# Image of downcoverted phonons

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

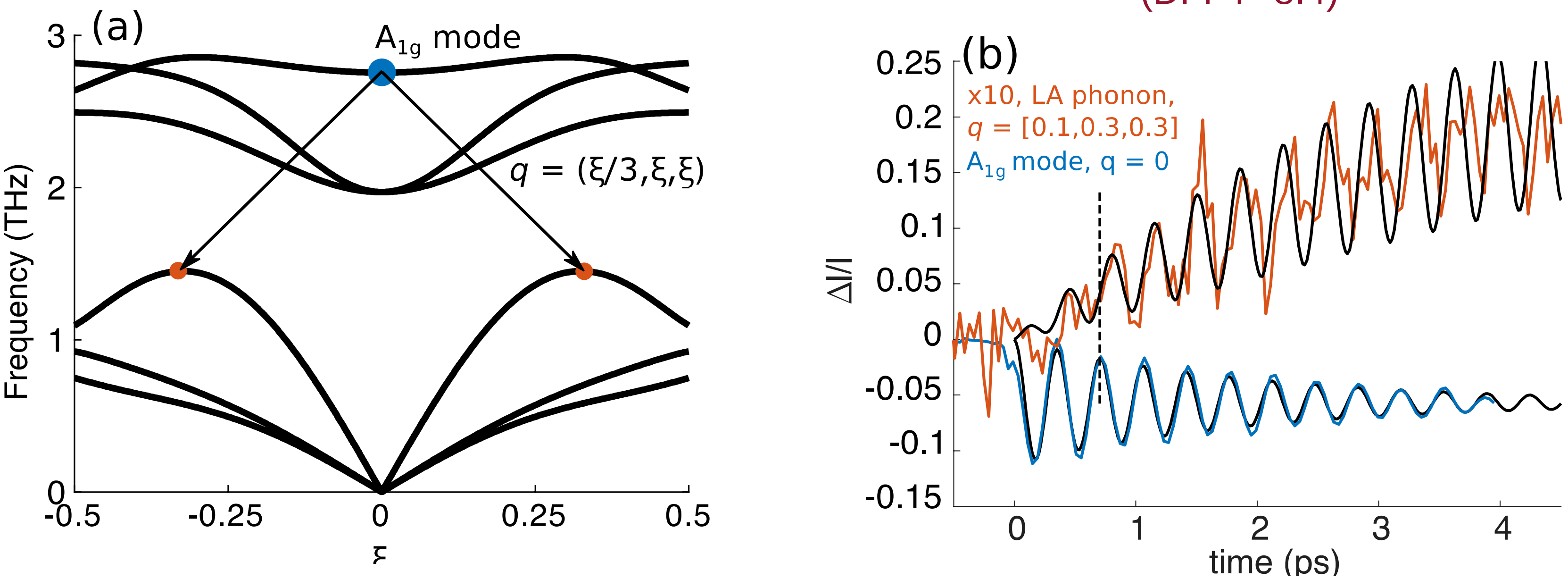



Teitlebaum, Trigo, Henighan et al.
PRL 121, 125901 (2018).

