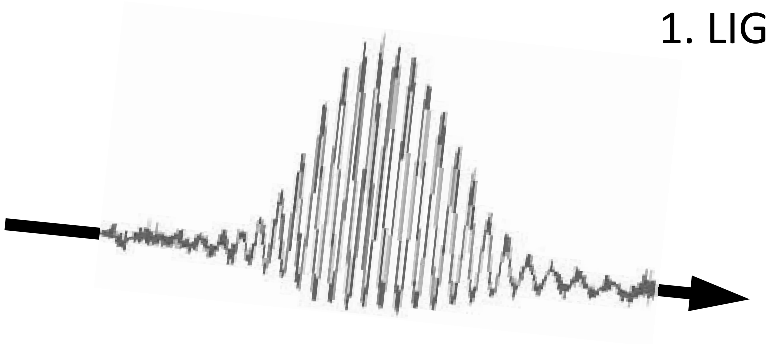
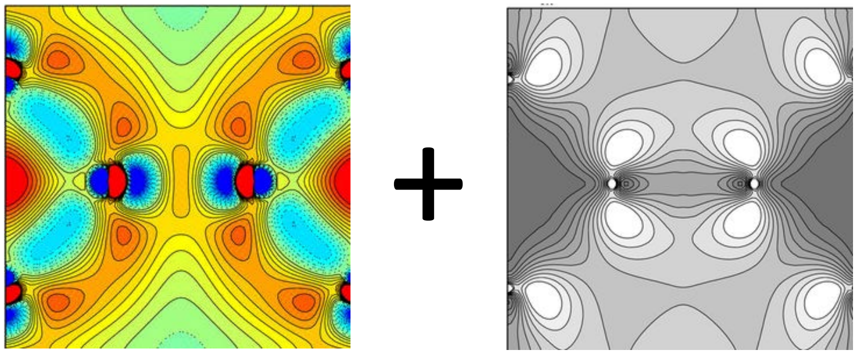
GENERATION OF VIBRATIONAL COHERENCES MEDIATED BY ELECTRONIC EXCITATIONS

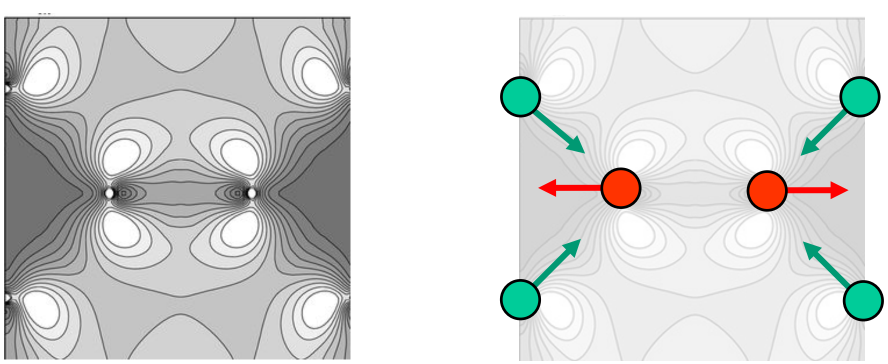


1. LIGHT CREATES A
CHARGE DENSITY FLUCTUATION $\propto |\mathbf{E}|^2$



$$\rho(\mathbf{r}) \quad + \quad \delta\rho(\mathbf{r},t)$$

2. ELECTRONIC CHARGE DENSITY FLUCTUATIONS
COUPLE TO AND DRIVE PHONONS OF THE SAME SYMMETRY



COUPLING MECHANISM

RAMAN-ACTIVE MODES ARE THE ONLY MODES THAT CAN CHANGE THE ELECTRON PERMITTIVITY !

THE TENSOR ε_{ij} TRANSFORMS LIKE THE PRODUCT OF TWO VECTORS

Q MUST TRANSFORMS LIKE THE PRODUCT OF TWO VECTORS (THUS, IT MUST BE RAMAN ACTIVE)

RAMAN SYMMETRIES ARE THE ONLY SYMMETRIES CHARGE-DENSITY FLUCTUATIONS CAN HAVE IF GENERATED BY LIGHT AND $\propto |\mathbf{E}|^2$!

RAMAN COUPLING TO PHONONS (TRANSPARENT MEDIA)

ELECTROMAGNETIC ENERGY DENSITY

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

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

⇩

CHANGE IN DIELECTRIC RESPONSE

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

⇩

CHANGE IN ENERGY DENSITY

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

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

⇩

FORCE DENSITY

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

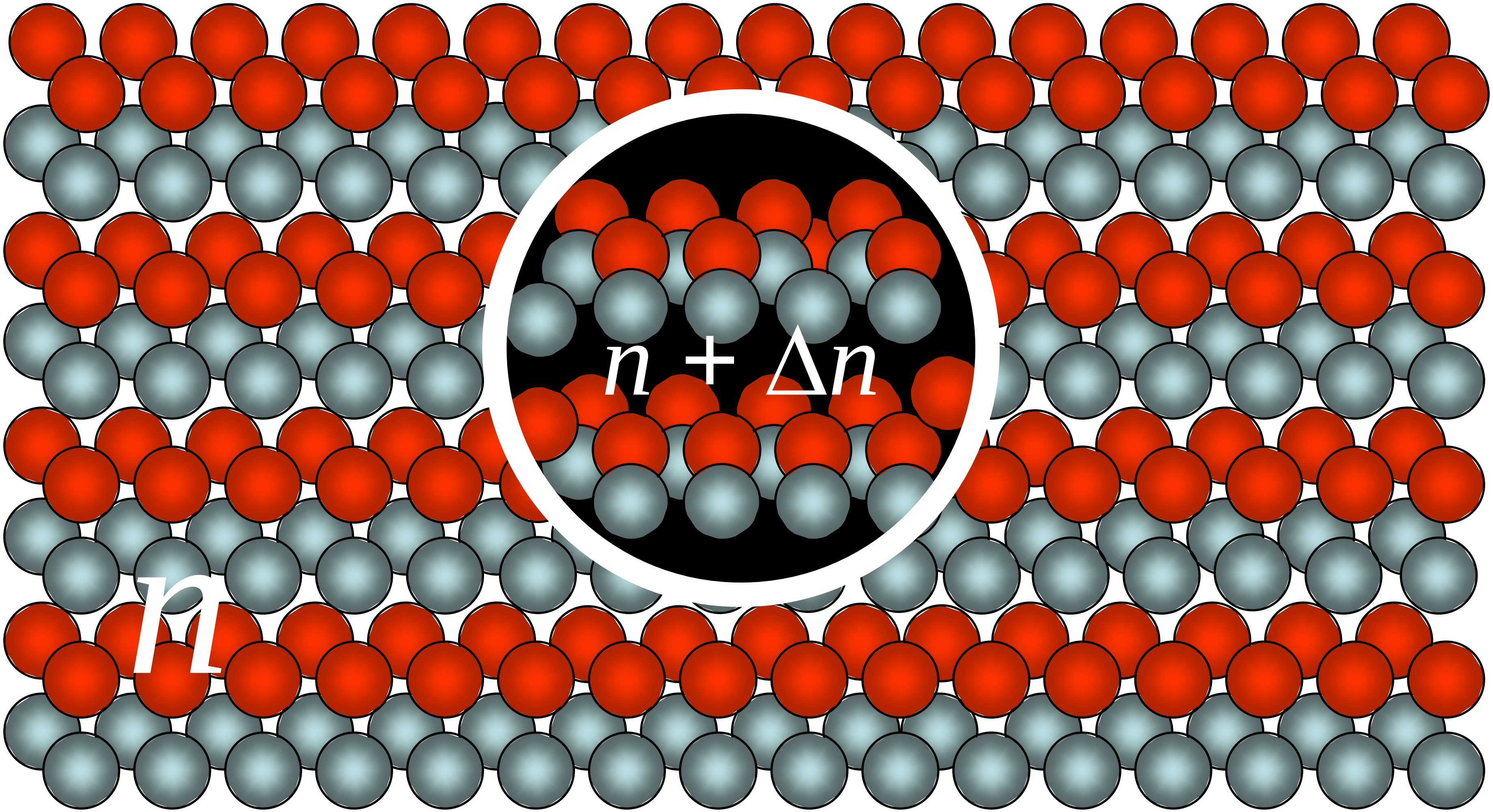
FIRST-ORDER IMPULSIVE FORCE

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

SECOND-ORDER IMPULSIVE CHANGE OF FREQUENCY

IMPULSIVE STIMULATED RAMAN SCATTERING

RAMAN COUPLING MECHANISM



$$n \rightarrow n + \Delta n \; (\Delta n \propto Q, Q^2, Q^3, \ldots)$$

COHERENT PHONONS

$$\ddot{Q} + \Omega^2 Q = \alpha \left| E^2(t) \right|$$

AND

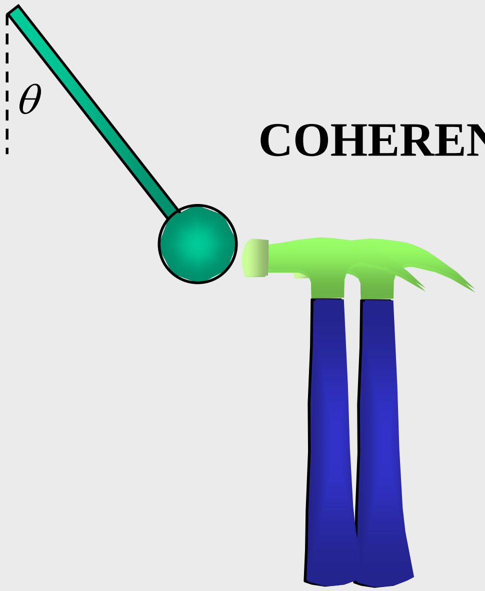
SQUEEZED PHONONS

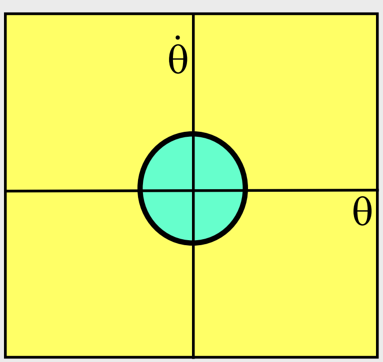
$$\ddot{Q} + \Omega^2 Q = \beta Q \left| E^2(t) \right|$$

