PULSE STANFORD

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Ultrafast processes in the solid state: Light scattering from elementary excitations

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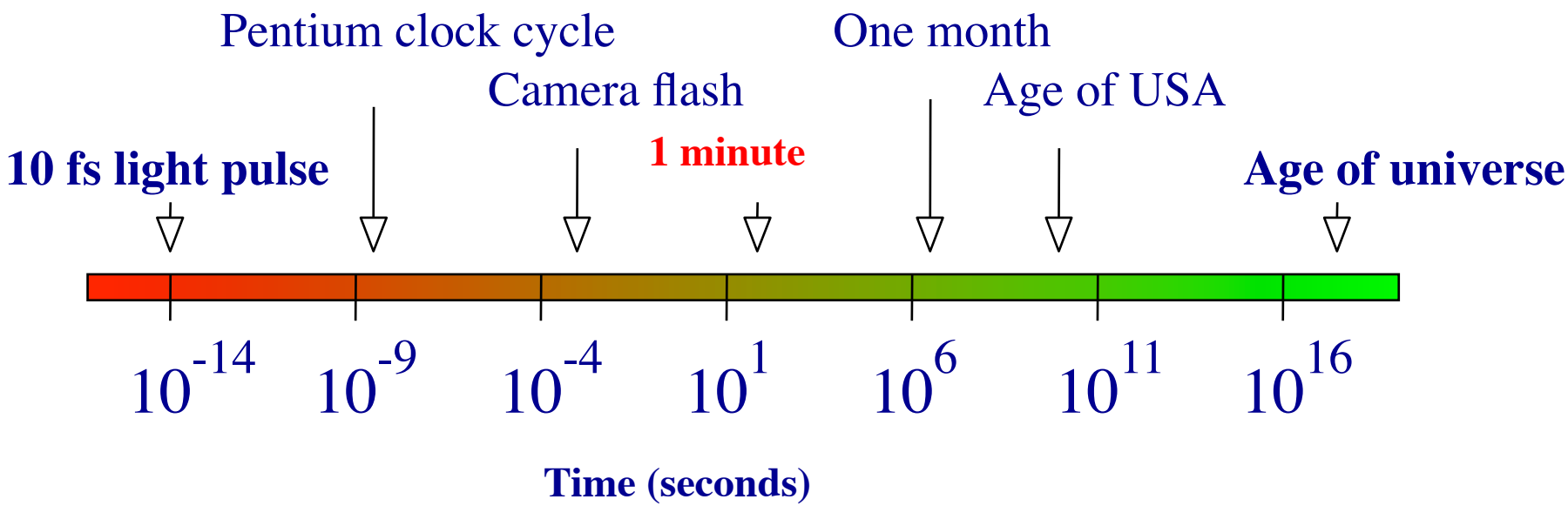
13th PSI Summer School August 9–15, 2014, Zug, Switzerland

Ultrafast Time Scales



Ultrafast Science explores the dynamics of the microscopic world:

- Making or breaking of chemical bonds
- Atomic and Electron dynamics in materials

attoseconds to femtoseconds:

electron motion/correlation,…

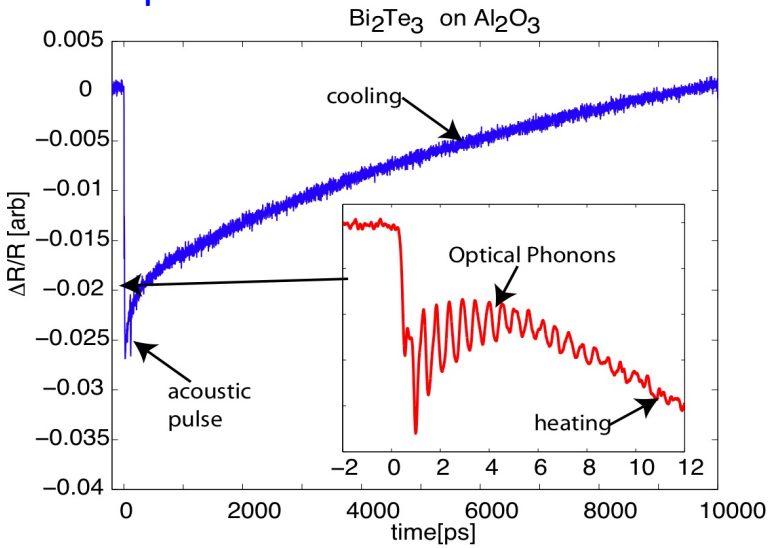
femtoseconds to picoseconds:

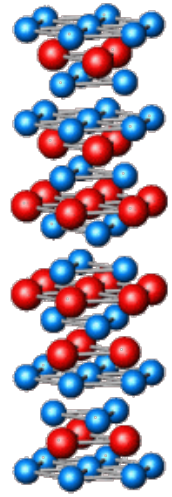
vibrational motion, electron-phonon and phonon-phonon scattering ,…

Adapted from M. Murnane

First, some advantages of time-domain measurements

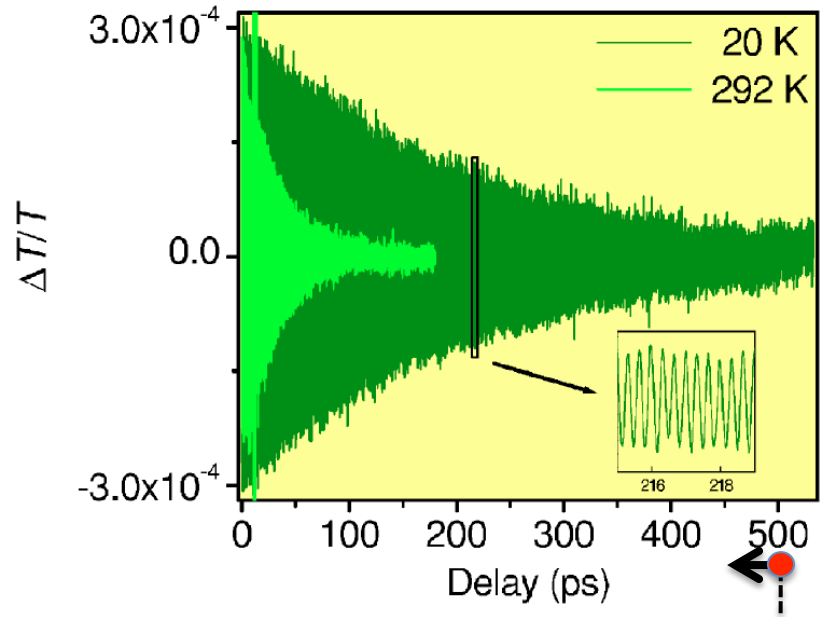
...separation of time-scales



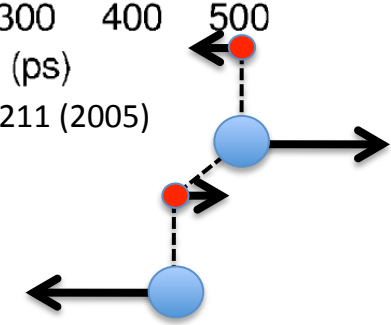


Sheu *et al.* unpublished

...sometimes just plain resolution!

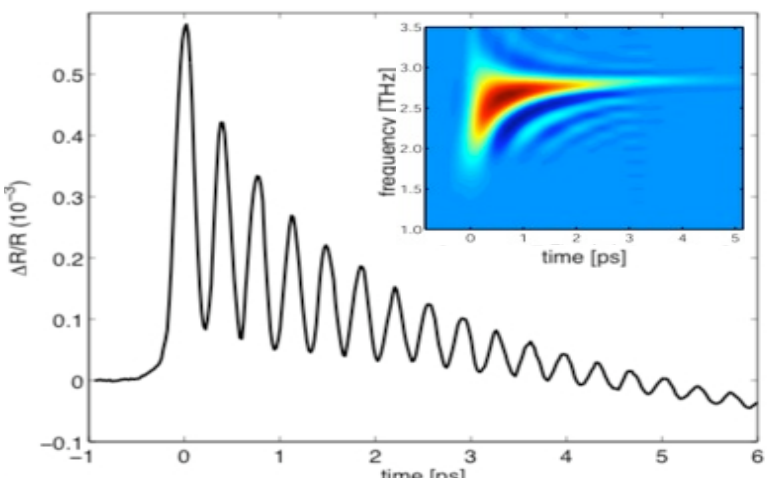


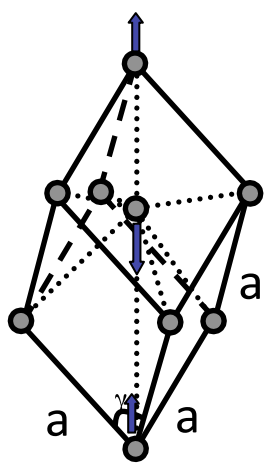
C. Aku-Leh, et al. PRB 71, 205211 (2005)