# Measurement of q-dependent coupling constants

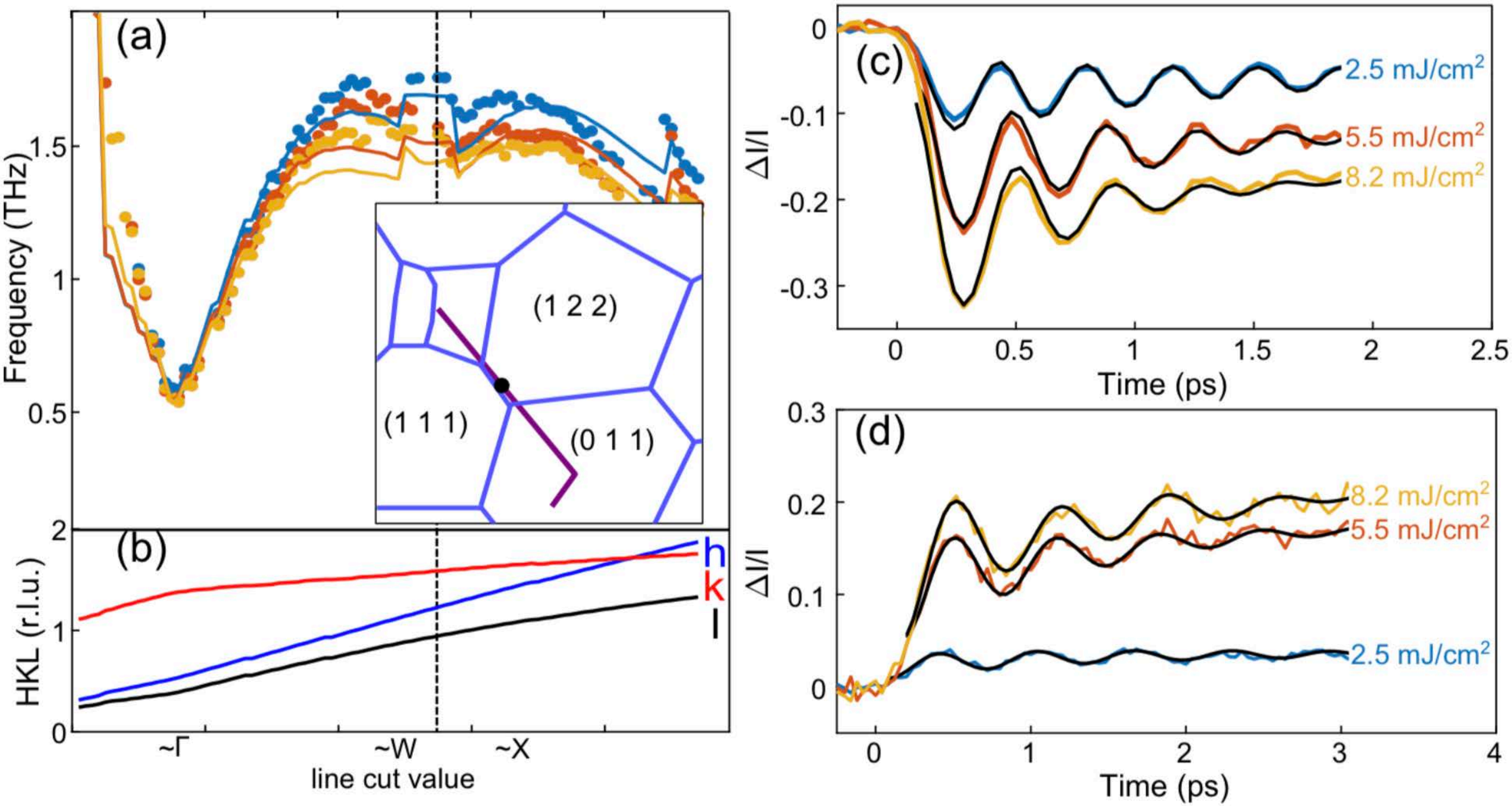


$g_q$ = -1.0
(DFPT -8.4)