CLASSICAL HARMONIC OSCILLATOR

$$mR\Delta(d\theta / dt) = \int F(t)dt$$

COHERENT STATE

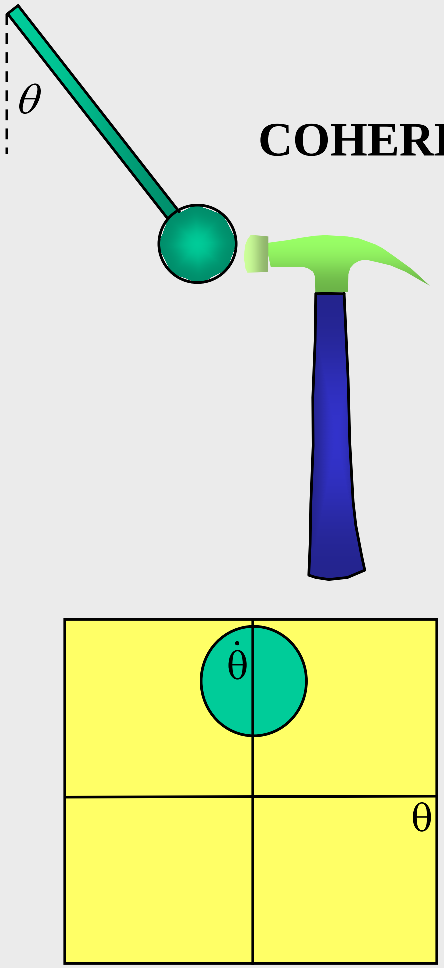




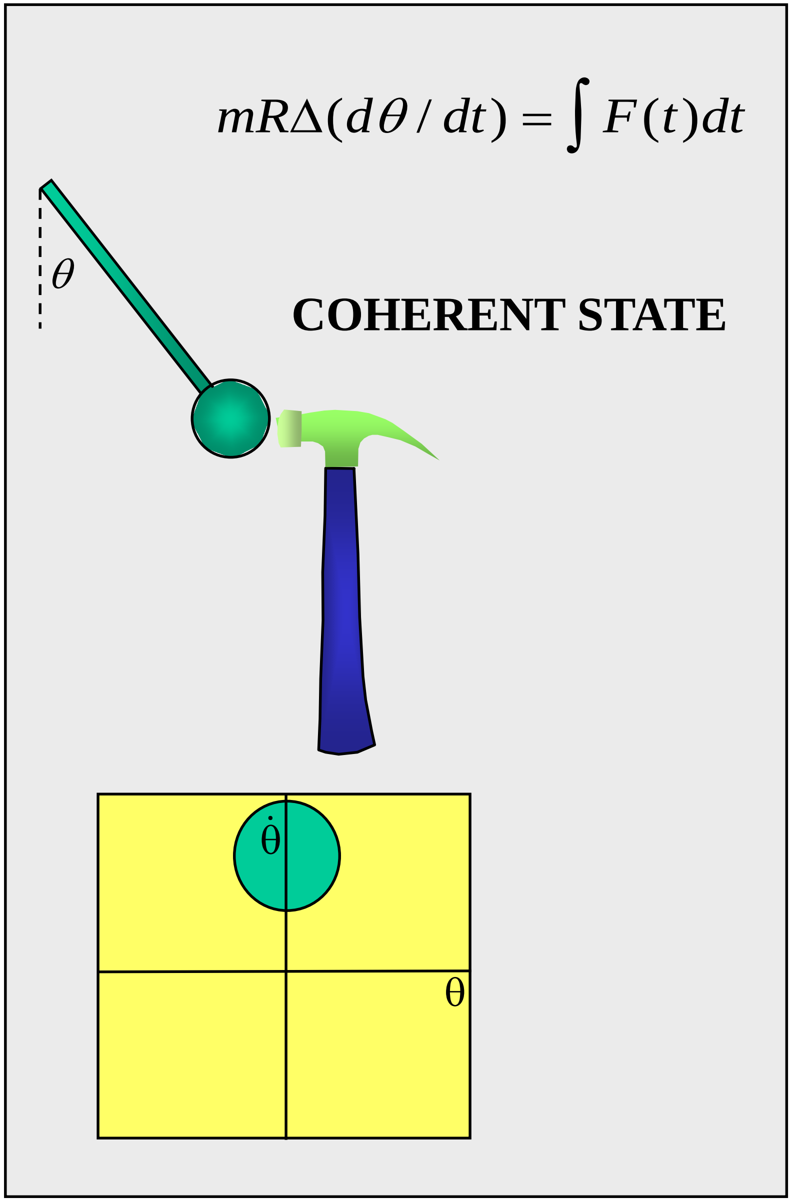
CLASSICAL HARMONIC OSCILLATOR

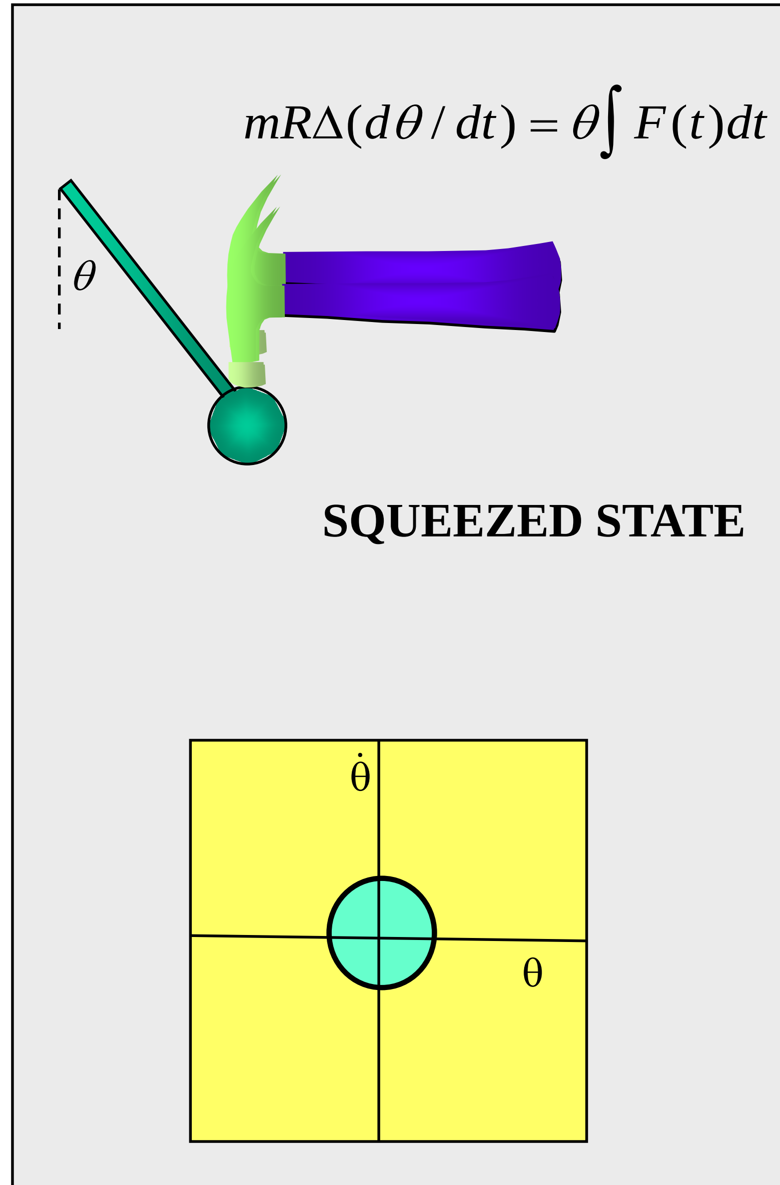
$$mR\Delta(d\theta / dt) = \int F(t)dt$$