f=(2.9787 ± 0.0002) THz
1/Γ= (211 ± 7) ps @ 5K

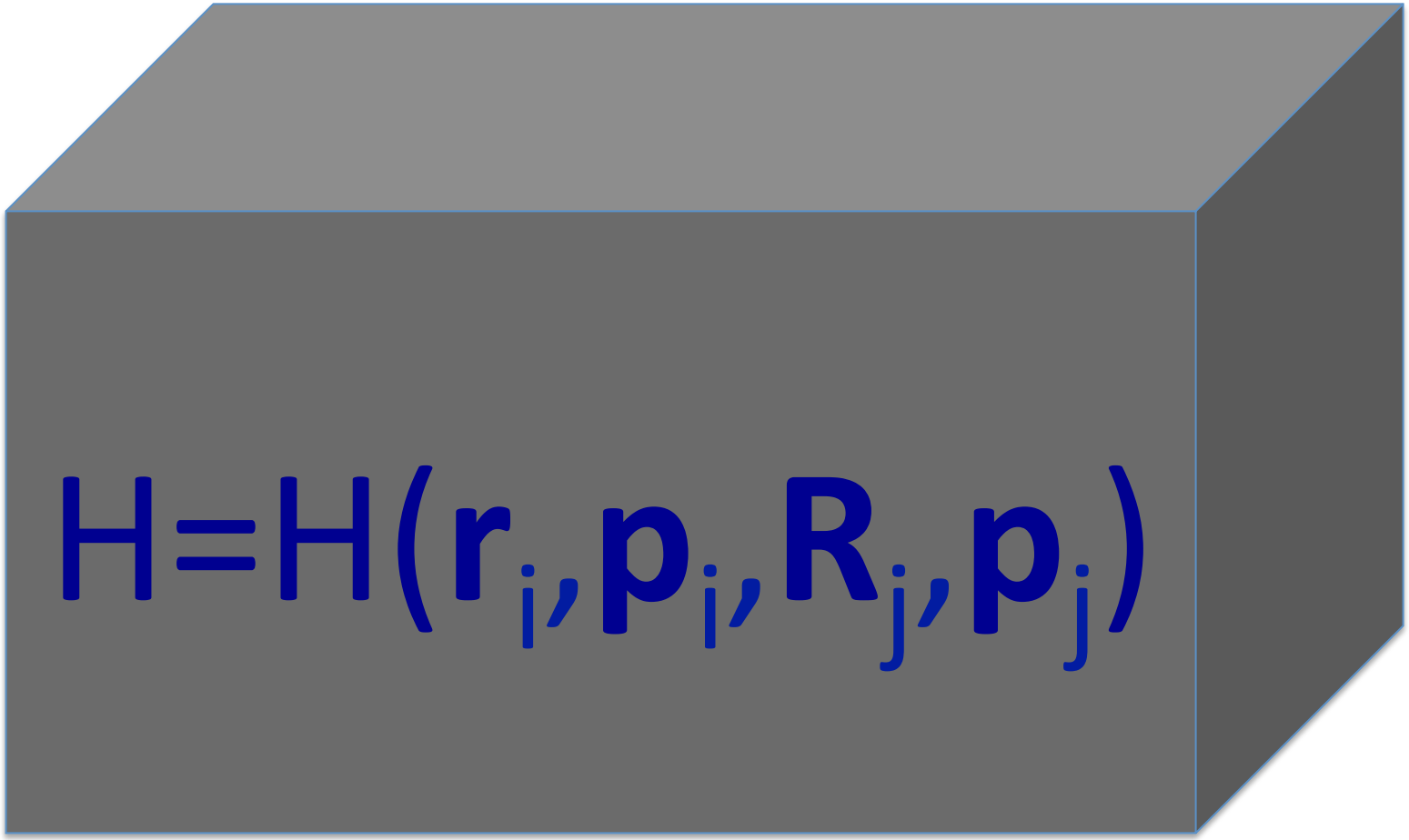
...Excited State Dynamics





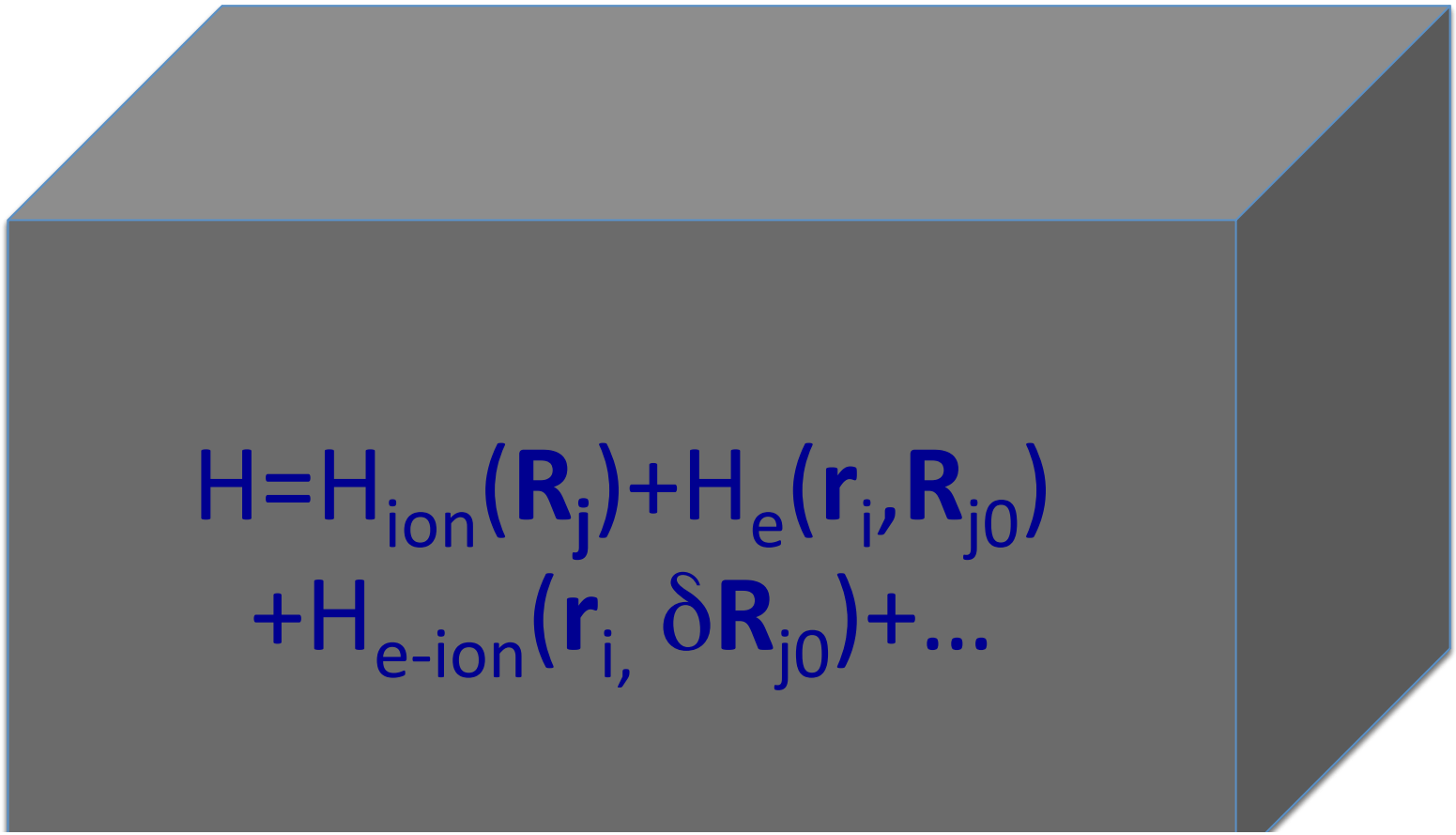
Murray *et al.* PRB 72, 060301 (R) 2005.

For perfect crystal, can write down Hamiltonian, but cannot solve.



$$H = \sum_i \frac{p_i^2}{2m_i} + \sum_j \frac{P_j^2}{2M_j} + \frac{1}{2}\sum_{j' \neq j} \frac{Z_{j'} Z_j e^2}{|\vec{R}_j - \vec{R}_{j'}|} - \sum_{i,j} \frac{Z_j e^2}{|\vec{r}_i - \vec{R}_j|} + \frac{1}{2}\sum_{i' \neq i} \frac{e^2}{|\vec{r}_i - \vec{r}_{i'}|}$$

For perfect crystal, can write down Hamiltonian, but cannot solve.



Born-Oppenheimer, valence/core and mean-field approximations,
+pert. theory with help from translational and point-group symmetries…
…single-particle excitations (electron-hole) and collective vibrations (phonons)

Elementary excitation soup

Stir in your favorite terms in the Hamiltonian…

Adapted from A. Warhol

Light Scattering and connection with elementary excitations

Response to Applied Field

$$P_i(\vec{r},t) = \int \chi_{ij}(\vec{r}',\vec{r},t',t)E_j(\vec{r}',t')\mathrm{d}\vec{r}'\mathrm{d}t'$$

$$\Updownarrow$$

$$P_i(\vec{q},\omega) = \chi_{ij}(\vec{q},\omega)E_j(\vec{q},\omega)$$

$$\epsilon_{ij}(\vec{q},\omega) = 1 + 4\pi\chi_{ij}(\vec{q},\omega)$$

- In linear Response, χ doesn't depend on E
- Causality, P follows E
- We will see excitation spectrum related to peaks in imaginary χ, ε
- Also related to the dynamical structure factor and van Hove correlations

At long wavelengths local response

$$P(t) = \int_0^\infty \chi(t')E(t-t')\mathrm{d}t'$$

$$\epsilon(\omega) = \epsilon_r(\omega) + i\epsilon_i(\omega) = 1 + \int_0^\infty 4\pi\chi(t')\mathrm{e}^{i\omega t'}\mathrm{d}t'$$

Kramers-Kronig, follows from causality

$$\epsilon_r(\omega) - 1 = \frac{2}{\pi} Pr \int_0^\infty \frac{\omega'\epsilon_i(\omega')}{\omega'^2 - \omega^2}\mathrm{d}\omega'$$

$$\epsilon_i(\omega) = -\frac{2\omega}{\pi} Pr \int_0^\infty \frac{\epsilon_r(\omega')}{\omega'^2 - \omega^2}\mathrm{d}\omega'$$

Can obtain real from imaginary part and vice versa, given knowledge everywhere
However, note that primary contribution near ω

Microscopic picture of light interacting with electrons/ions

Microscopic picture: Light-matter interaction as perturtabation to (single electron, or ion) Hamiltonian

$$H_0 = \frac{p^2}{2m} + V(\vec{r})$$

$$\vec{E} = -\frac{1}{c}\frac{\partial \vec{A}}{\partial t}, \vec{B} = \nabla \times \vec{A} \qquad \Phi = 0, \nabla \cdot \vec{A} = 0$$

$$\vec{p} \rightarrow \vec{p} - \frac{e\vec{A}}{c}$$

$$H = H_0 + H_{eR} = \frac{p^2}{2m} + V(\vec{r}) + \frac{e}{mc}\vec{A} \cdot \vec{p} + \frac{e^2 A^2}{2mc^2}$$

- In dipole approx. p•A equivallent to μ•E
- A^2 term typically neglected at optical wavelengths in linear response regime
- A^2 dominant for x-ray scattering except very near resonance

Long-wavelength make dipole approximation

$$\lambda \gg a, q \approx 0 \rightarrow \vec{A} \approx \vec{A}_0 \mathrm{e}^{i\omega t} + c.c.$$

Time dependent perturbation theory gives transition rate for SPE...

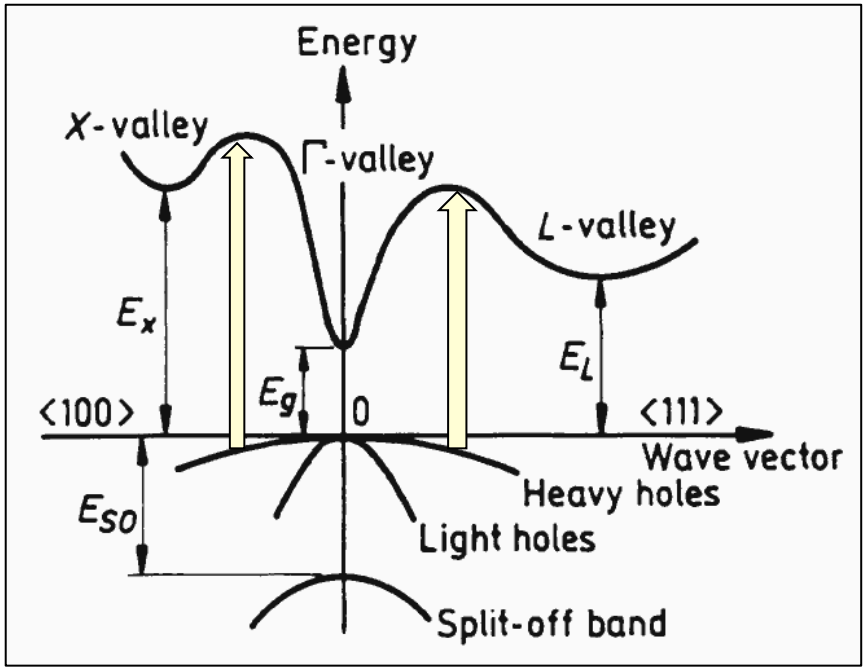
$$w(\omega) = \frac{2\pi}{\hbar}\left(\frac{e}{m\omega}\right)^2\left(\frac{E(\omega)}{2}\right)^2 \sum_{\vec{k}} |P_{cv}|^2 \delta(E_c(\vec{k}) - E_v(\vec{k}) - \hbar\omega)$$

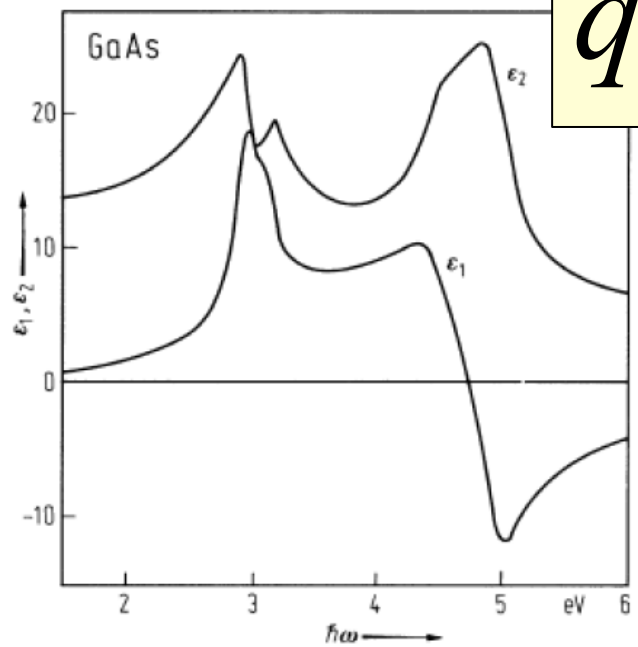
Same matrix elements appear in Im{ε}

$$\epsilon_i(\omega) = \left(\frac{2\pi e}{m\omega}\right)^2 \sum_{\vec{k}} |P_{cv}|^2 \delta(E_c(\vec{k}) - E_v(\vec{k}) - \hbar\omega)$$

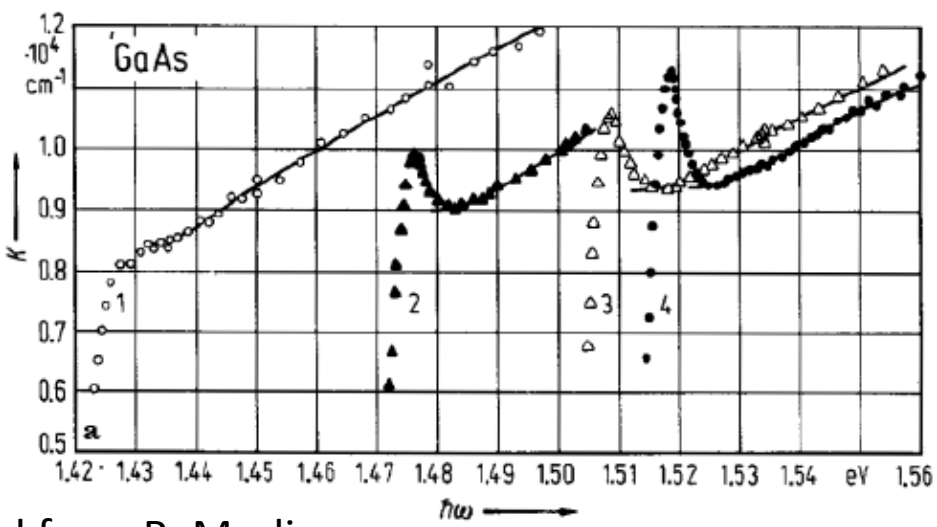
- Similar for phonons, plasmons and other excitations
- More generally for finite q

FIRST-ORDER COUPLING TO ELECTRONS





$$q_e \approx q_h$$



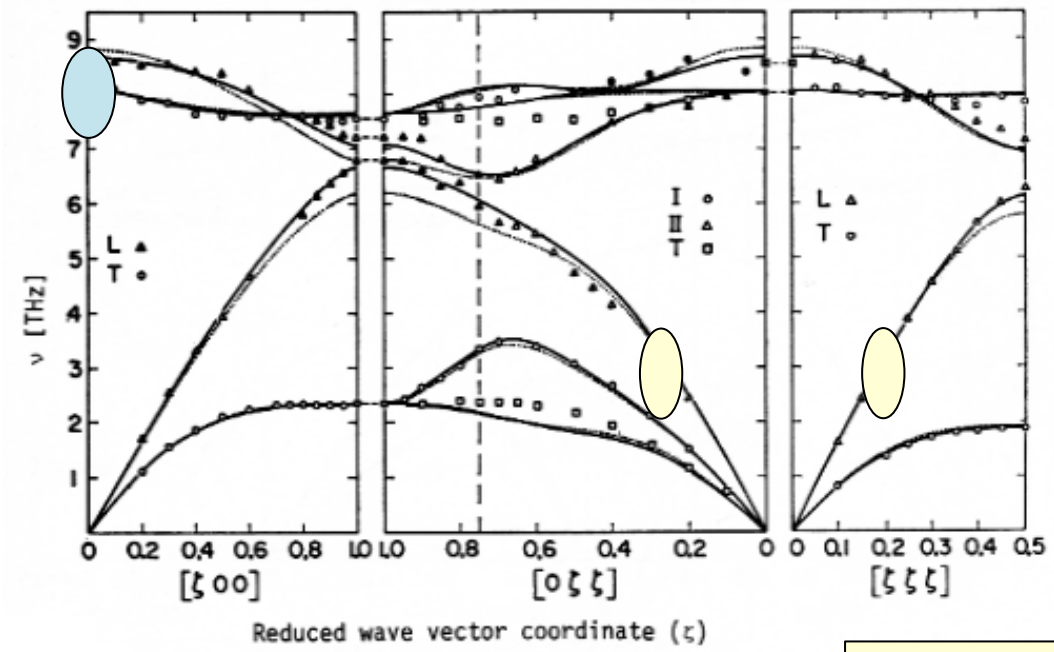
Adapted from R. Merlin.