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

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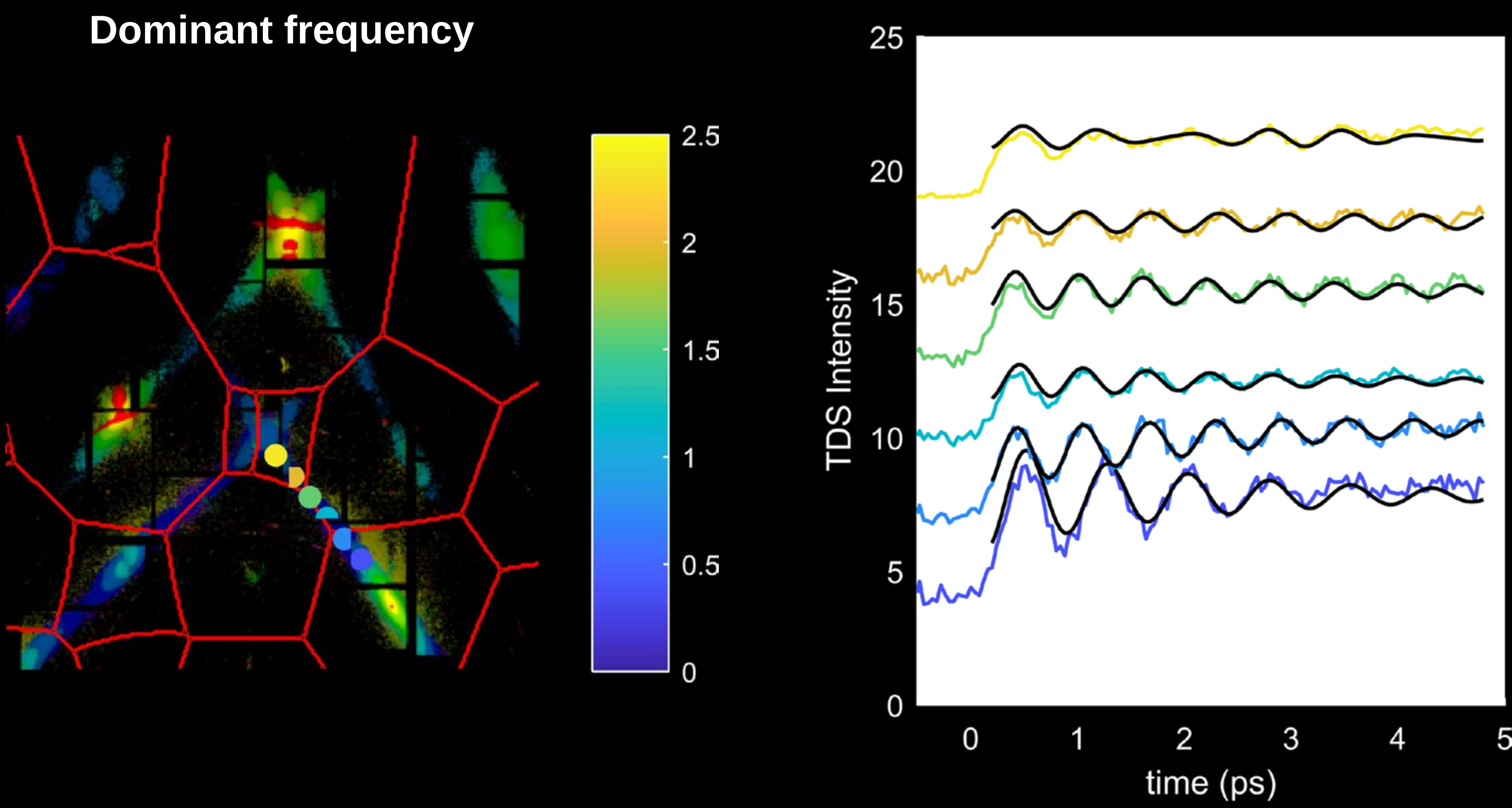