COHERENT STATE



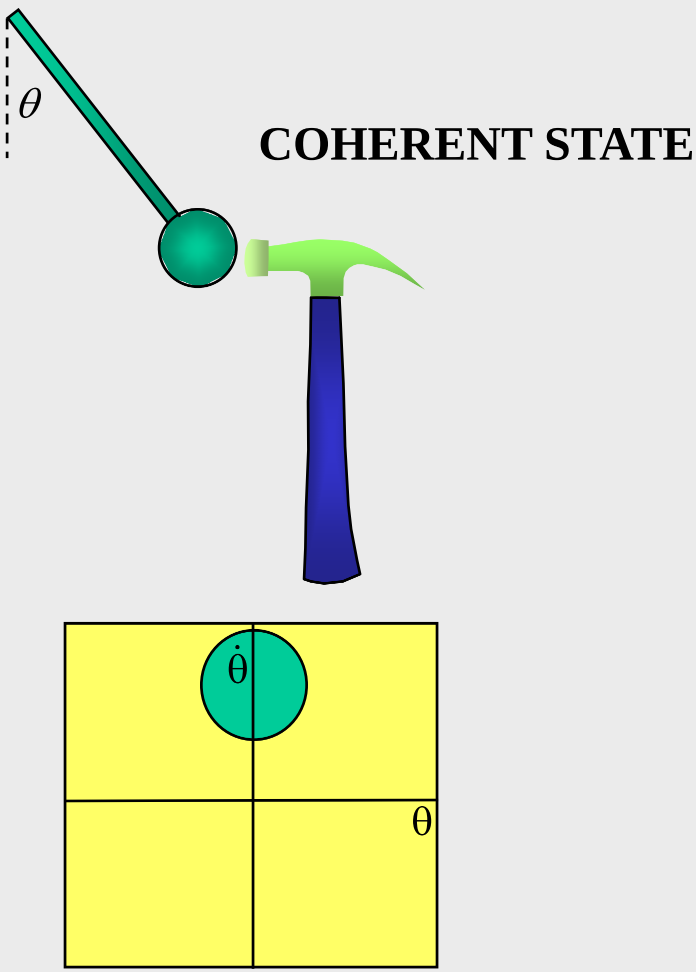
CLASSICAL HARMONIC OSCILLATOR



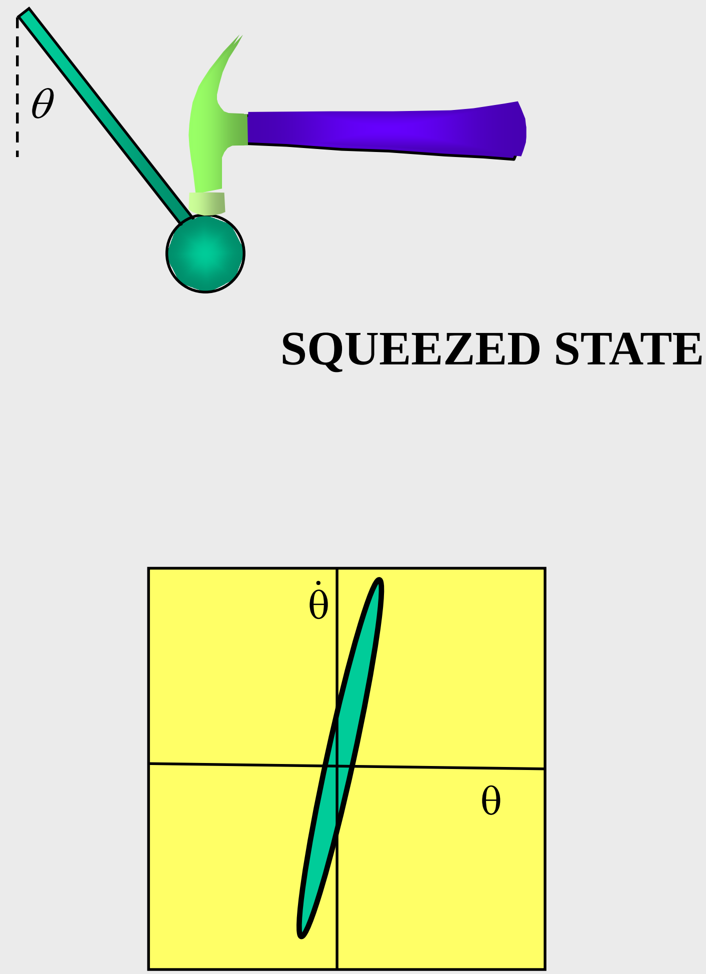


CLASSICAL HARMONIC OSCILLATOR

$$mR\Delta(d\theta / dt) = \int F(t)dt$$



$$mR\Delta(d\theta / dt) = \theta\int F(t)dt$$

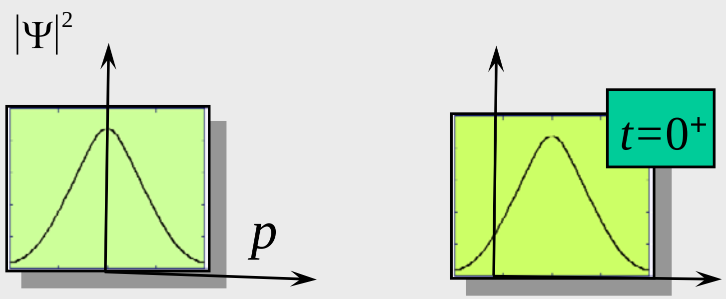


QUANTUM HARMONIC OSCILLATOR

COHERENT STATE

$$\frac{-\hbar^2}{2m}\left(d^2\Psi/dx^2\right)+\left(\frac{m\Omega^2}{2}x^2+\alpha\delta(t)x\right)\Psi = i\hbar\dot{\Psi}$$

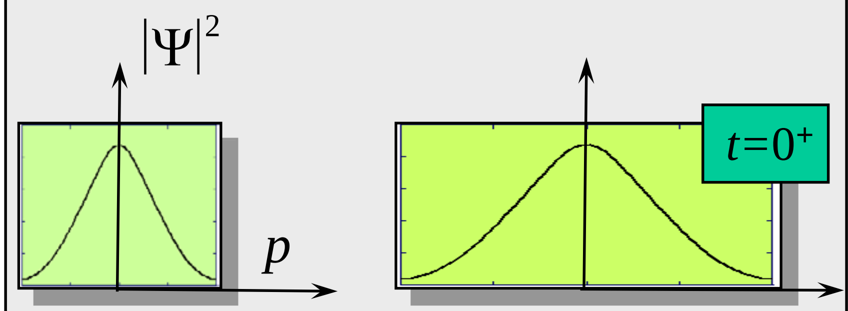
$$\Psi(0^+) = \exp\left(i\alpha x\right)\Psi(0^-)$$



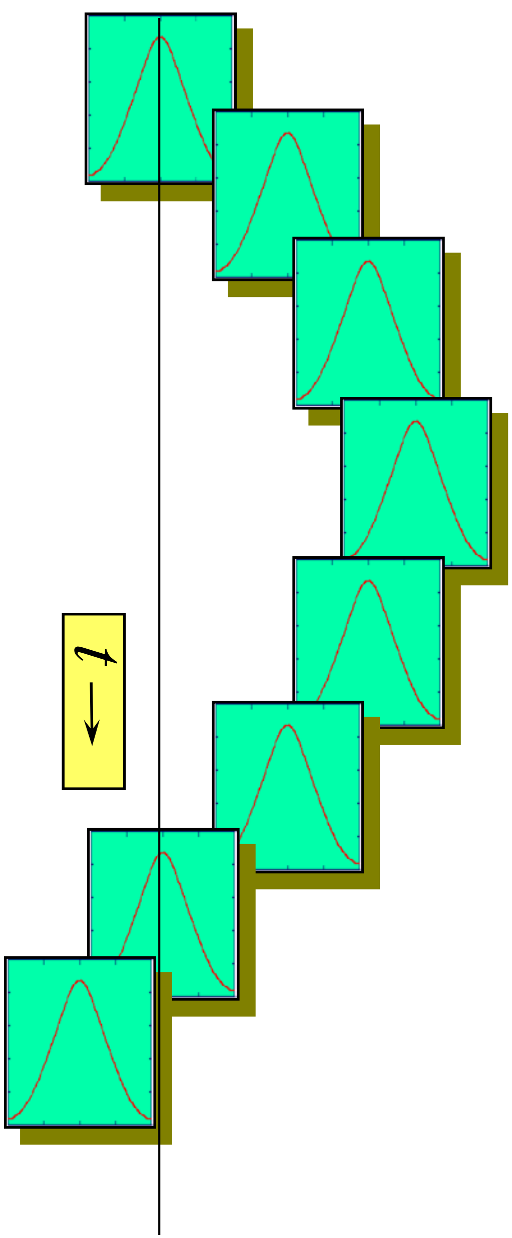
SQUEEZED STATE

$$\frac{-\hbar^2}{2m}\left(d^2\Psi/dx^2\right)+\left(\frac{m\Omega^2}{2}x^2+\beta\delta(t)x^2\right)\Psi = i\hbar\dot{\Psi}$$