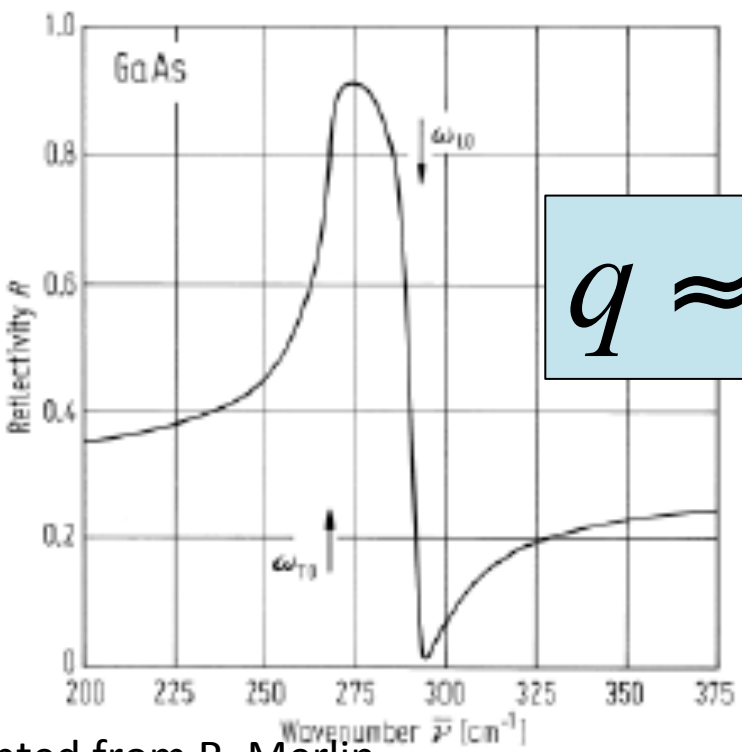
q=0 excitons

DISCRETE (exciton) vs. (interband) CONTINUUM

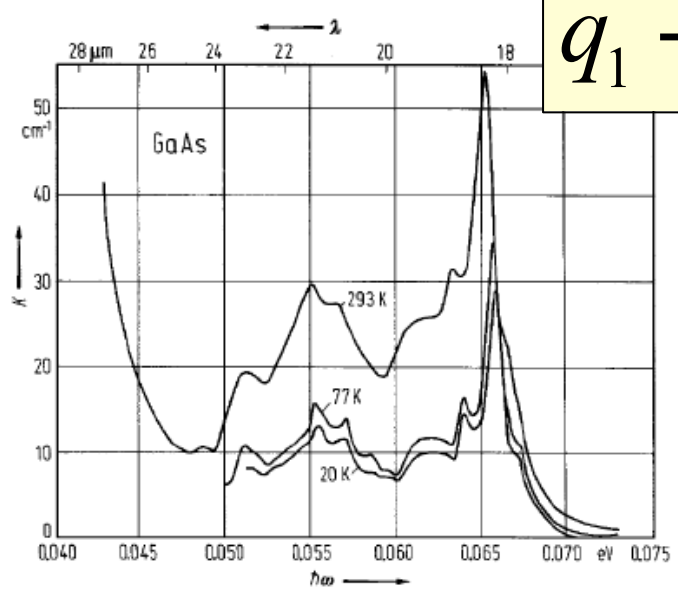
FIRST-ORDER COUPLING TO PHONONS (FAR TO MID INFRARED)

GaAs





$$q \approx 0$$

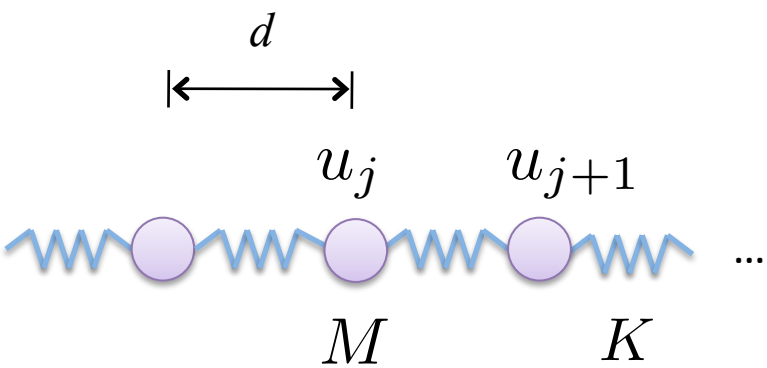


$$q_1 + q_2 \approx 0$$

DISCRETE (1 phonon) vs. (2 phonon) CONTINUUM

Adapted from R. Merlin.

Phonons in 1D



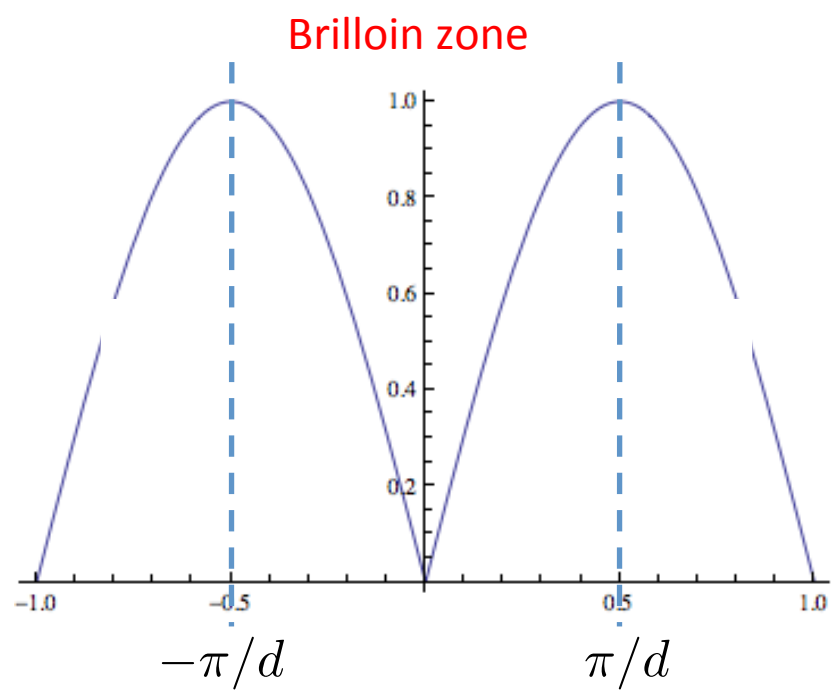
$$M\frac{d^2u}{dt^2} = K(u_{j+1} - 2u_j + u_{j-1})$$

Propose solutions: $u_j = \epsilon e^{i(qR_j - \omega t)}$

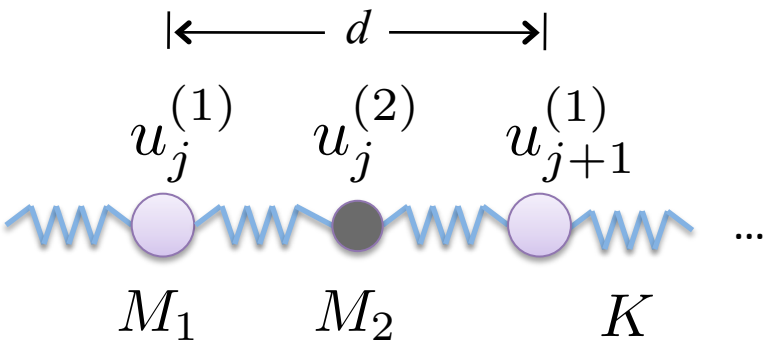
relationship between ω and **q**, dispersion relation:

$$\omega = 2\sqrt{\frac{K}{M}}|\sin(qd/2)|$$

ω linear for $|q| << 1/d$: acoustic phonons



Phonons in 1D



Now $M_1 \neq M_2$

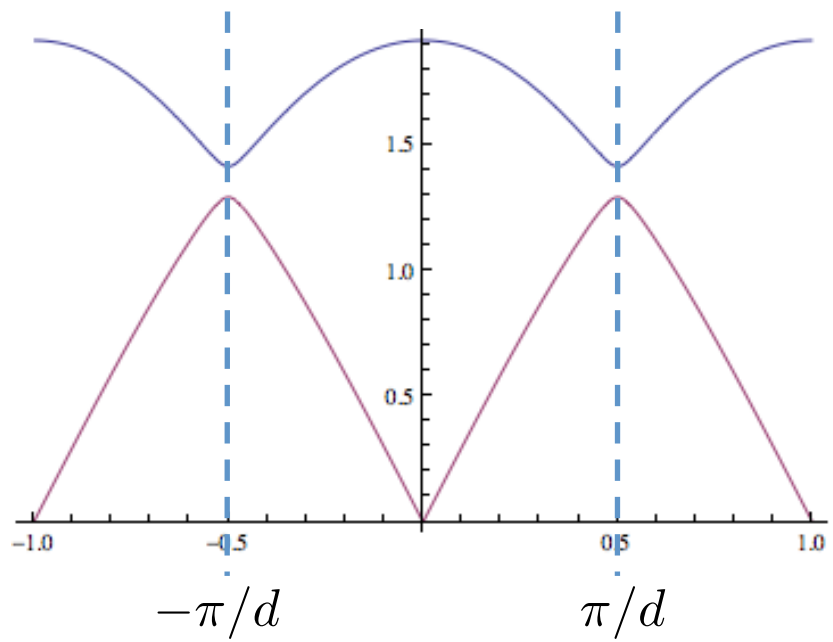
$$M_1 \frac{d^2 u^{(1)}}{dt^2} = K(u_j^{(2)} - 2u_j^{(1)} + u_{j-1}^{(2)})$$

$$M_2 \frac{d^2 u^{(2)}}{dt^2} = K(u_{j+1}^{(1)} - 2u_j^{(2)} + u_j^{(1)})$$

Propose solutions: $u_j^{(1,2)} = \epsilon^{(1,2)} e^{i(qR_j - \omega t)}$

dispersion relation:

Brilloin zone



FIRST-ORDER COUPLING TO PHONONS (EXCITONS)

$$H = -\vec{\mu}_{\text{ions}}.\vec{\mathbf{E}} = -\left(\vec{\alpha}Q_0 + \sum_q \vec{\beta}^q Q_q^2 + \ldots\ldots\right).\vec{\mathbf{E}}$$

$$\ddot{Q}_0 + \Omega_0^2 Q_0 = \sum_i \alpha_i E_i$$

Driven Oscillator

Parametric Resonance

$$\ddot{Q}_q + \Omega_q^2 Q_q = \left(\sum_i \beta_i^q E_i\right) Q_{-q}$$

RAMAN COUPLING TO PHONONS (TRANSPARENT MEDIA)

DISPLACEMENTS
($u \equiv$ ions ; $Q \equiv$ phonons)

⇩

CHANGE IN DIELECTRIC RESPONSE

⇩

CHANGE IN ENERGY DENSITY

⇩

FORCE DENSITY

ELECTROMAGNETIC ENERGY DENSITY

$$U = \varepsilon \left| E(\mathbf{r},t) \right|^2 / 8\pi$$

$$\delta\varepsilon = \boxed{\sum_{im} (\partial\varepsilon / \partial u_{im})\ u_{im}} + \boxed{\sum_{ijmn} (\partial^2\varepsilon / \partial u_{im} \partial u_{jn})\ u_{im} u_{jn}} + \ldots$$

$$\delta U \approx \delta\varepsilon_{\mathbf{q}=0} \left| E(\mathbf{r},t) \right|^2 / 8\pi = \frac{\left| E(\mathbf{r},t) \right|^2}{8\pi} \times$$

$$\boxed{\sum_{s} (\partial\varepsilon / \partial Q_{s,\mathbf{q}=0})\ Q_{s,\mathbf{q}=0}} + \boxed{\sum_{st,\mathbf{q}} (\partial^2\varepsilon / \partial Q_{s,\mathbf{q}} \partial Q_{t,-\mathbf{q}}) Q_{s,\mathbf{q}} Q_{t,-\mathbf{q}}} + \ldots.$$

$$\boxed{F \propto \left| E^2(t) \right|}$$

FIRST-ORDER IMPULSIVE FORCE

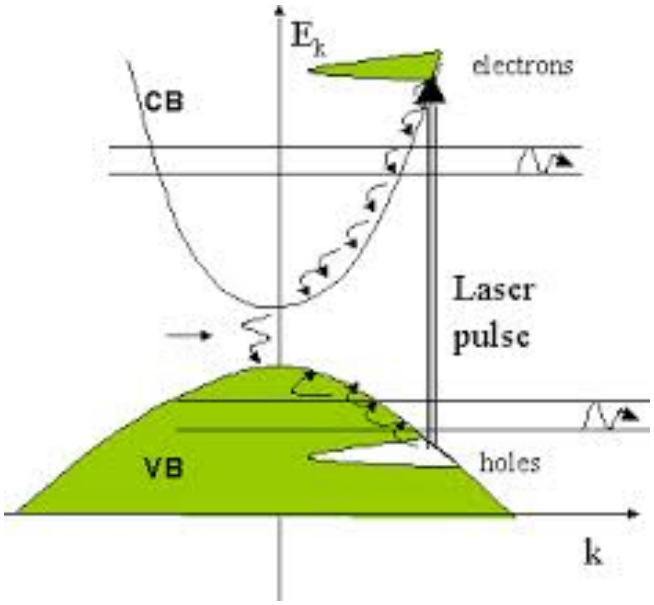
$$\boxed{F \propto Q_{\mathbf{k}} \left| E^2(t) \right|}$$

SECOND-ORDER IMPULSIVE CHANGE OF FREQUENCY

IMPULSIVE STIMULATED RAMAN SCATTERING

Non-Raman Mechanisms?

Displacive Excitation of Coherent Phonons



(WORKS ONLY FOR FULLY-SYMMETRIC MODES)

Adapted from R. Merlin.