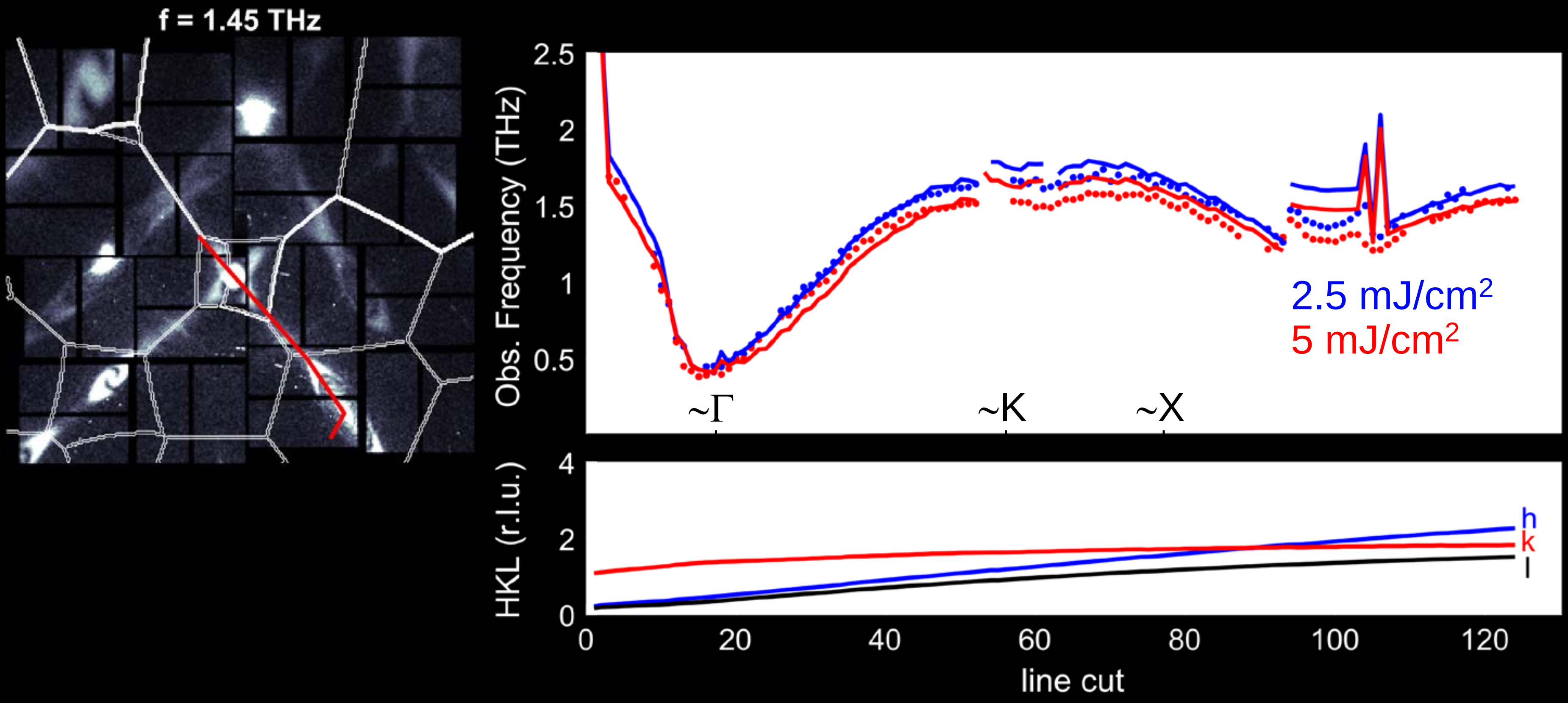
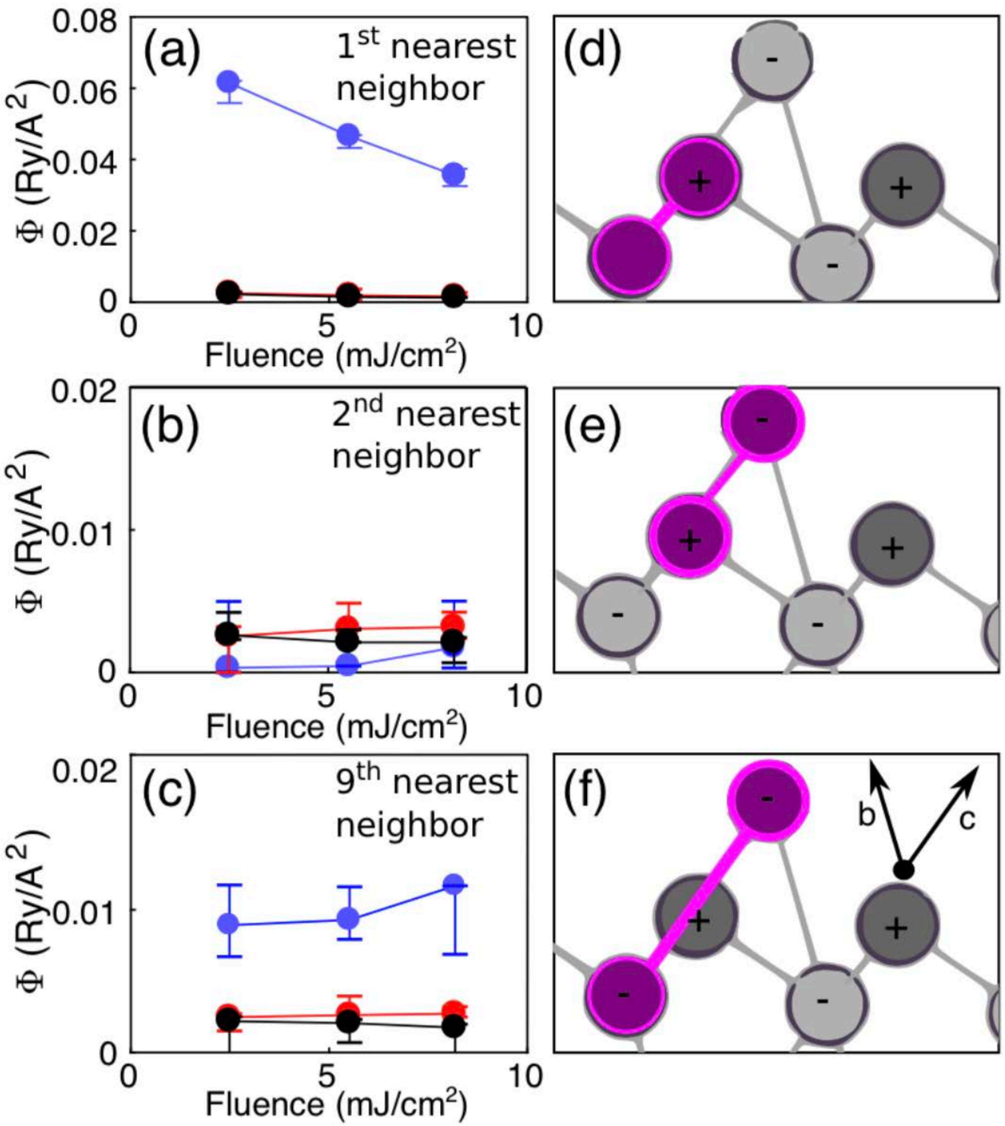