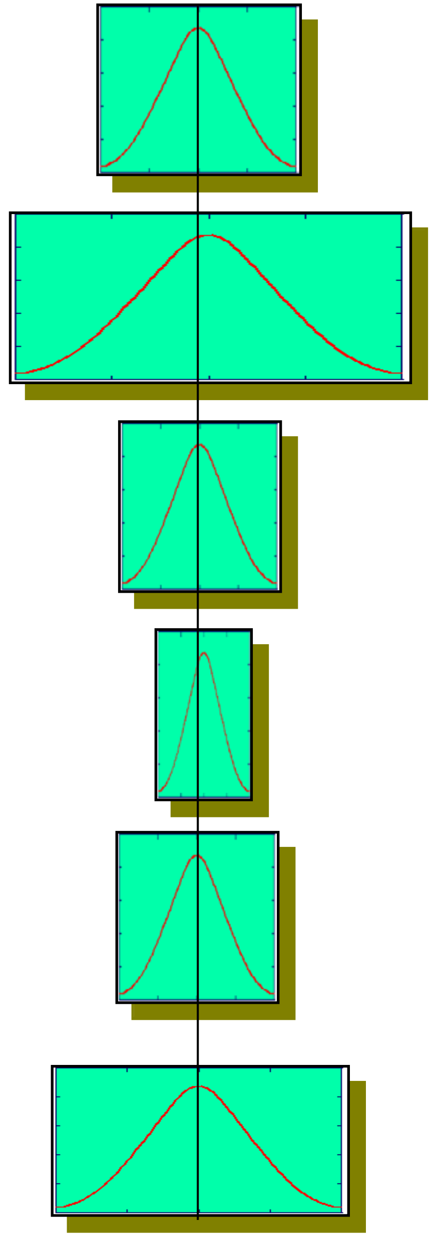
$$\Psi(0^+) = \exp\left(i\beta x^2\right)\Psi(0^-)$$



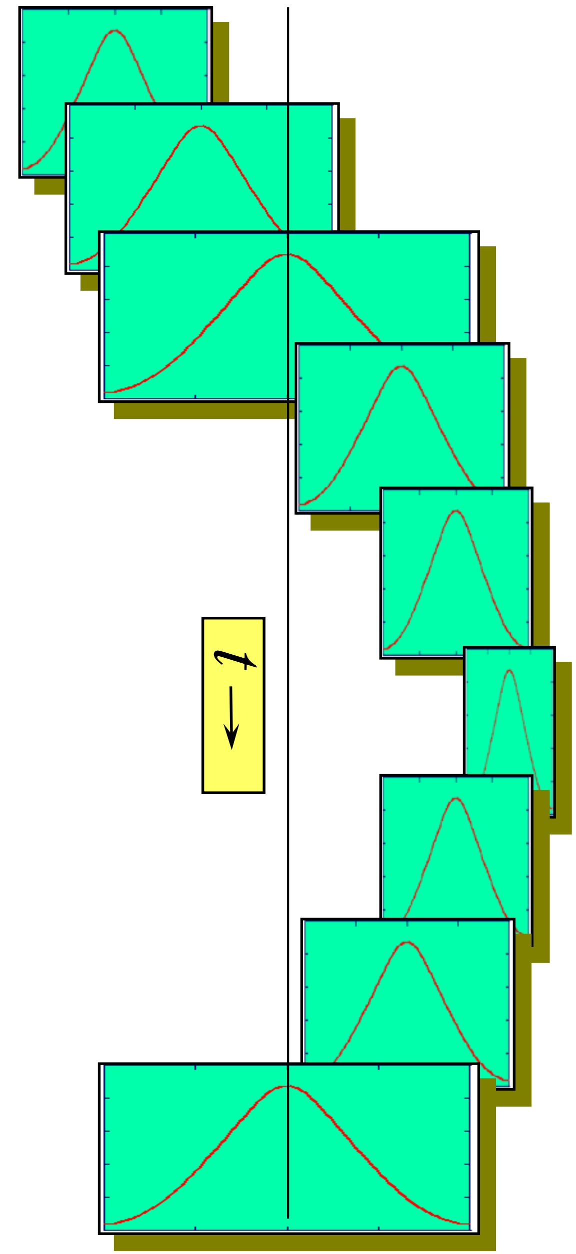
COHERENT STATE



SQUEEZED VACUUM

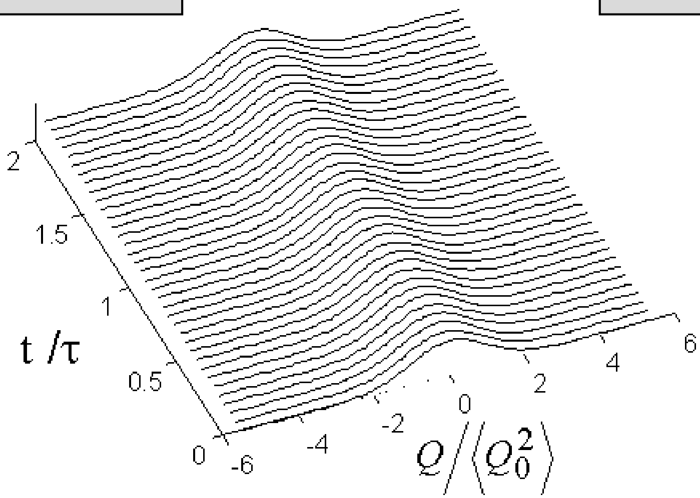


SQUEEZED STATE

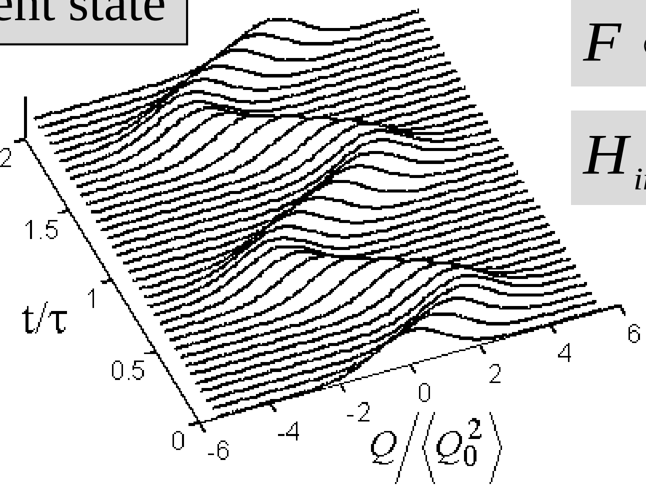


Harmonic Oscillator

ground state



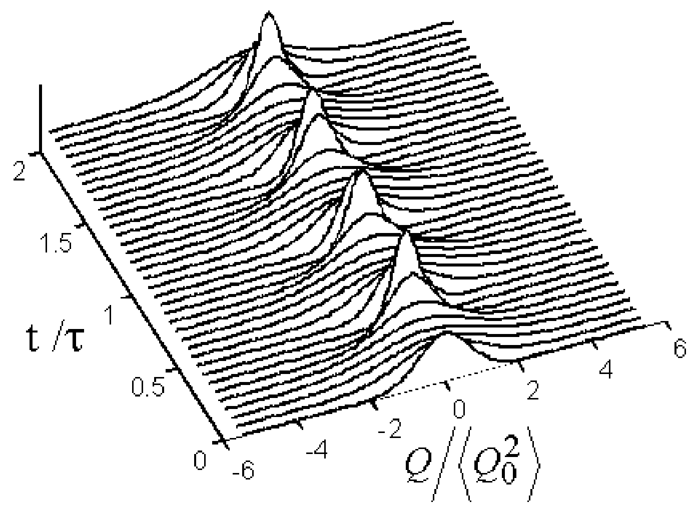
coherent state



$$F \propto \delta(t)$$

$$H_{int} \propto \delta(t)Q$$

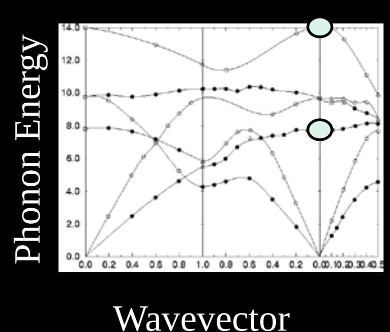
squeezed state



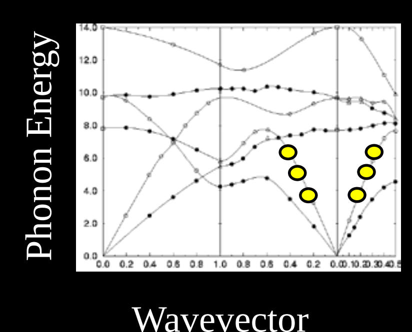
$$F \propto \delta(t)Q$$

$$H_{int} \propto \delta(t)Q^2$$

phonons → Impulsive Raman Scattering



$$\mathbf{q} = 0$$



$$\mathbf{q}_1 + \mathbf{q}_2 = 0$$

BELOW THE GAP: IMPULSIVE EXCITATION

Coherent Phonons

$$\ddot{Q}_0 + \Omega_0^2 Q_0 = F(t) \equiv \sum_i R_{ij}^{(1)} E_i E_j \propto \delta(t)$$

Squeezed Phonons

$$\ddot{Q}_q + \Omega_q^2 Q_q = \left(\sum_i R_{ij}^{(2)} E_i E_j \right) Q_q \propto \delta(t) Q_q$$

PULSE WIDTH $<< \Omega^{-1}$

ABOVE THE GAP: TWO RAMAN TENSORS

$$H = \sum_b \varepsilon_b c_b^+ c_b + \frac{1}{2}\sum_q (P_q^2 + \Omega_q^2 Q_q^2) + \sum \Xi_{kk'} Q_{k-k'} c_k^+ c_{k'}$$

$$\langle \ddot{Q}_q \rangle + \Omega_0^2 \langle Q_q \rangle = F(t) \equiv -\sum_k \Xi_{k,k-q} \langle c_k^+ c_{k-q} \rangle \qquad (q \approx 0)$$

$$F(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} e^{i\Omega t} F(\Omega) d\Omega$$

$$F(\Omega) \propto \left[\frac{d\,\mathrm{Re}(\varepsilon)}{d\omega} + 2i\,\mathrm{Im}\,\varepsilon / \Omega \right] \int_{-\infty}^{+\infty} e^{i\Omega t} \, |E(t)|^2 \, dt$$

Impulsive, Displacive, …….