Ti_2O_3

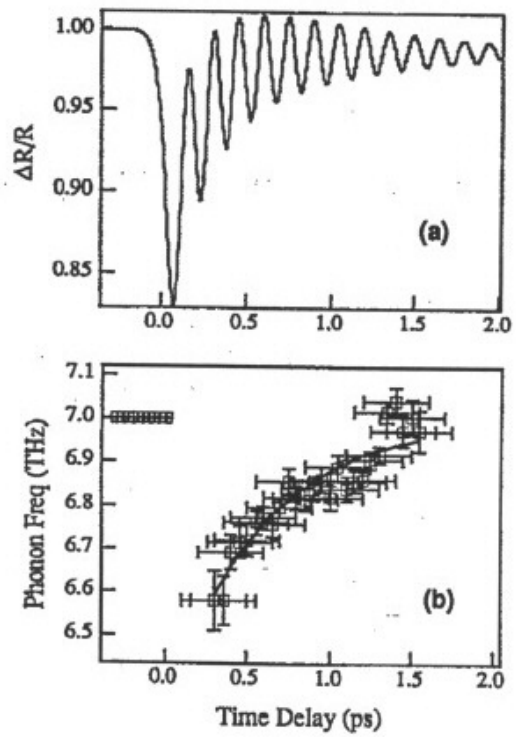
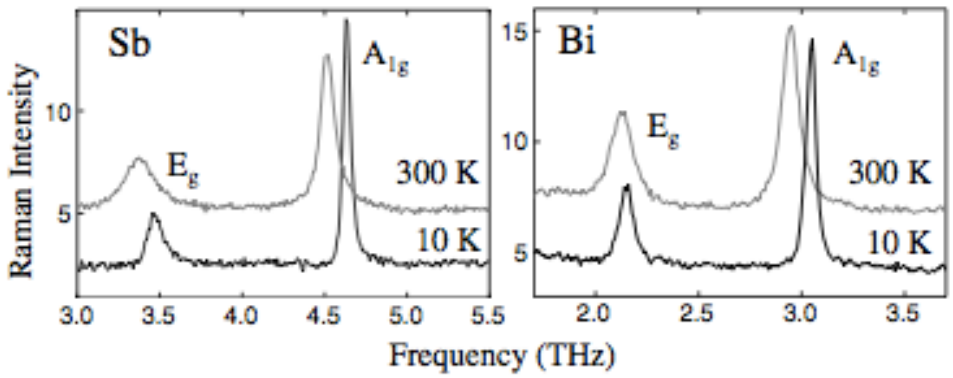


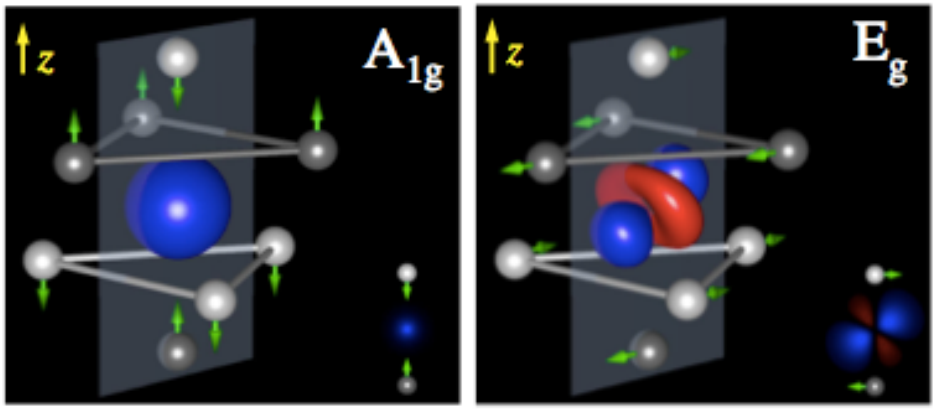
FIG. 3. (a) The pump-induced coherent phonon amplitude produces a $\Delta R/R$ as large as 12% initially. This plot convolved with the optical pulse intensity profile yields the least-squares fit to the data in Fig. 2. The fitting function is taken to be an exponentially damped cosine, superimposed on an exponentially decaying background (Ref. 6). (b) The coherent phonon frequency is initially down-shifted by 7%, but subsequently decays back to the 7.0 THz Raman frequency. This plot was obtained by fitting 0.5 ps blocks of the data from Fig. 2 to a decaying sinusoid, superimposed on an exponentially decaying background.

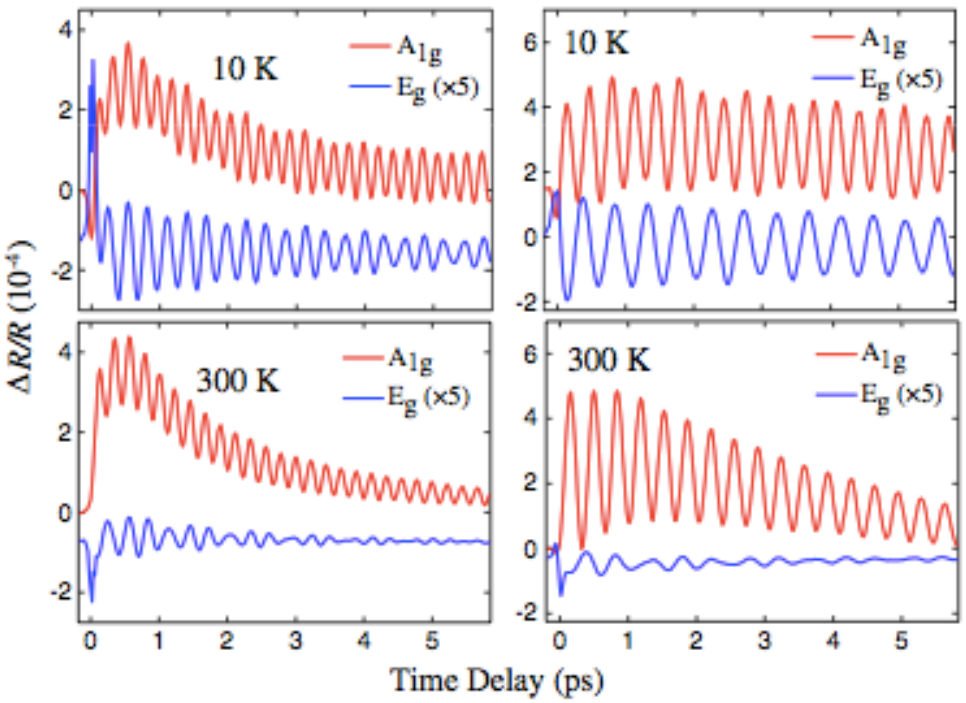
Chen et al., Appl. Phys. Lett. **62**, 1901 (1993)

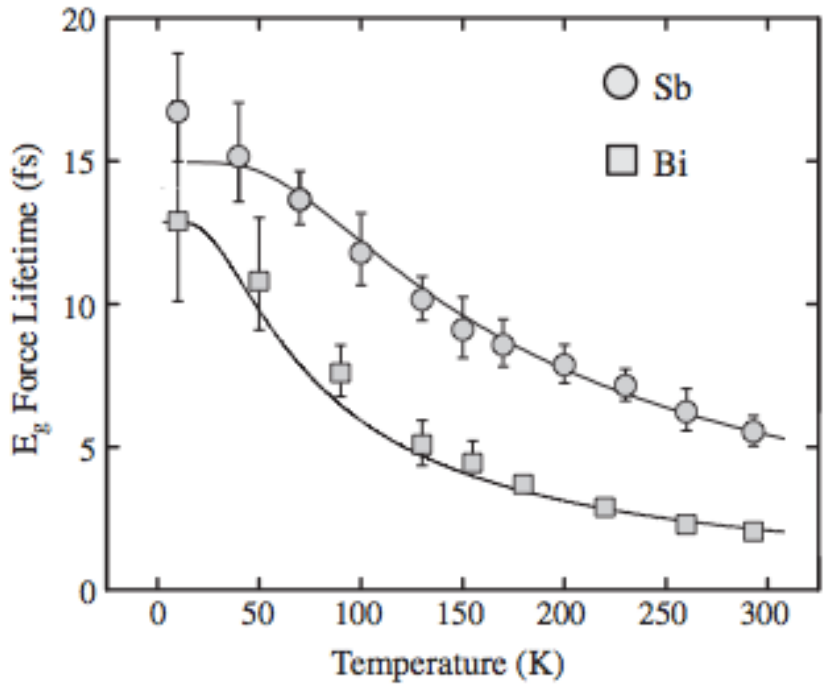
Excitation has coherent and incoherent contribution

Comparison of spontaneous and pump-probe yield femtosecond decay of Raman coherences (Force)









Li *et al., Phys Rev. Lett.* ***110****, 047401 (2013)*

Squeezed Phonons, q=0 excitation and measurement

S. L. Johnson et al., Phys. Rev. Lett. **102**, 175563 (2009)

Vacuum Squeezing of Solids: Macroscopic Quantum States Driven by Light Pulses

G. A. Garrett, A. G. Rojo, A. K. Sood,* J. F. Whitaker, R. Merlin†

Femtosecond laser pulses and coherent two-phonon Raman scattering were used to excite $KTaO_3$ into a squeezed state, nearly periodic in time, in which the variance of the atomic displacements dips below the standard quantum limit for half of a cycle. This nonclassical state involves a continuum of transverse acoustic modes that leads to oscillations in the refractive index associated with the frequency of a van Hove singularity in the phonon density of states.

Squeezing refers to a class of quantum mechanical states of the electromagnetic field and, more generally, of harmonic oscillators for which the fluctuations in two conjugate variables oscillate out of phase and become alternatively squeezed below the values for the vacuum state for some fraction of a cycle (1). Thus, a squeezed electromagnetic field provides a way for experimental measurements to overcome the standard quantum

crystal, $KTaO_3$, with an ultrafast pulse of light. The measurements were performed with the standard pump-probe setup (Fig. 1). Second-order coupling of the photons with the lattice vibrations [specifically, transverse acoustic (TA) modes] amounts to an impulsive change in the phonon frequency that gives rise to squeezing; this mechanism is closely related to that used to generate two-photon coherent states in

Fig. 2. (A) $\Delta P = \langle P^2(t)\rangle^{1/2}$ versus $\Delta Q = \langle Q^2(t)\rangle^{1/2}$ in units of $(\hbar\Omega/2)^{1/2}$ and $(\hbar/2\Omega)^{1/2}$, respectively. Dots denote values immediately before ($t = 0^-$) and after ($t = 0^+$) the pulse is applied. The circular arc is the trajectory for $t > 0$. Shaded regions represent quadrature-squeezed states. Minimum-uncertainty states lie on the hyperbola (dashed curve). (B) Classical phase-space diagrams showing the circular ($t < 0$) and elliptical ($t > 0$) noise distributions.

Fig. 3. (A) Normalized transmitted intensity of the probe pulse as a function of the delay for the A_{1g}-symmetry configuration. (B) Fourier transform of the time-domain data. (C) Weighted second-order Raman cross section $\mathcal{H}(\Omega)\exp(-\Omega^2\tau_p^2/2)/C(2\Omega)$ obtained at 810.0 nm.

Fig. 4. Experimental time dependence of the squeezing factor $\mathcal{F} = 1 - \langle \mathcal{R}^2(t)\rangle/\langle \mathcal{R}^2(0)\rangle^{1/2}$ at integrated pulse intensity $I_0 = 19\ \mu J/cm^2$. (Inset) Amplitude of $\mathcal{F}$ as a function of I_0.

Garrett, Science **275**, 1638 (1997)

Adapted from R. Merlin.

Selected for a Viewpoint in *Physics*
PRL 102, 175503 (2009) PHYSICAL REVIEW LETTERS week ending 1 MAY 2009

Directly Observing Squeezed Phonon States with Femtosecond X-Ray Diffraction

S. L. Johnson,[1,*] P. Beaud,[1] E. Vorobeva,[1] C. J. Milne,[2] É. D. Murray,[3] S. Fahy,[4] and G. Ingold[1]
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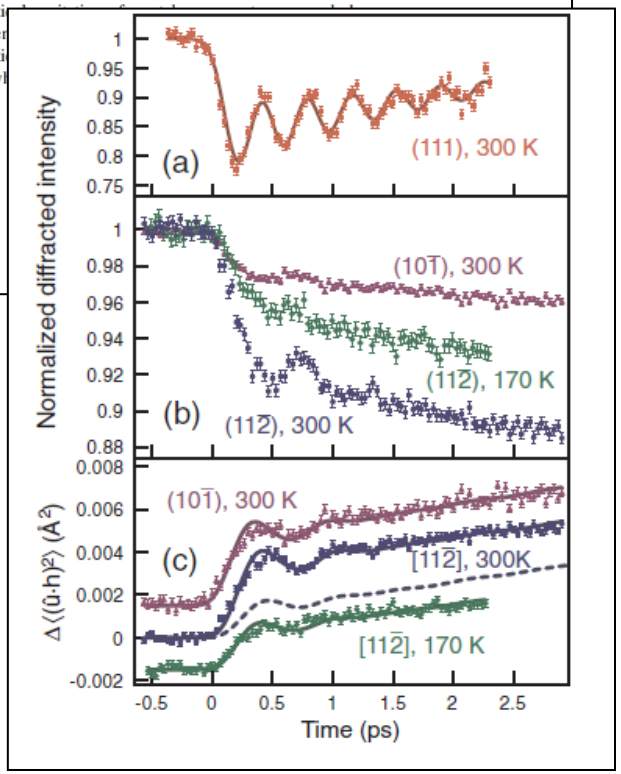
[2]*Laboratoire de Spectroscopie Ultrarapide, Ecole Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland*
[3]*Department of Physics and Astronomy, Rutgers University, Piscataway, New Jersey 08854-8019, USA*
[4]*Tyndall National Institute and Department of Physics, University College, Cork, Ireland*
(Received 13 December 2008; published 27 April 2009)

Squeezed states are quantum states of a harmonic oscillator in which the variance of two conjugate variables each oscillate out of phase. Ultrafast opti
states, where the variance of the atomic displacemen
bonding strength. With femtosecond x-ray diffracti
conclude that they are consistent with a model in wl
a constant scaling factor.