# …Fit modified dispersion to IF-model



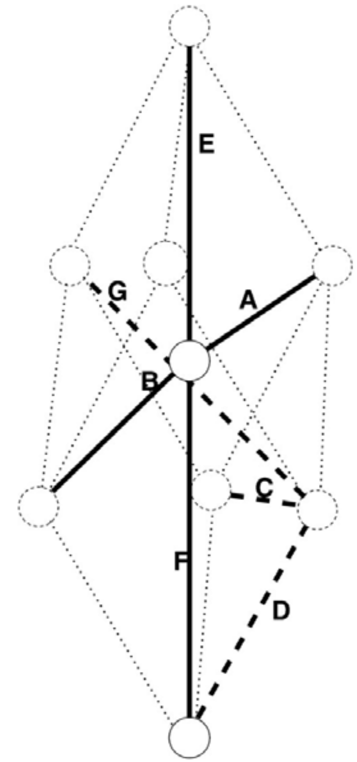
# Excited-state interatomic forces of Bi




A: 1nn

B: 2nn

G: 9nn



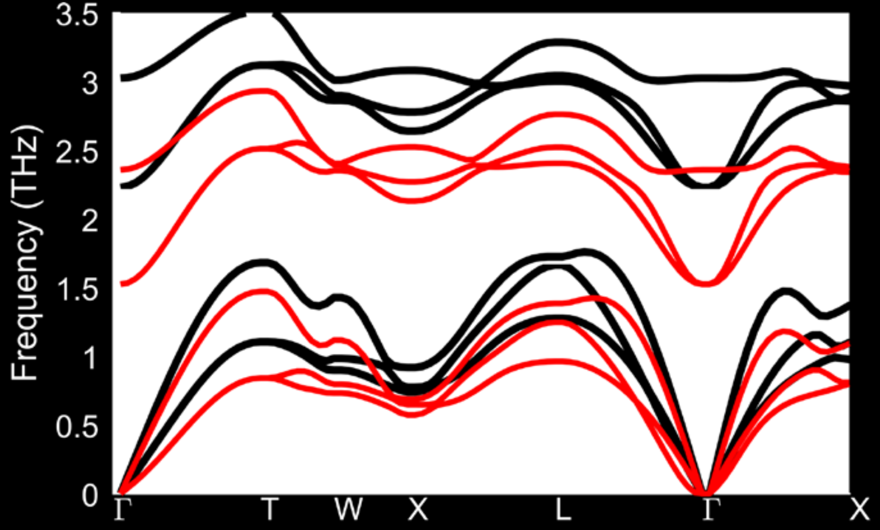

Theory (Ha/bohr)

| | | IFC Trace | | Distance | |
|---|---|---|---|---|---|
| Pair | NN | $n$=0% | $n$=1% | $n$=0% | $n$=1% |
| A | First | −33.23 | −27.74 | 5.766 | 5.807 |
| B | Second | −0.88 | −0.52 | 6.604 | 6.552 |
| C | Third | −0.67 | −0.86 | 8.544 | 8.544 |
| D | Fourth | −0.30 | −0.50 | 8.873 | 8.873 |
| E | Sixth | −0.26 | −0.16 | 10.360 | 10.439 |
| F | Eighth | −0.43 | −0.26 | 11.766 | 11.687 |
| G | Ninth | −4.16 | −4.40 | 12.318 | 12.318 |

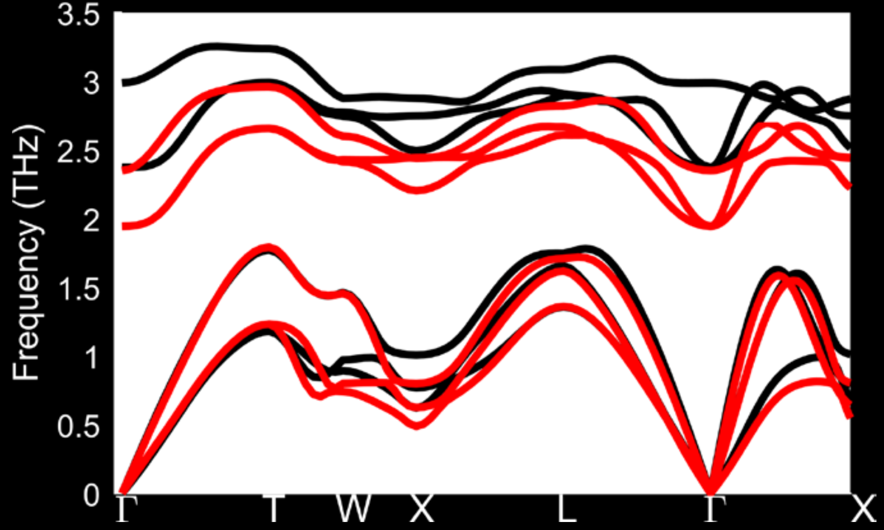
Preliminary, S.Teitelbaum et al. 2019

# Comparison with DFT calculations

Experimental (Reconstruction)




DPFT Calculations*




*ED Murray et al, PRB, (2007)

# Acknowledgments

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(Stanford PULSE Institute)

Recent past members (working on non-equilibrium phonons):
**Tom Henighan**, **Mason Jiang**, Mike Kozina, Crystal Bray, Jian Chen

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)