T. Stevens, J. Kuhl and RM, Phys. Rev. B **65**, 144304 (2002)

ABOVE THE GAP: TWO TENSORS

GENERATION: π^R

$$F(t)=\frac{Nv_c}{4\pi}\sum_{kl}\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty}e^{-i\Omega t}E_l(\omega)\pi^R_{kl}(\omega,\omega-\Omega)$$
$$\times E_k^*(\omega-\Omega)d\omega\, d\Omega,$$

$$F(t)\equiv\langle\hat{\Xi}\rangle=\frac{1}{2\pi\hbar^2}\sum_{mn}\int_{-\infty}^{+\infty}d\omega\int_{-\infty}^{+\infty}d\Omega e^{-i\Omega t}$$
$$\times\left\{\frac{1}{2}\frac{\Xi_{mn}[\boldsymbol{\Delta}_{n0}\cdot\mathbf{E}(\omega)][\boldsymbol{\Delta}_{0m}\cdot\mathbf{E}^*(\omega-\Omega)]}{(\omega_m+i\gamma_{mn}-\omega+\Omega)(\omega_n-i\gamma_{mn}-\omega)}\right. + \ldots$$

$$\pi^R(\omega+\Omega,\omega)\approx\frac{\Xi_0}{4\pi\hbar\Omega}[\varepsilon(\omega+\Omega)-\varepsilon^*(\omega)]$$
$$\approx\frac{\Xi_0}{4\pi\hbar}\left[\frac{d\,\mathrm{Re}(\varepsilon)}{d\omega}+2i\,\mathrm{Im}(\varepsilon)/\Omega\right]$$

New Tensor: Extremely sensitive to lifetime of electronic coherence

J. J. Li, J. Chen, D. A. Reis, S. Fahy, and RM. Phys. Rev. Lett. **110**, 047401 (2013)

DETECTION: χ^R

$$P_k^R(t)=\frac{1}{2\pi}\sum_{l}\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty}e^{-i(\omega-\Omega)t}\chi^R_{kl}(\omega,\omega-\Omega)$$
$$\times E_l(\omega)Q^*(\Omega)d\omega\, d\Omega$$

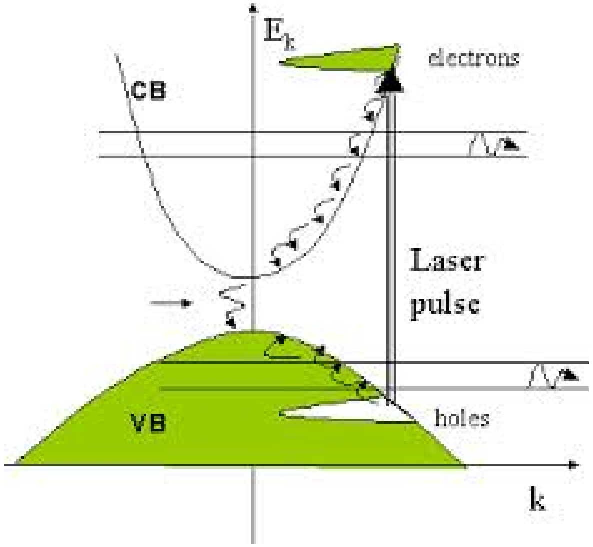
$$\mathbf{P}^R(t)\equiv\frac{\langle\boldsymbol{\Delta}\rangle^R}{Nv_c}=\frac{1}{2\pi\hbar^2 Nv_c}\sum_{mn}\int_{-\infty}^{+\infty}d\omega\int_{-\infty}^{+\infty}d\Omega e^{-i(\omega-\Omega)t}Q^*(\Omega)$$
$$\times\left\{\frac{\Xi_{mn}\boldsymbol{\Delta}_{0m}|\boldsymbol{\Delta}_{n0}\cdot\mathbf{E}(\omega)|}{(\omega_m-i\gamma_m-\omega+\Omega)(\omega_n-i\gamma_n-\omega)}\right. + \ldots$$

$$\chi^R(\omega,\omega+\Omega)\approx\frac{\Xi_0}{4\pi\hbar\Omega}[\varepsilon(\omega+\Omega)-\varepsilon(\omega)]$$
$$\approx\frac{\Xi_0}{4\pi\hbar}\left[\frac{d\,\mathrm{Re}(\varepsilon)}{d\omega}+i\frac{d\,\mathrm{Im}(\varepsilon)}{d\omega}\right]$$

Conventional Raman Tensor: Not very sensitive to lifetime of electronic intermediate states (except at resonances)

NON-RAMAN MECHANISMS

DISPLACIVE EXCITATION OF COHERENT PHONONS



(WORKS ONLY FOR FULLY-SYMMETRIC MODES)

Ti_2O_3

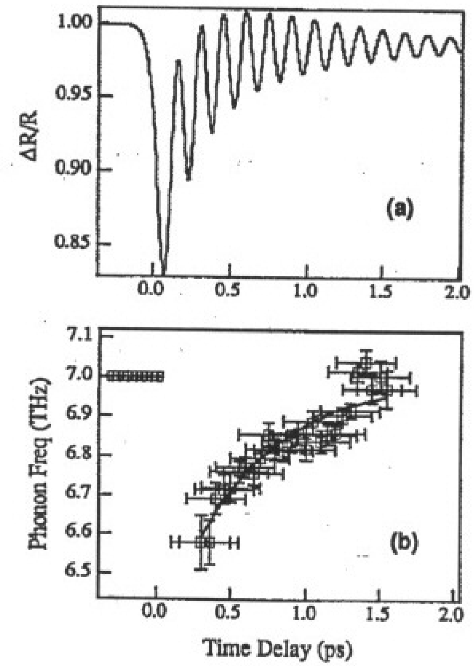


FIG. 3. (a) The pump-induced coherent phonon amplitude produces a $\Delta R/R$ as large as 12% initially. This plot convolved with the optical pulse intensity profile yields the least-squares fit to the data in Fig. 2. The fitting function is taken to be an exponentially damped cosine, superimposed on 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.

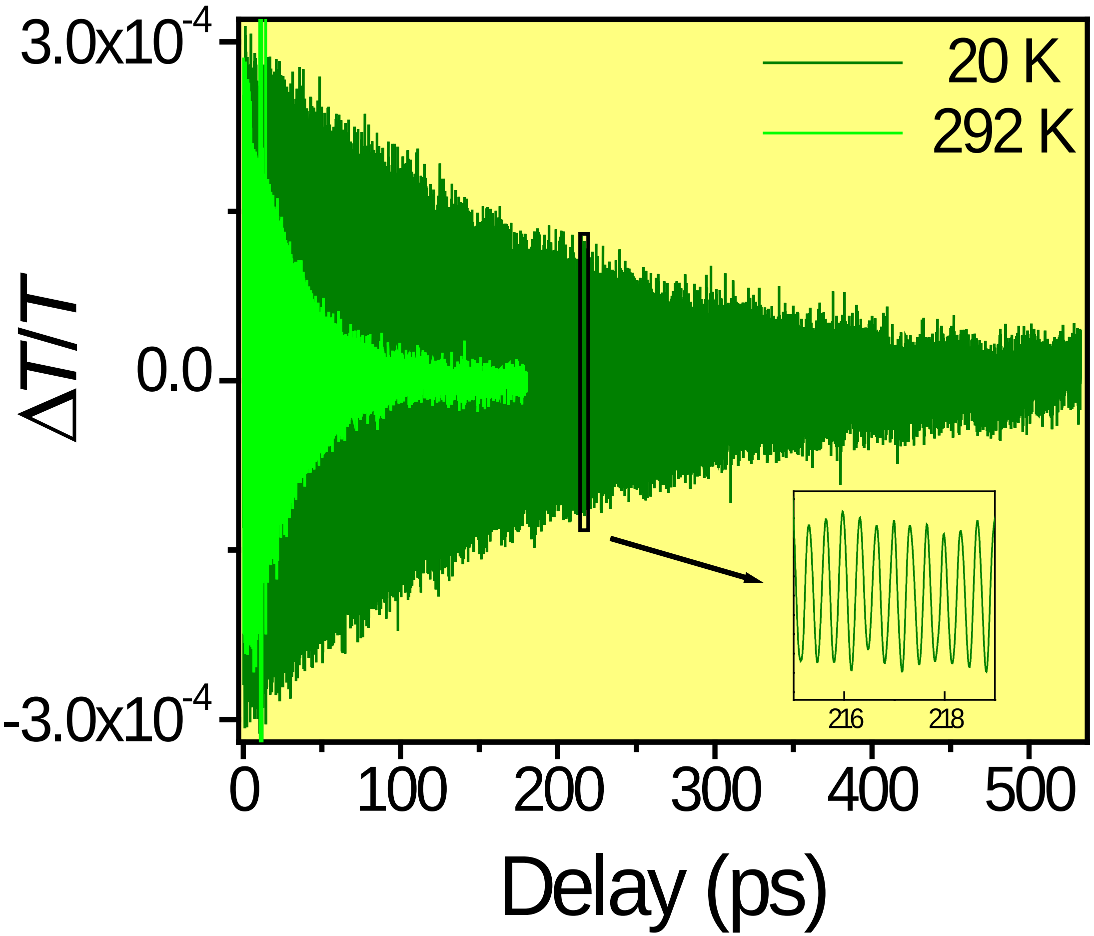
Chen et al., Appl. Phys. Lett. **62**, 1901 (1993)