DOI: 10.1103/PhysRevLett.102.175503

Squeezed states, where the variance of two conjugate variables oscillate out of phase in time, are a centerpiece of modern quantum optics [1,2]. The physics of squeezing is not limited to the electromagnetic field, however, and indeed there has been much interest in the squeezing of



$KTaO_3$

Bi

Johnson et al., Phys. Rev. Lett **102**, 175503(2009)

X-ray Scattering and connection with elementary excitations

Inelastic X-ray (and Neutron) Scattering

PHYSICAL REVIEW VOLUME 95, NUMBER 1 JULY 1, 1954

Correlations in Space and Time and Born Approximation Scattering in Systems of Interacting Particles

LÉON VAN HOVE
Institute for Advanced Study, Princeton, New Jersey
(Received March 16, 1954)

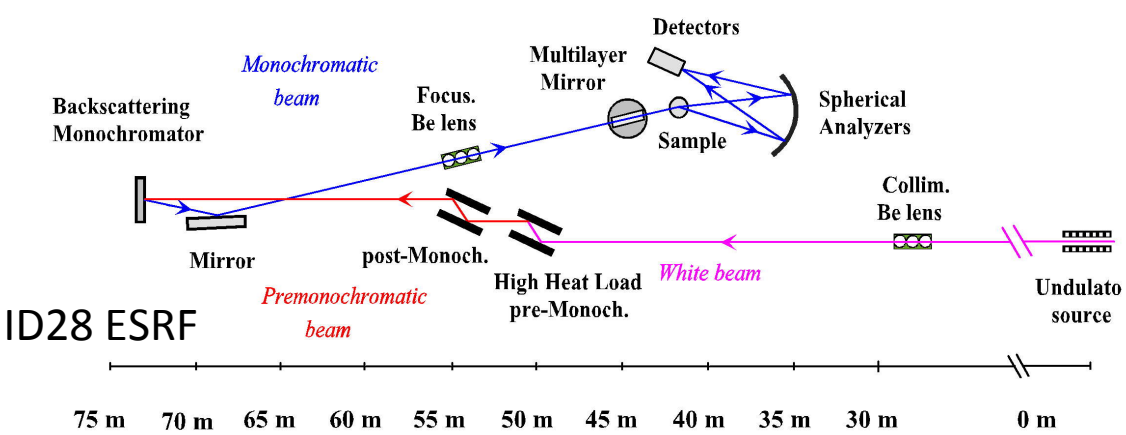
$$\frac{d^2\sigma}{d\Omega d\epsilon} = A\,\mathcal{S}(\boldsymbol{\kappa},\omega),$$

$$\mathcal{S}(\boldsymbol{\kappa},\omega) = (2\pi)^{-1}N\int \exp[i(\boldsymbol{\kappa}\cdot\mathbf{r}-\omega t)]\cdot G(\mathbf{r},t)d\mathbf{r}dt,$$

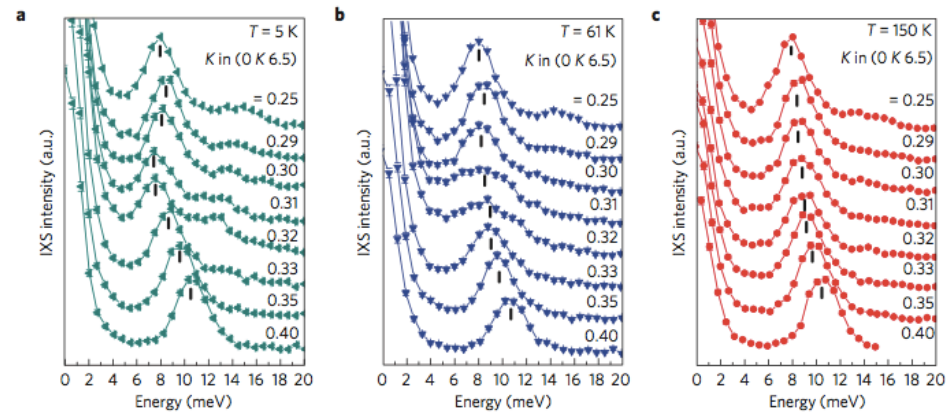
$$G(\mathbf{r},t) = (2\pi)^{-3}N^{-1}\sum_{l,j=1}^{N}\int d\boldsymbol{\kappa}\exp(-i\boldsymbol{\kappa}\cdot\mathbf{r})$$
$$\cdot\langle\exp\{-i\boldsymbol{\kappa}\cdot\mathbf{r}_l(0)\}\cdot\exp\{i\boldsymbol{\kappa}\cdot\mathbf{r}_j(t)\}\rangle.$$

S(Q,ω) Related to the imaginary part of density-density response function

Inelastic X-ray Scattering: $S(\vec{Q},\omega) \propto \sum_j \int \mathrm{d}t e^{i\omega t} \langle u_{j,\vec{Q}}(0) u_{j,-\vec{Q}}(t) \rangle$

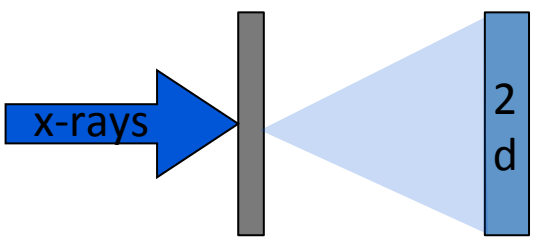


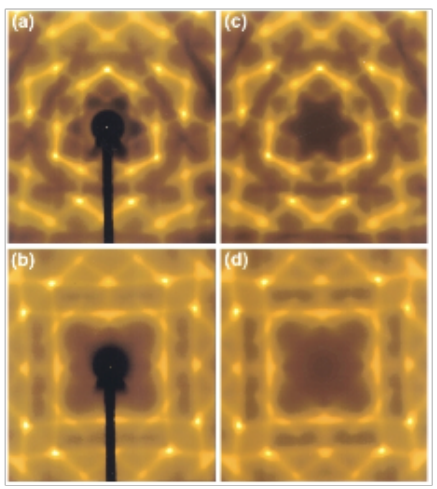
Underdoped YBCO



M. Le Tacon et. al, Nat. Phys. 10,52 (2014)

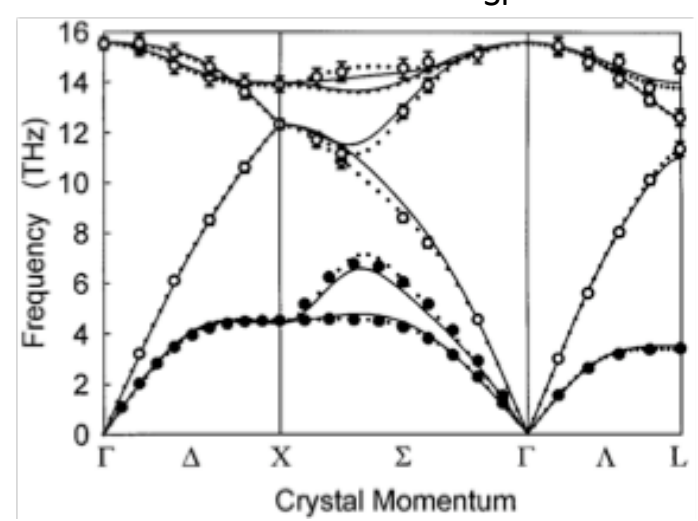
X-ray Diffuse Scattering: $S(\vec{Q}) \propto \sum_j \langle u_{j,\vec{Q}}(0) u_{j,-\vec{Q}}(0) \rangle$





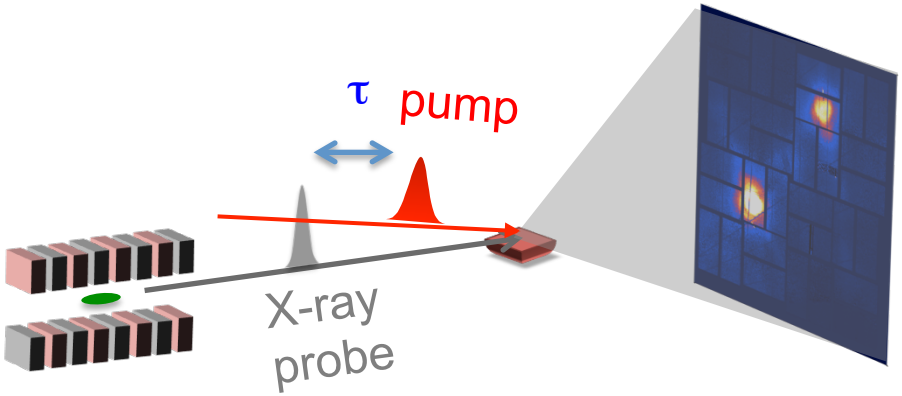
fit to Bose-Einstein distribution.

Si



M. Holt et al., PRL 83 (1999).

Time and momentum-domain x-ray scattering

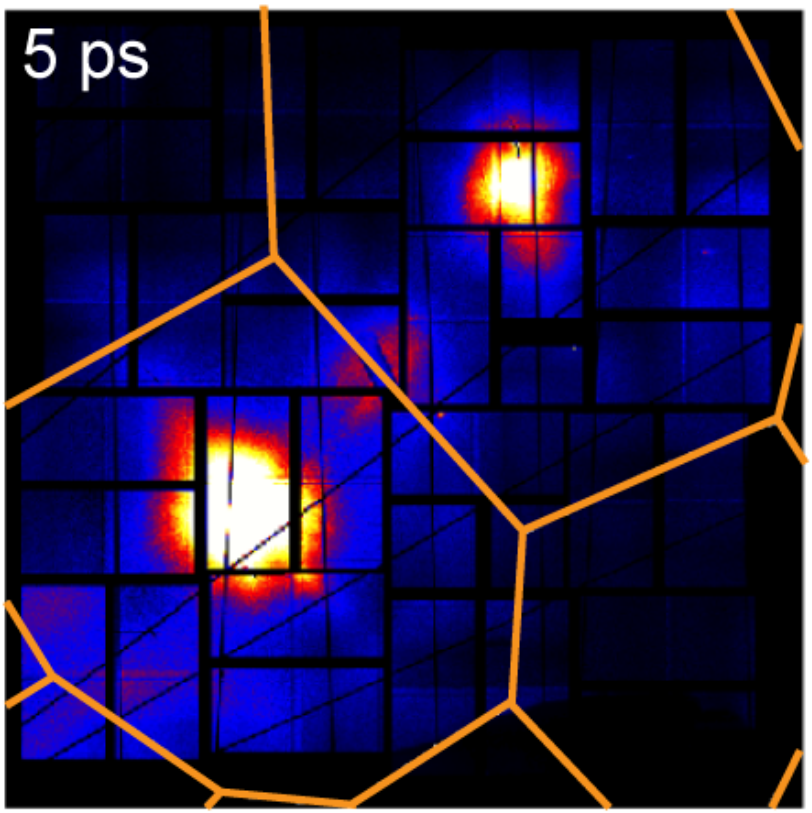


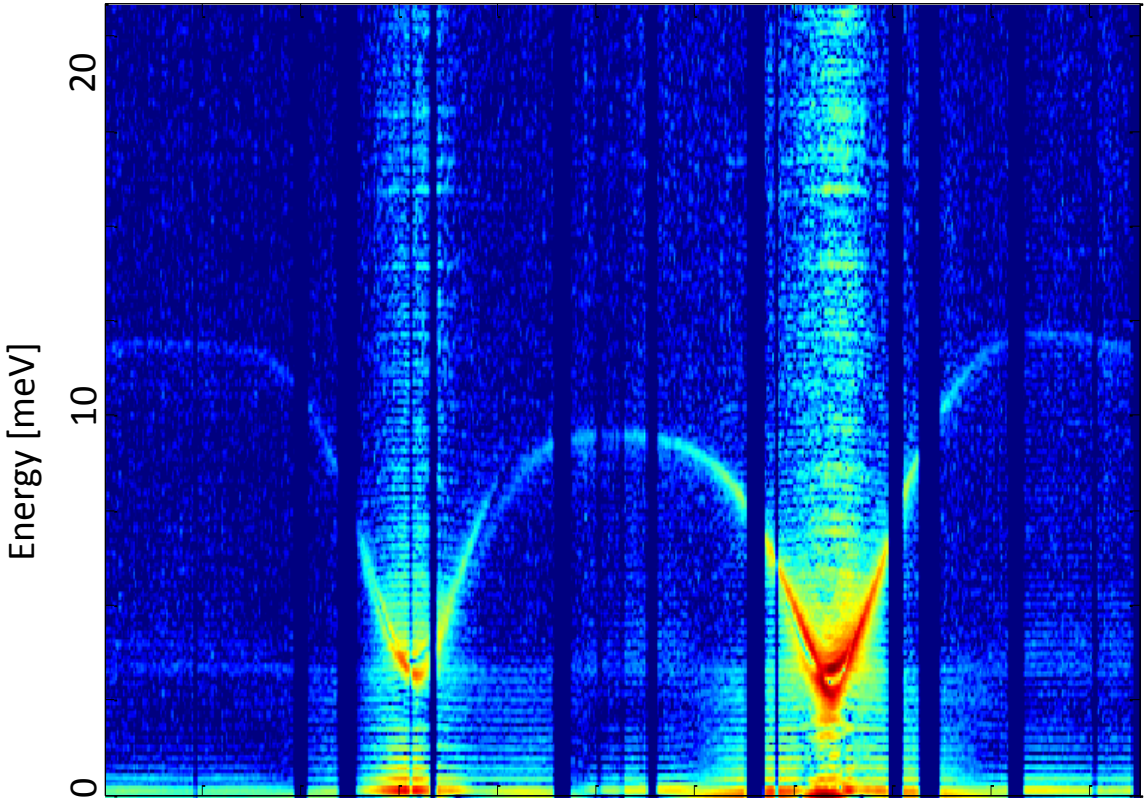
$$S(\vec{Q};\tau) \propto \sum_{j,j'} \langle u_{j,\vec{Q}}(\tau) u_{j',-\vec{Q}}(\tau) \rangle$$