- Coherent Optical Phonons
- Coherent Acoustic Phonons
- Squeezed Phonons
- Coherent Polaritons

- Optical Phonons
- Acoustic Phonons
- Squeezed Phonons
- Polaritons

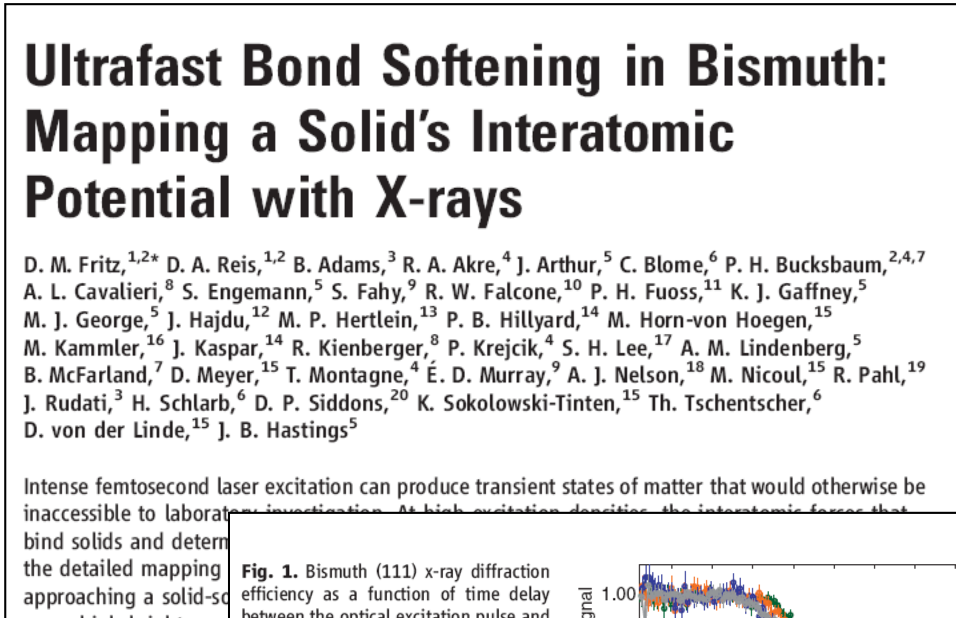
COHERENT OPTICAL PHONONS

ZnO



C. Aku-Leh, J. Zhao, RM, J. Menendez and M. Cardona, Phys. Rev. B **71**, 205211 (2005)

COHERENT OPTICAL PHONONS



Ultrafast Bond Softening in Bismuth: Mapping a Solid's Interatomic Potential with X-rays

D. M. Fritz,[1,2*] D. A. Reis,[1,2] B. Adams,[3] R. A. Akre,[4] J. Arthur,[5] C. Blome,[6] P. H. Bucksbaum,[2,4,7] A. L. Cavalieri,[8] S. Engemann,[5] S. Fahy,[9] R. W. Falcone,[10] P. H. Fuoss,[11] K. J. Gaffney,[5] M. J. George,[5] J. Hajdu,[12] M. P. Hertlein,[13] P. B. Hillyard,[14] M. Horn-von Hoegen,[15] M. Kammler,[16] J. Kaspar,[14] R. Kienberger,[8] P. Krejcik,[4] S. H. Lee,[17] A. M. Lindenberg,[5] B. McFarland,[7] D. Meyer,[15] T. Montagne,[4] É. D. Murray,[9] A. J. Nelson,[18] M. Nicoul,[15] R. Pahl,[19] J. Rudati,[3] H. Schlarb,[6] D. P. Siddons,[20] K. Sokolowski-Tinten,[15] Th. Tschentscher,[6] D. von der Linde,[15] J. B. Hastings[5]

Intense femtosecond laser excitation can produce transient states of matter that would otherwise be inaccessible to laboratory investigation. At high excitation densities, the interatomic forces that bind solids and determ

the detailed mapping

approaching a solid-sc

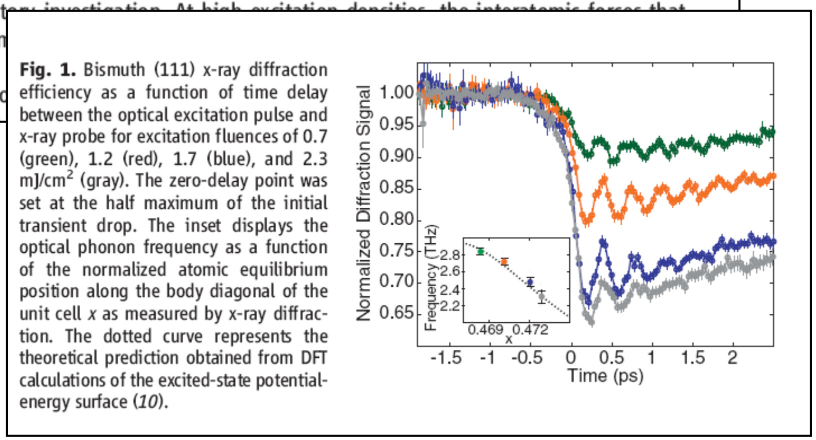
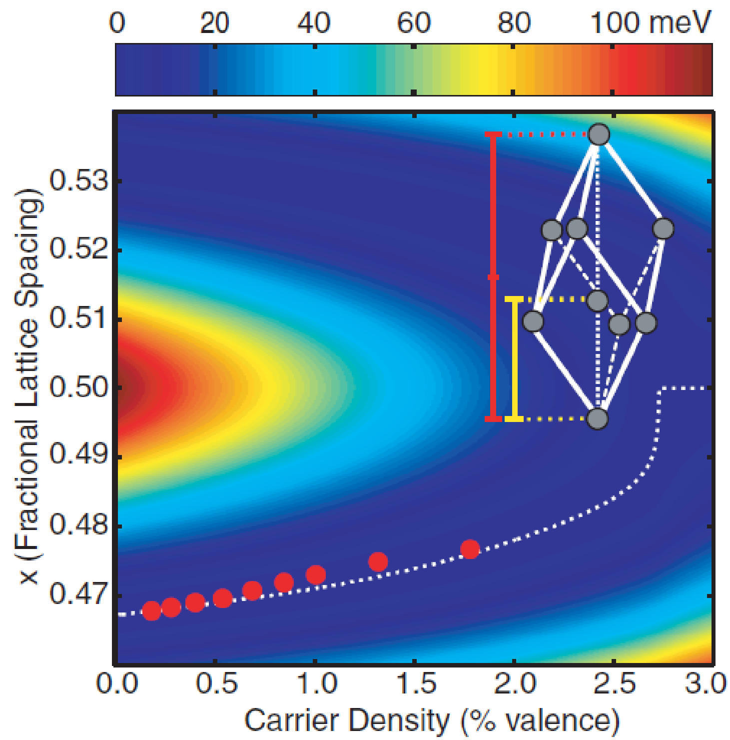


Fig. 1. Bismuth (111) x-ray diffraction efficiency as a function of time delay between the optical excitation pulse and x-ray probe for excitation fluences of 0.7 (green), 1.2 (red), 1.7 (blue), and 2.3 mJ/cm^2 (gray). The zero-delay point was set at the half maximum of the initial transient drop. The inset displays the optical phonon frequency as a function of the normalized atomic equilibrium position along the body diagonal of the unit cell *x* as measured by x-ray diffraction. The dotted curve represents the theoretical prediction obtained from DFT calculations of the excited-state potential-energy surface (*10*).



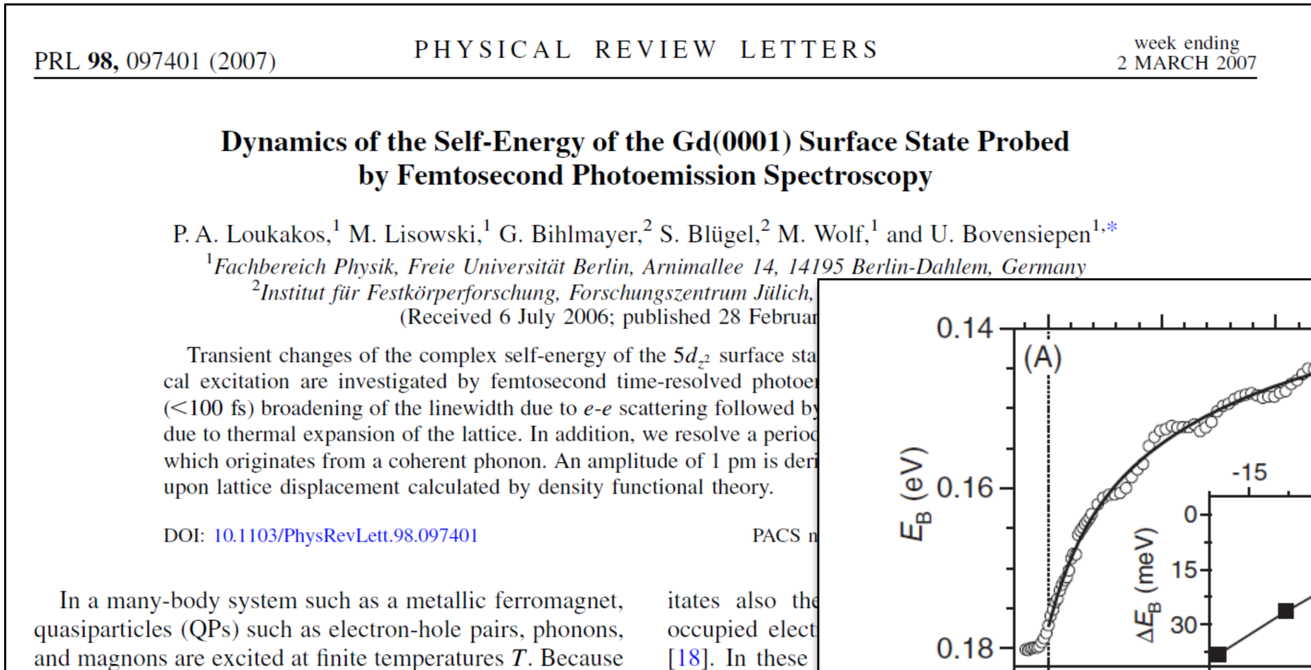
Bi

Fritz et al., Science **315**, 633 (2007)

PUMP = LIGHT

PROBE = X-RAYS

COHERENT OPTICAL PHONONS



PRL **98,** 097401 (2007) PHYSICAL REVIEW LETTERS week ending 2 MARCH 2007

Dynamics of the Self-Energy of the Gd(0001) Surface State Probed by Femtosecond Photoemission Spectroscopy

P. A. Loukakos,[1] M. Lisowski,[1] G. Bihlmayer,[2] S. Blügel,[2] M. Wolf,[1] and U. Bovensiepen[1,*]
[1]*Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin-Dahlem, Germany*
[2]*Institut für Festkörperforschung, Forschungszentrum Jülich,*
(Received 6 July 2006; published 28 Februar

Transient changes of the complex self-energy of the $5d_{z^2}$ surface sta
cal excitation are investigated by femtosecond time-resolved photoe
(<100 fs) broadening of the linewidth due to *e-e* scattering followed by
due to thermal expansion of the lattice. In addition, we resolve a perio
which originates from a coherent phonon. An amplitude of 1 pm is deri
upon lattice displacement calculated by density functional theory.

DOI: 10.1103/PhysRevLett.98.097401 PACS n

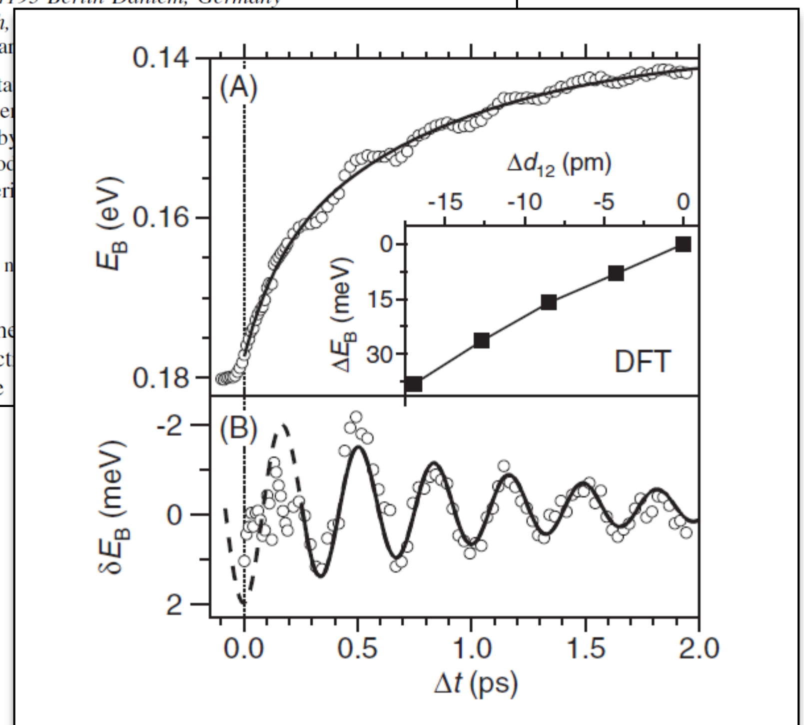
In a many-body system such as a metallic ferromagnet, quasiparticles (QPs) such as electron-hole pairs, phonons, and magnons are excited at finite temperatures *T*. Because

itates also the
occupied elect
[18]. In these

Gd

PUMP = LIGHT

PROBE = XPS



Loukakos et al., Phys. Rev. Lett. **98**, 097401 (2007)

COHERENT AMPLITUDONS

IOP Institute of Physics DEUTSCHE PHYSIKALISCHE GESELLSCHAFT

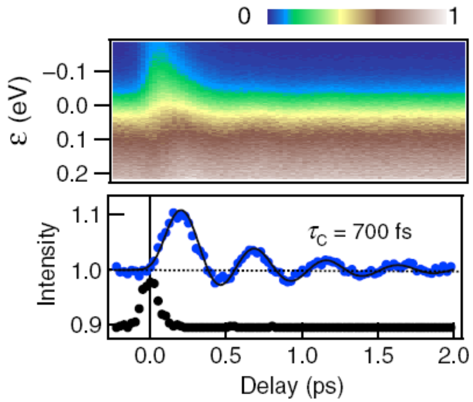


Figure 5. Upper panel: intensity map of the pl
in the metallic phase ($T_1 = 300$ K). Lower panel:
photoelectron counts in the energy interval 0 eV <
based on equation (3) (black line) and cross-corre
probe pulse (black marks).

TaS_2

New Journal of Physics

The open-access journal for physics

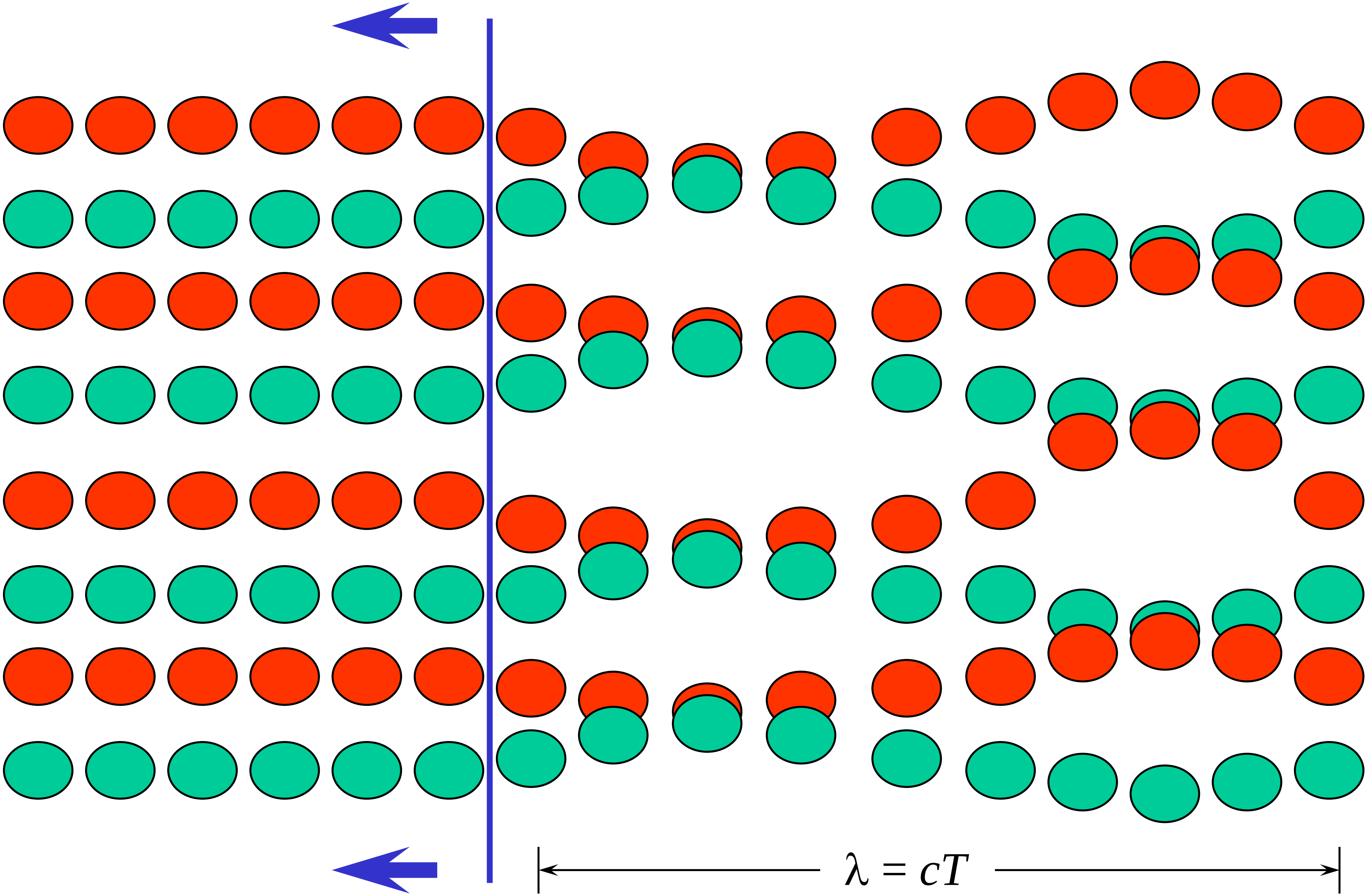
Femtosecond dynamics of electronic states in the Mott insulator 1T-TaS_2 by time resolved photoelectron spectroscopy