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

Non-equillibrium populations

Non-equillibrium frequency (forces)





phonon-phonon interactions

Electron-phonon interactions

Classical Picture of X-ray Scattering from Phonons

$$\vec{u}_n = \vec{u}\cos(\Omega t + \delta)\cos(\vec{q}\cdot\vec{R}_n)$$

$$z_n = z(t) = \vec{K}\cdot\vec{u}\cos(\Omega t + \delta)$$

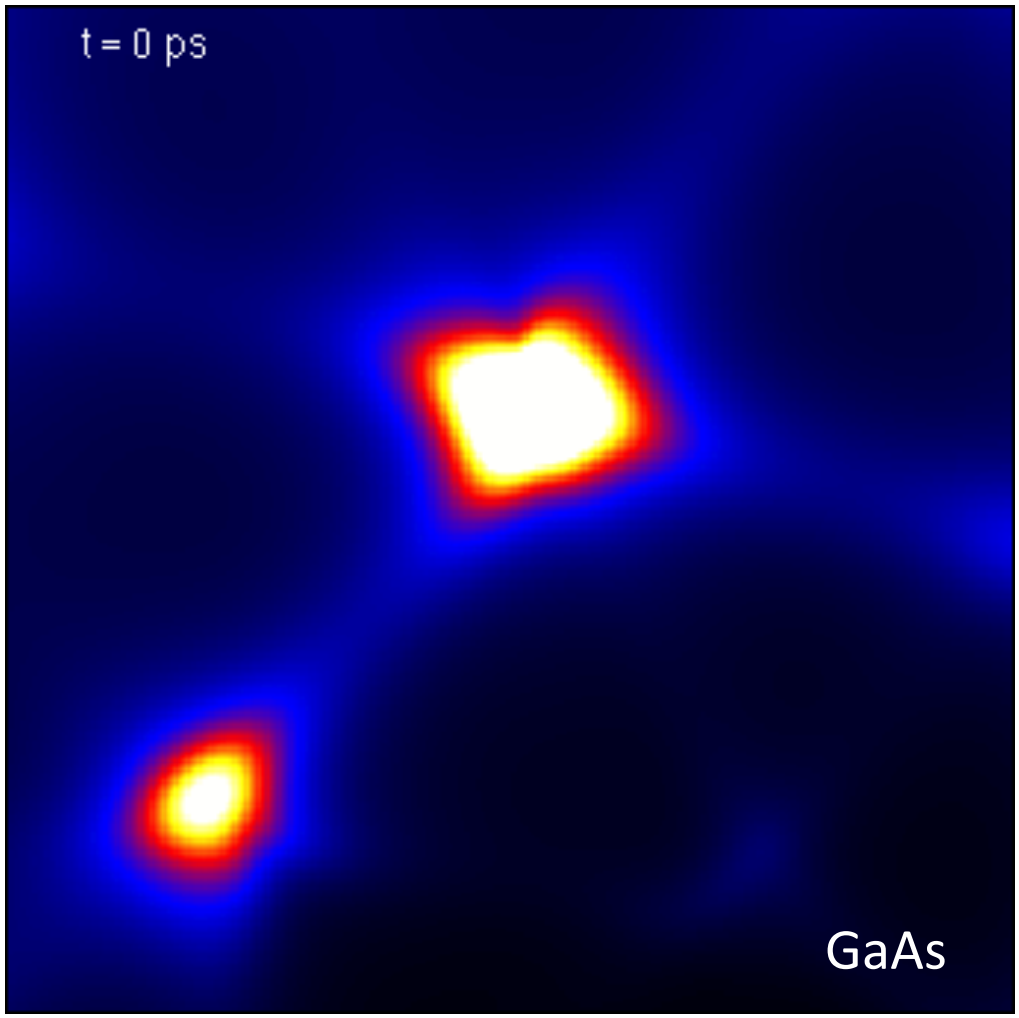
$$I(\vec{K},t) = I_e|f|^2 \boxed{\mathrm{e}^{-\frac{1}{2}z^2(t)}}\Big($$

$$\boxed{N^2\delta(\vec{K}-\vec{G})+}$$

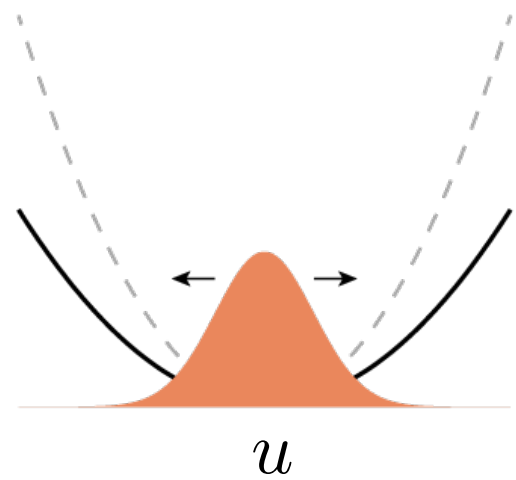
$$\boxed{N^2 z^2(t)\delta(\vec{K}-\vec{G}\pm\vec{q})}+$$

$$+Nz(t))\delta(\vec{K}-\vec{G}\pm\vec{q})\Big) + \ldots$$

Change in forces induces temporal coherence



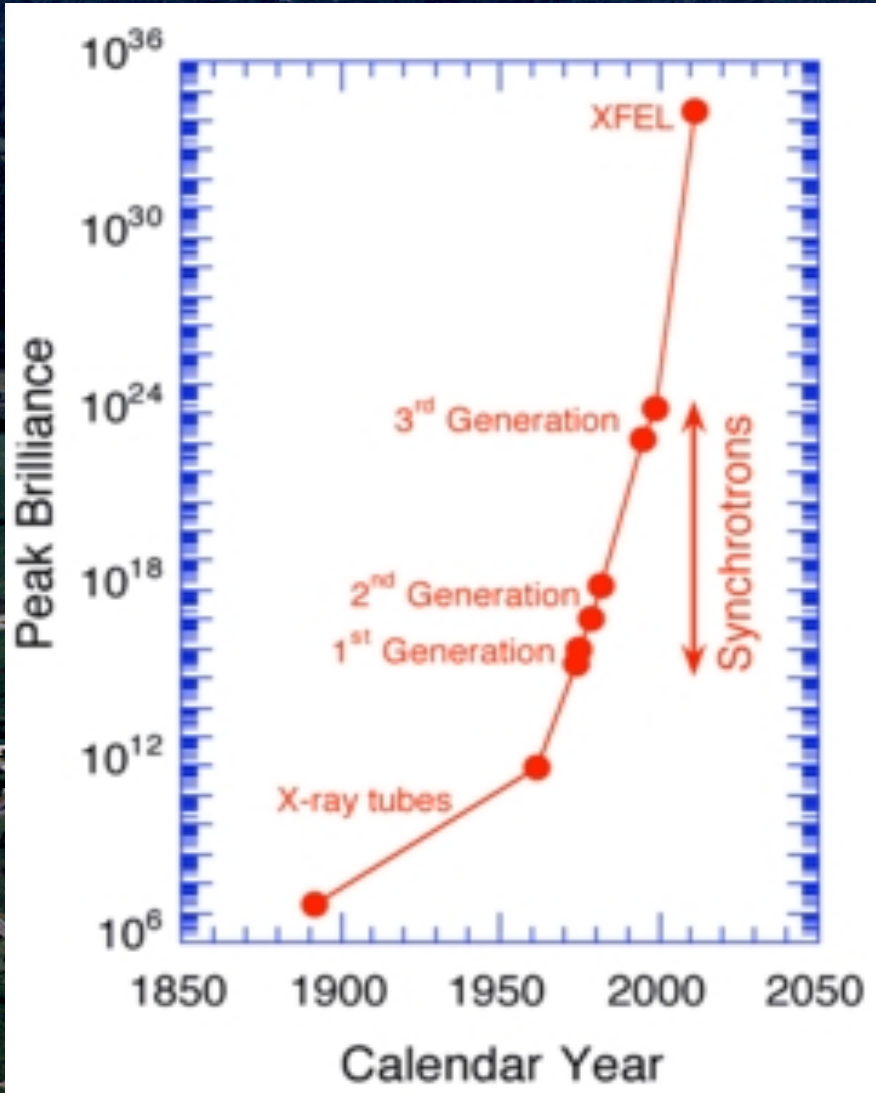
$$\left\langle (\vec{Q} \cdot u_{-\vec{q}}(t))(\vec{Q} \cdot u_{\vec{q}}(t)) \right\rangle$$



Sudden Softening

squeezed thermal vibrations

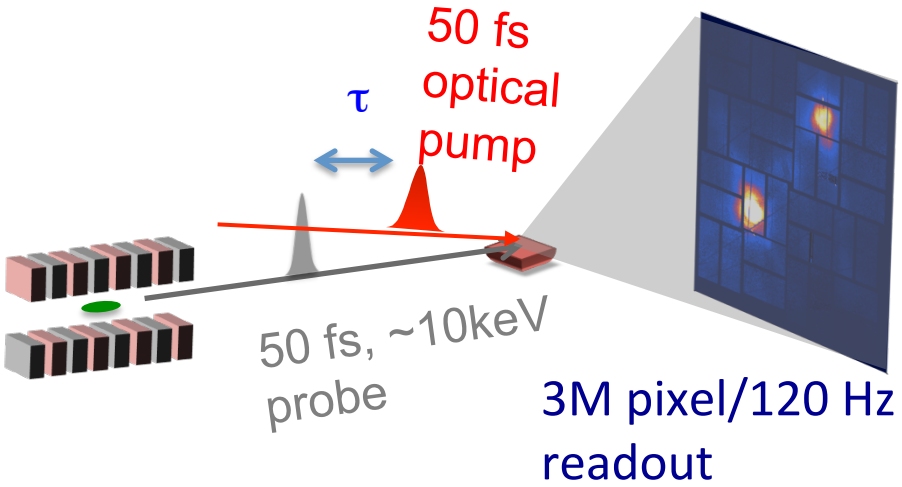
A new era of hard x-ray sources



http://www.psi.ch/swissfel/why-swissfel

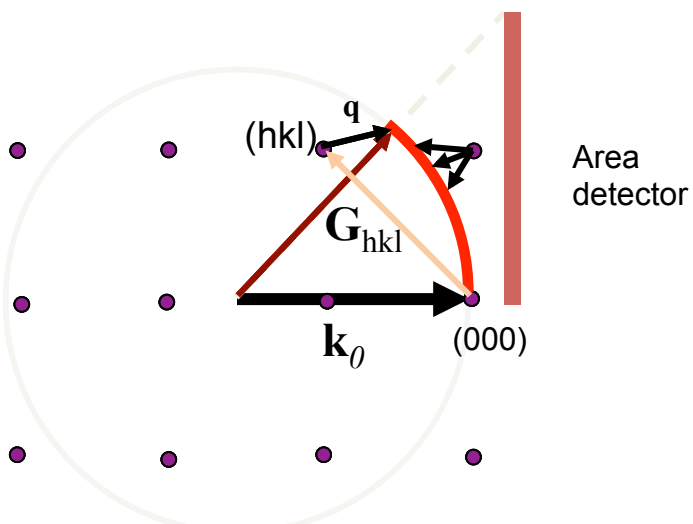
x-ray free-electron lasers (version 1):

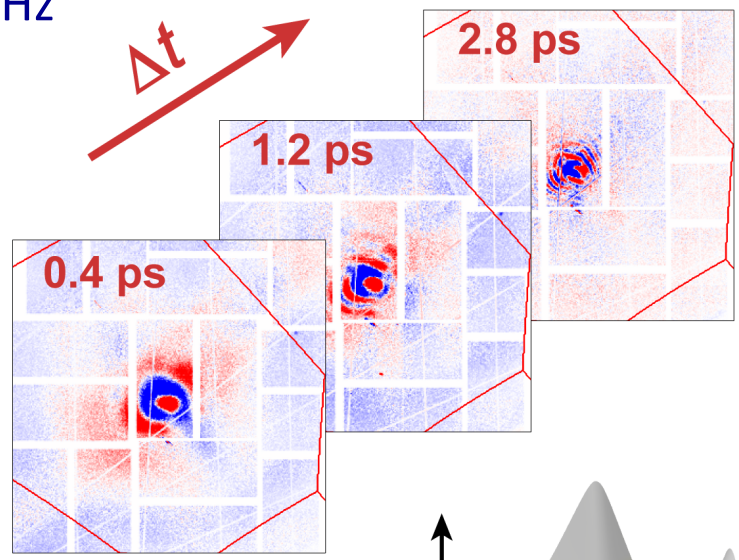
Time and momentum-domain x-ray scattering:

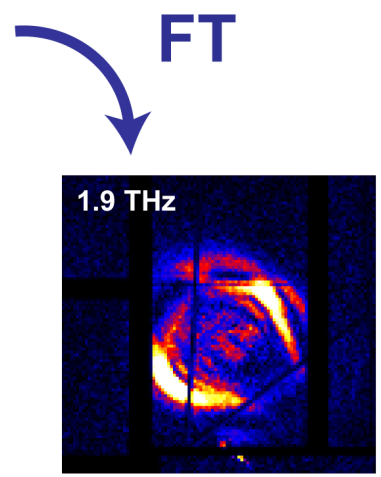


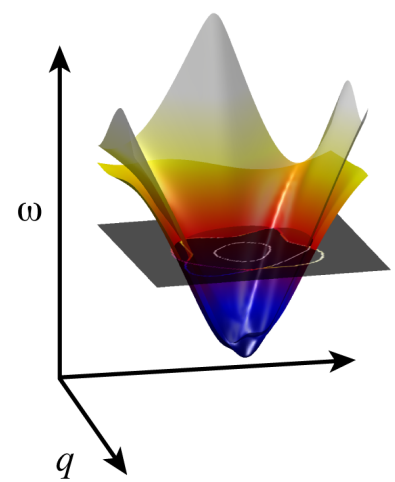
$$S(\vec{Q};\tau) \propto \sum_{j,j'} \langle u_{j,\vec{Q}}(\tau) u_{j',-\vec{Q}}(\tau) \rangle$$

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





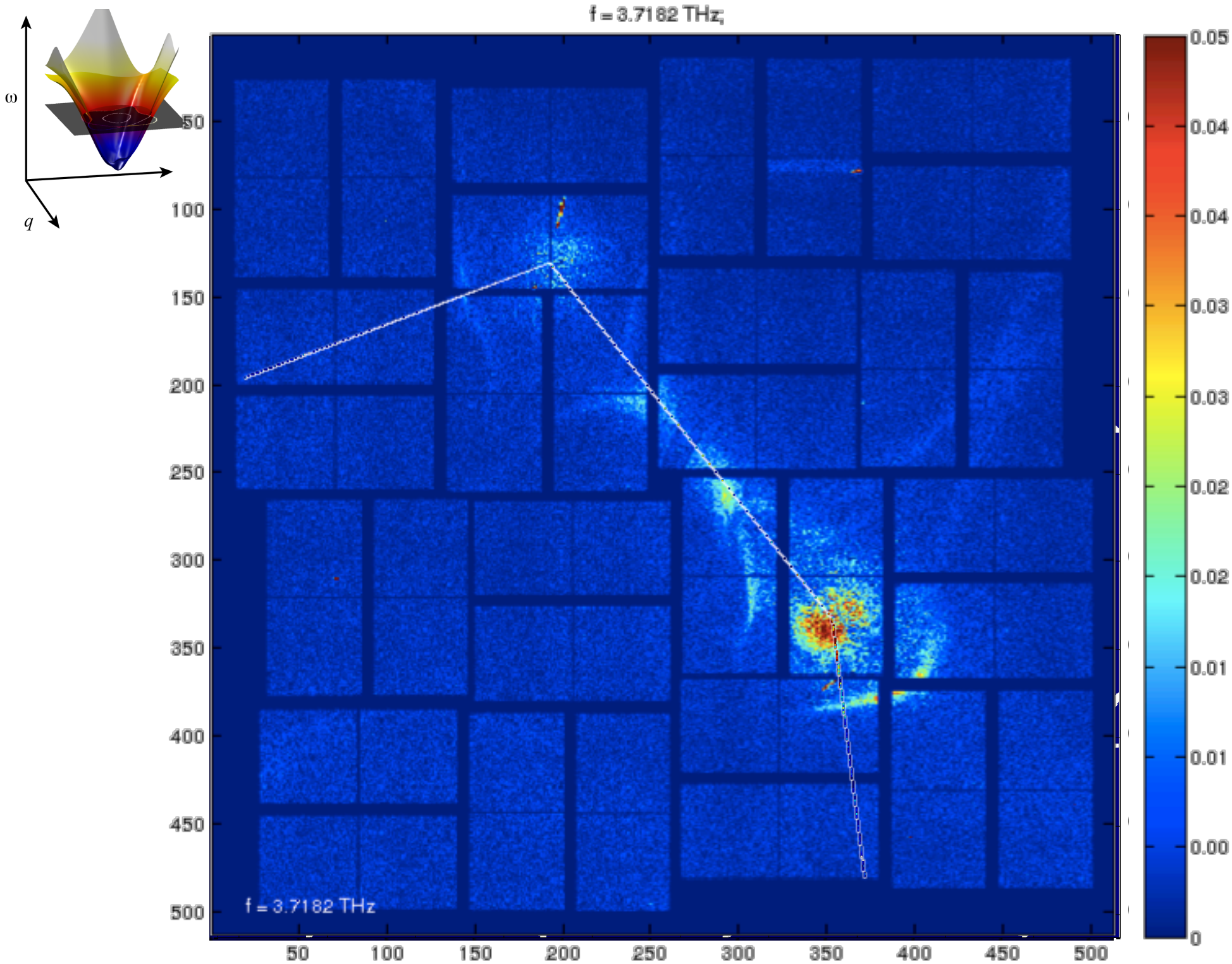




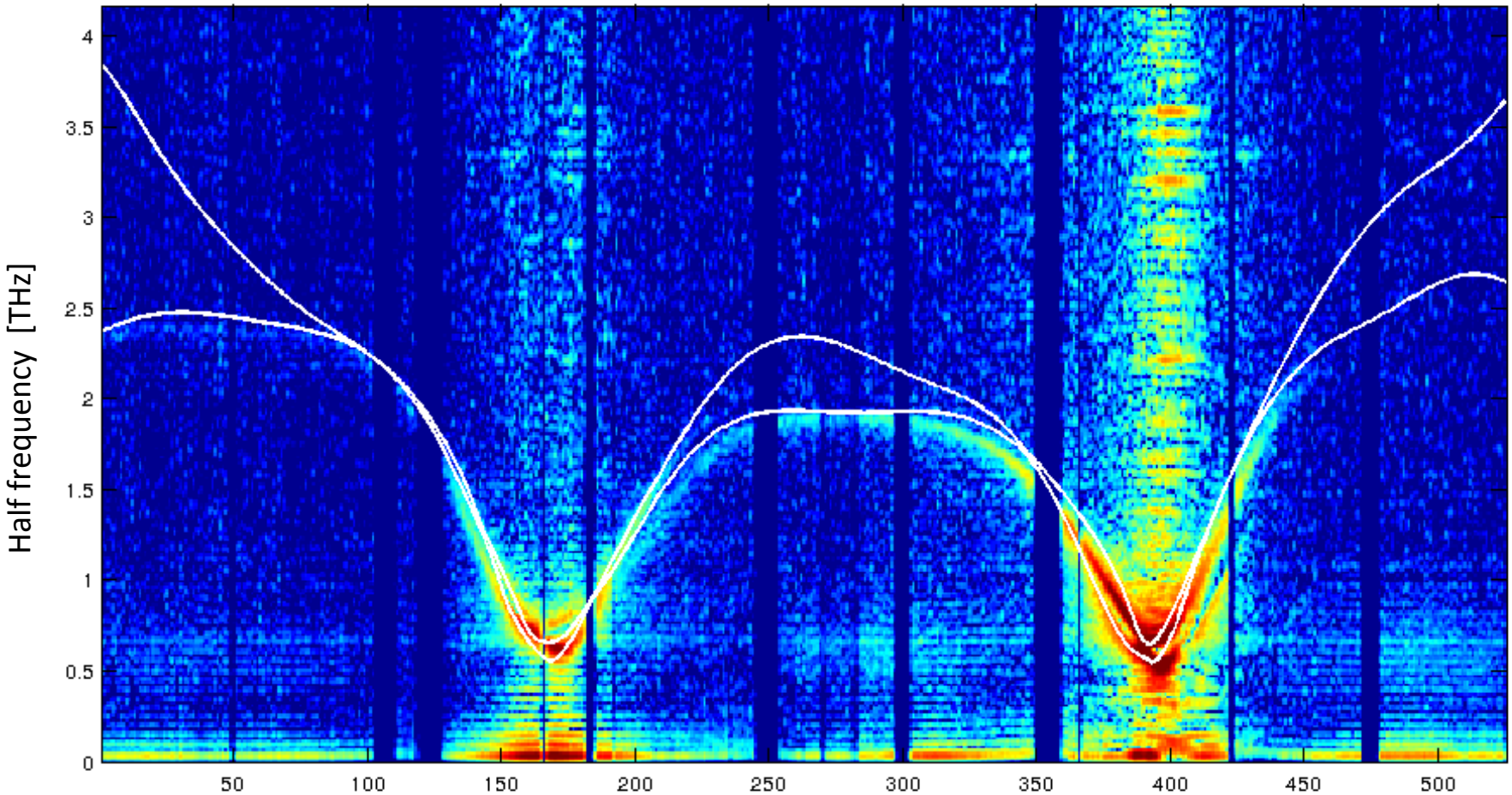
Femtosecond time domain diffuse images:

t = -2 ps

Ge, differential signal after high-pass filter (for emphasis)


f = 3.7182 THz
ω
q
f = 3.7182 THz
50
100
150
200
250
300
350
400
450
500
0.05
0.04
0.04
0.03
0.03
0.02
0.02
0.01
0.01
0.00
0

extracted TA phonon dispersion (Ge)



independent modes (oscillation at twice frequency):

$$\langle u_q u_{-q} \rangle = \frac{1}{4m\omega_q}\left(\left(1+\frac{\omega_q^2}{\omega_q^{2\prime}}\right)+\left(1-\frac{\omega_q^{2\prime}}{\omega_q^2}\right)\cos(2\omega'\tau)\right)$$

Trigo, Zhu, et al. in preparation

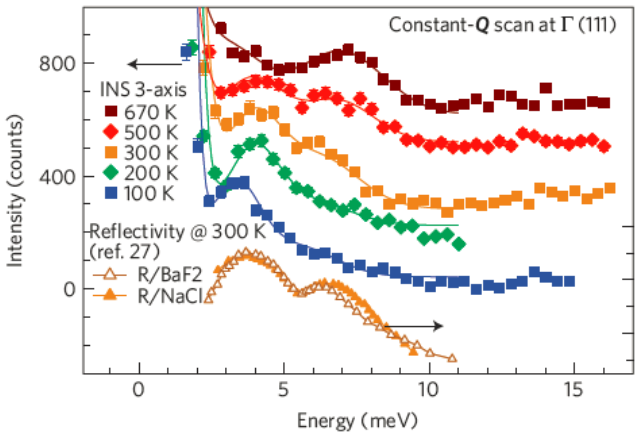
Trigo et al. Nature Physics. 9, 790, 2013

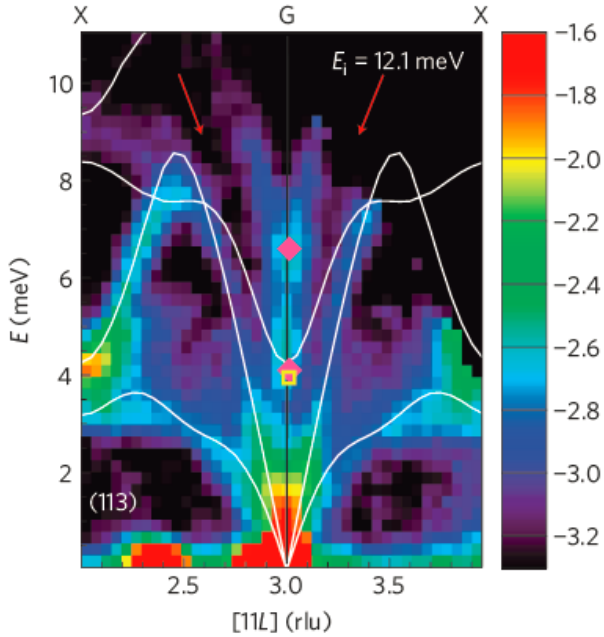
Lattice dynamics of PbTe

ARTICLES
PUBLISHED ONLINE: 5 JUNE 2011 | DOI: 10.1038/NMAT3035
nature materials

Giant anharmonic phonon scattering in PbTe

O. Delaire[1]*, J. Ma[1], K. Marty[1], A. F. May[2], M. A. McGuire[2], M-H. Du[2], D. J. Singh[2], A. Podlesnyak[1], G. Ehlers[1], M. D. Lumsden[1] and B. C. Sales[2]

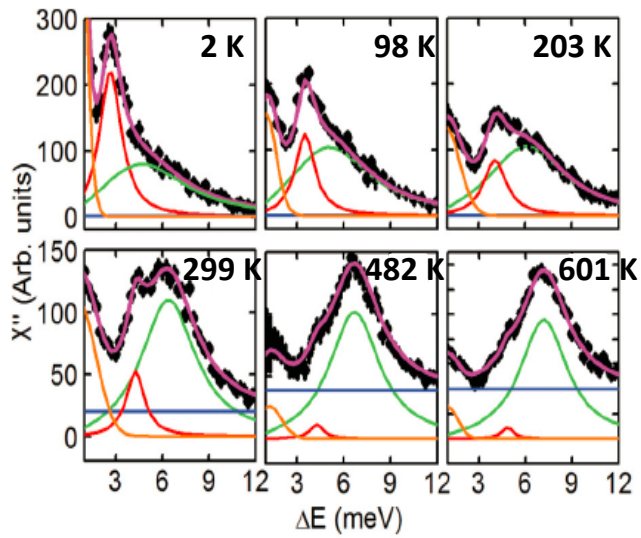


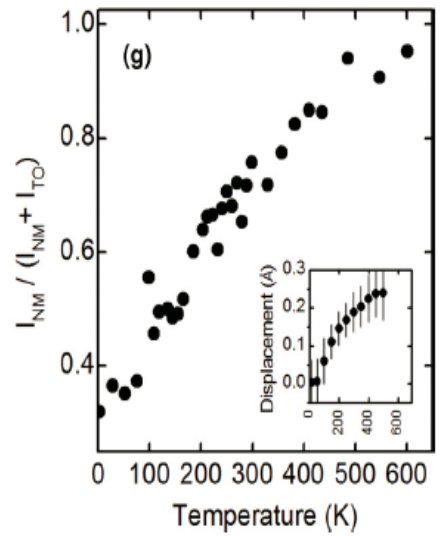


PHYSICAL REVIEW B 86, 085313 (2012)

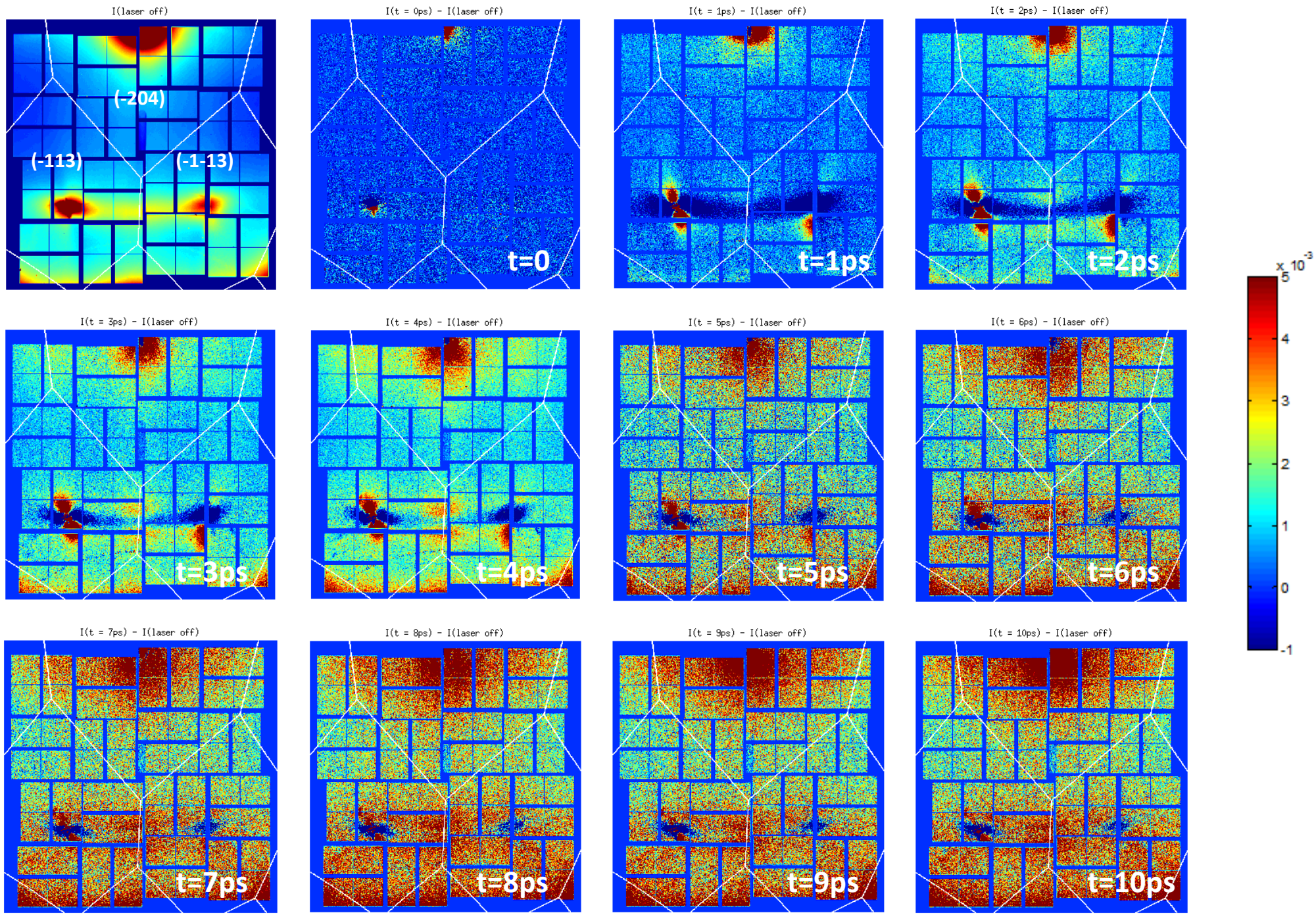
Lattice dynamics reveals a local symmetry breaking in the emergent dipole phase of PbTe

Kirsten M. Ø. Jensen,[1] Emil S. Božin,[2] Christos D. Malliakas,[3] Matthew B. Stone,[4] Mark D. Lumsden,[4] Mercouri G. Kanatzidis,[3,5] Stephen M. Shapiro,[2] and Simon J. L. Billinge[2,6]



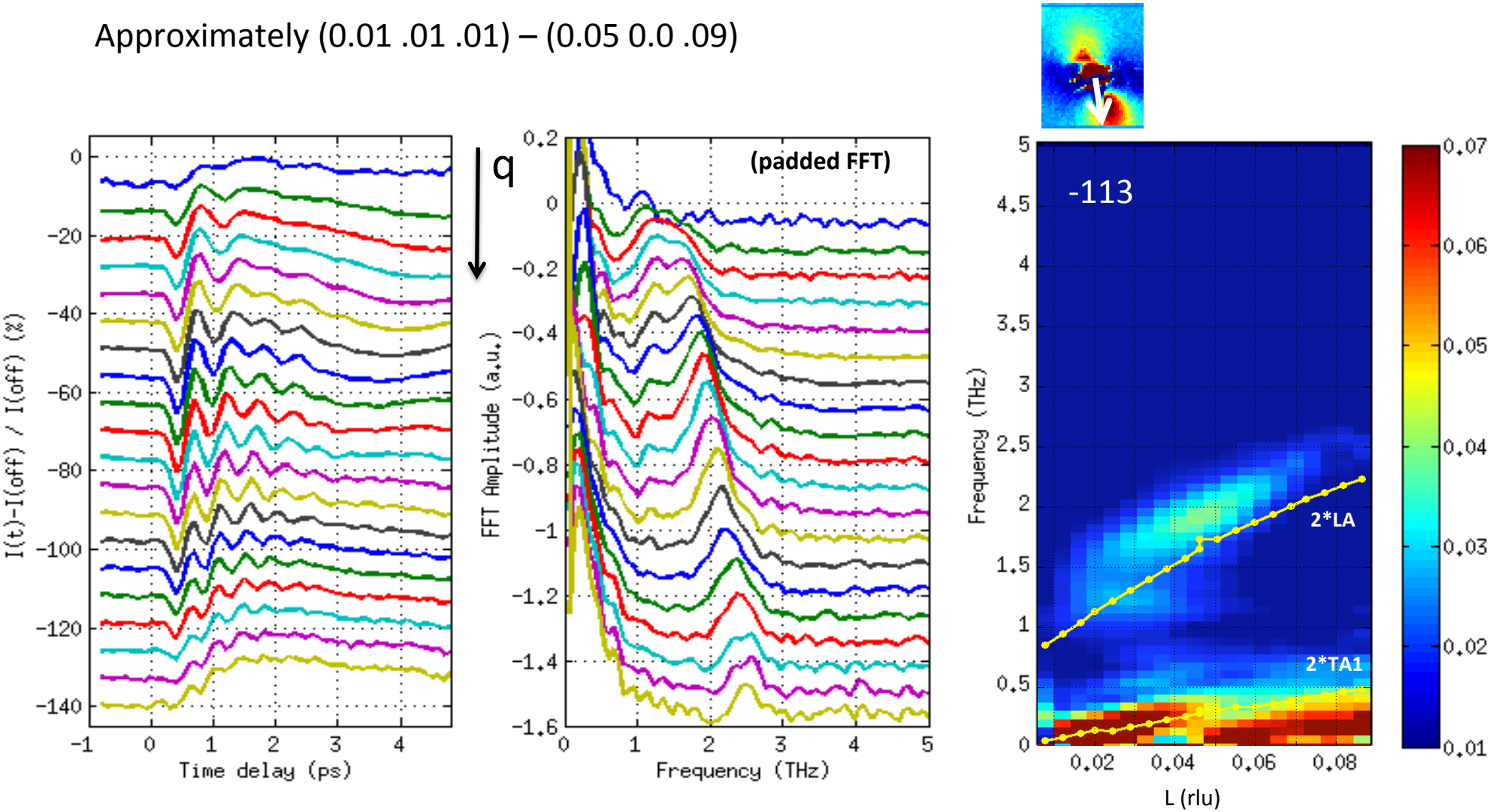


PbTe differential scattering (2 μm pump, 1.4 Å probe)



PbTe near zone-center 100 fs steps (re-binned)

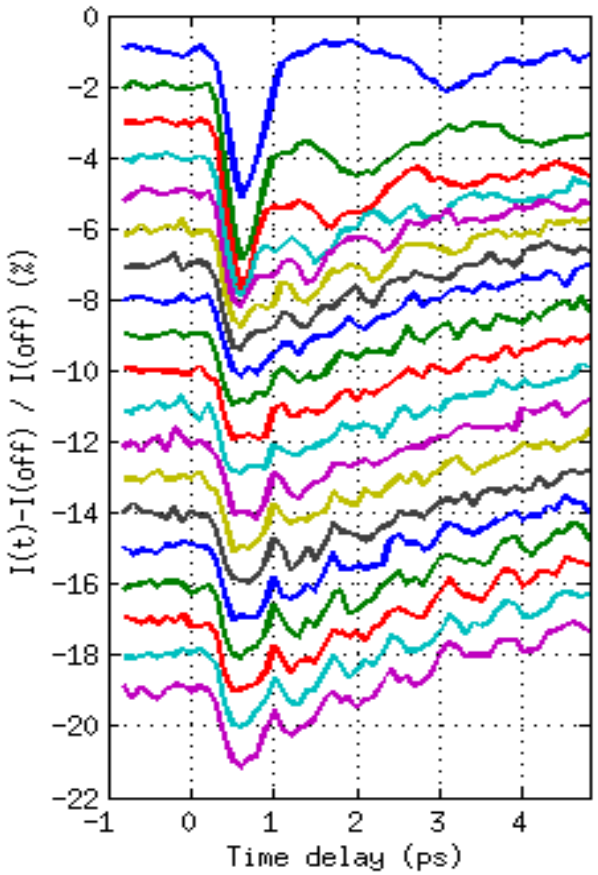
Approximately (0.01 .01 .01) – (0.05 0.0 .09)



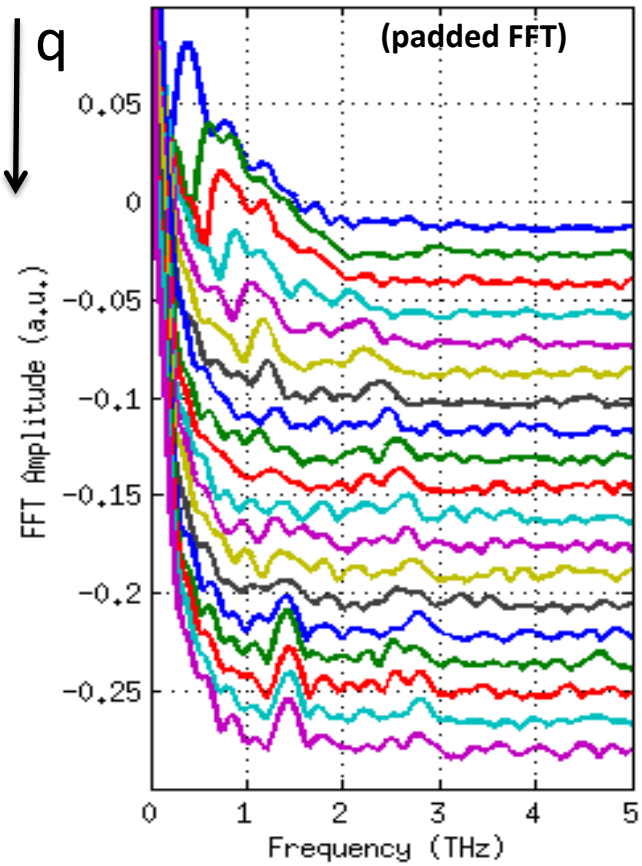
M.P. Jiang et al., in preparation

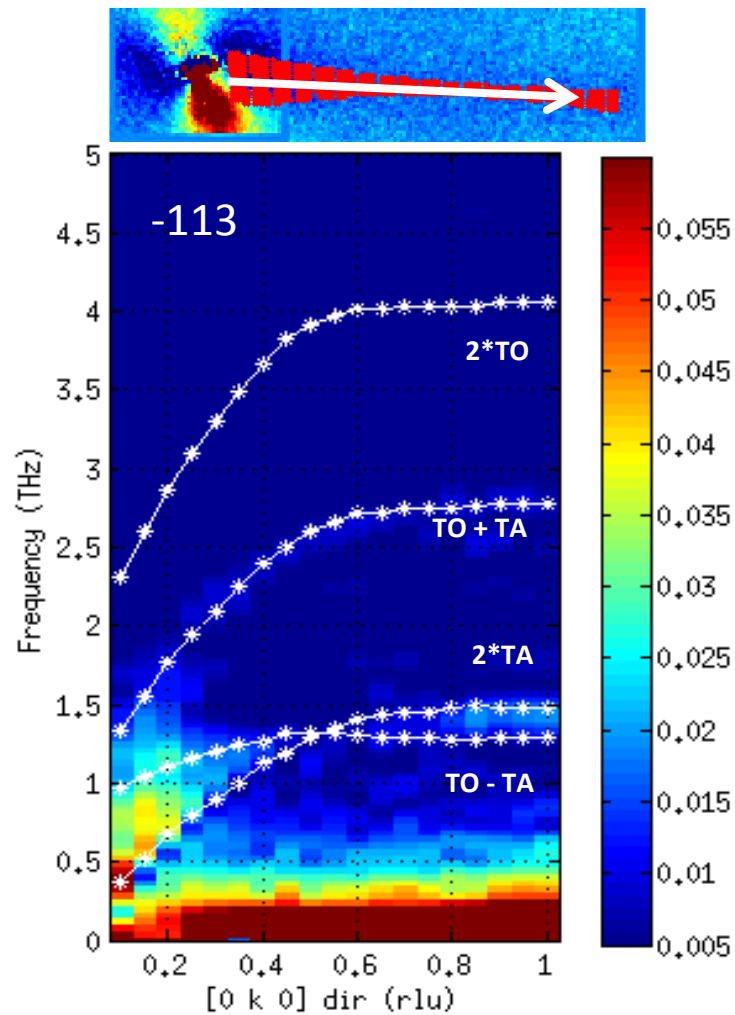
PbTe (Γ to X) 100 fs steps (re-binned)

Near Γ [0 0.1 0] to X [0 1 0] in 0.05 steps
(with tolerance levels +/- 0.03)



q





Identification based on Cohran,
Proc. R. Soc. Lond. A **293**: 433 (1966)

M.P. Jiang et al., in preparation

Comments on FT-IXS

- Femtosecond x-ray scattering from large wave-vector phonon pairs excited by long-wavelength laser.
  - harmonic crystal (Ge): coherences in $\langle x_{TA,q} x_{TA,-q} \rangle$ at $2\omega_q$
  - anharmonic crystal (PbTe): coherences in $\langle x_{j,q} x_{j',-q} \rangle$ two-phonon combination modes ($j \neq j'$ as well as $j=j$).
- Resolution limited by maximum delay (sub-meV demonstrated). Range limited by time-resolution (sub-80 fs demonstrated)
- Broad-band x rays, no crystal analyzers and parallel detection of momentum transfer.
- Applicable near and far from equilibrium.

Acknowledgements (Ge expt.)

LETTERS
PUBLISHED ONLINE: 27 OCTOBER 2013 | DOI: 10.1038/NPHYS2788

nature physics

Fourier-transform inelastic X-ray scattering from time- and momentum-dependent phonon–phonon correlations

M. Trigo[1,2]*, M. Fuchs[1,2], J. Chen[1,2], M. P. Jiang[1,2], M. Cammarata[3], S. Fahy[4], D. M. Fritz[3], K. Gaffney[2], S. Ghimire[2], A. Higginbotham[5], S. L. Johnson[6], M. E. Kozina[2], J. Larsson[7], H. Lemke[3], A. M. Lindenberg[1,2,8], G. Ndabashimiye[2], F. Quirin[9], K. Sokolowski-Tinten[9], C. Uher[10], G. Wang[10], J. S. Wark[5], D. Zhu[3] and D. A. Reis[1,2,11]*

[1]Stanford Institute for Materials and Energy Sciences, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA, [2]Stanford PULSE Institute, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA, [3]Linac Coherent Light Source, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA, [4]Tyndall National Institute and Department of Physics, University College, Cork, Ireland, [5]Department of Physics, Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, UK, [6]Physics Department, ETH Zurich, 8093 Zurich, Switzerland, [7]Department of Physics, Lund University, S-22100 Lund, Sweden, [8]Department of Materials Science and Engineering, Stanford University, Stanford, California 94305, USA, [9]Faculty of Physics and Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, 47048, Duisburg, Germany, [10]Department of Physics, University of Michigan, Ann Arbor, Michigan 48109, USA, [11]Departments of Photon Science and Applied Physics, Stanford University, Stanford, California 94305, USA. *e-mail: mtrigo@slac.stanford.edu; dreis@slac.stanford.edu

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Diling Zhu

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Andrew May
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Roberto Merlin

Takahiro SATO

Stephen Fahy
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Ivana Savic

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M. Trigo, et al., Fourier-transform inelastic x-ray scattering from time- and momentum-dependent phonon-phonon correlations. *Nature Physics*, 9,790 2013.

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