L Perfetti[1,4], P A Loukakos[1], M Lisowski[1],
U Bovensiepen[1], M Wolf[1], H Berger[2],
S Biermann[3] and A Georges[3]

PUMP = LIGHT

PROBE = XPS

COHERENT PHONON FIELD



- Optical Phonons
- Acoustic Phonons
- Squeezed Phonons
- Polaritons

COHERENT ACOUSTIC MODES (Sound Waves)

2562 IEEE JOURNAL OF QUANTUM ELECTRONICS, VOL. 25, NO. 12, DECEMBER 1989

Picosecond Ultrasonics

HOLGER T. GRAHN, HUMPHREY J. MARIS, AND JAN TAUC

(Invited Paper)

GENERATION:
ABSORPTION-INDUCED STRESS
(no dependence on light polarization)

DETECTION:
STIMULATED BRILLOUIN SCATTERING
(selection rules)

GRAHN *et al.*: PICOSECOND ULTRASONICS

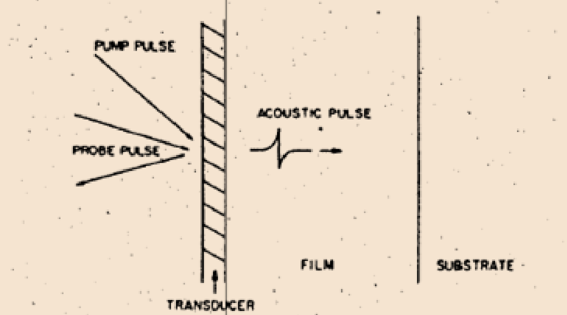
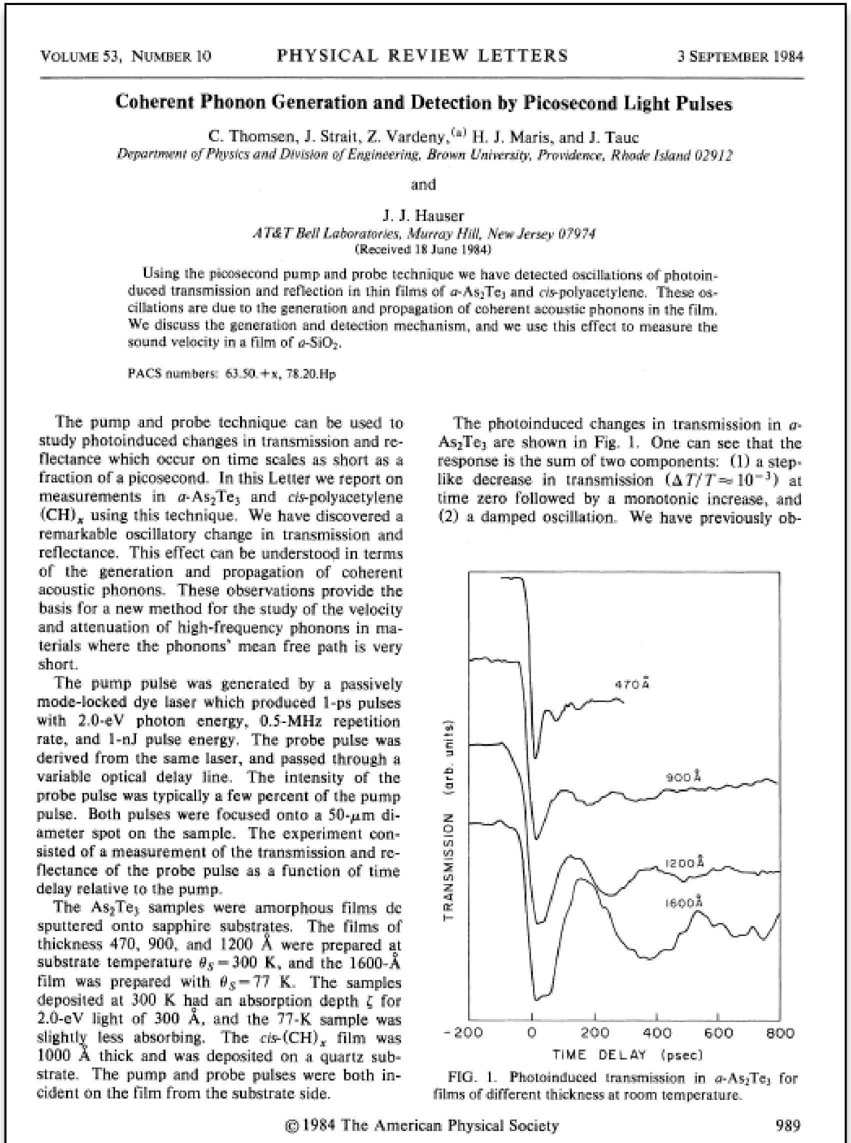


Fig. 4. Schematic diagram for the transducer method to measure transparent films.

TWO TYPES OF OSCILLATIONS

- GEOMETRICAL (PERIOD DEPENDS ON SAMPLE THICKNESS)
- STIMULATED BRILLOUIN SCATTERING (PERIOD DEPENDS ON LASER WAVELENGTH)

ACOUSTIC PHONONS



VOLUME 53, NUMBER 10 PHYSICAL REVIEW LETTERS 3 SEPTEMBER 1984

Coherent Phonon Generation and Detection by Picosecond Light Pulses

C. Thomsen, J. Strait, Z. Vardeny,[a] H. J. Maris, and J. Tauc
Department of Physics and Division of Engineering, Brown University, Providence, Rhode Island 02912

and

J. J. Hauser
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
(Received 18 June 1984)

Using the picosecond pump and probe technique we have detected oscillations of photoinduced transmission and reflection in thin films of a-As_2Te_3 and *cis*-polyacetylene. These oscillations are due to the generation and propagation of coherent acoustic phonons in the film. We discuss the generation and detection mechanism, and we use this effect to measure the sound velocity in a film of a-SiO_2.

PACS numbers: 63.50.+x, 78.20.Hp

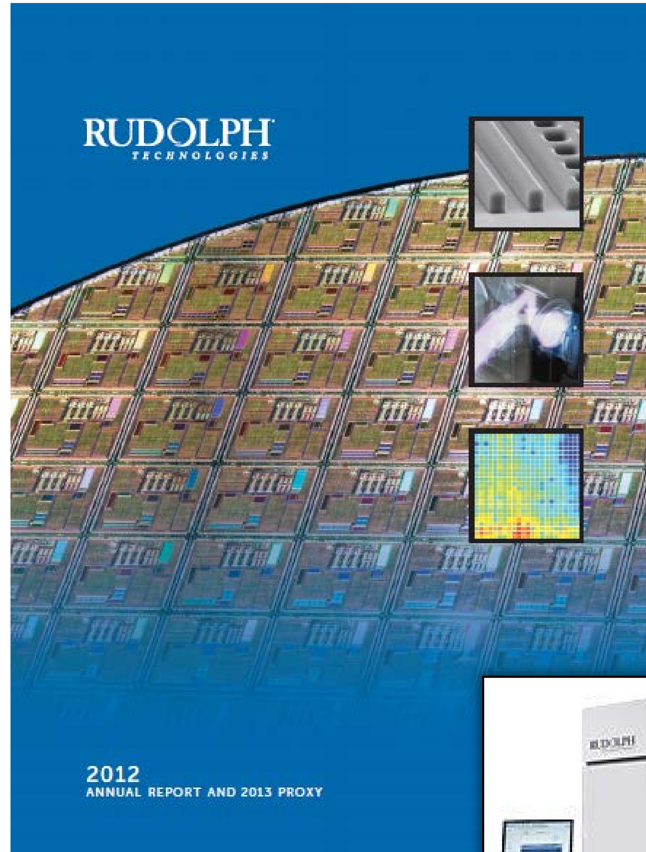
The pump and probe technique can be used to study photoinduced changes in transmission and reflectance which occur on time scales as short as a fraction of a picosecond. In this Letter we report on measurements in a-As_2Te_3 and *cis*-polyacetylene $(CH)_x$ using this technique. We have discovered a remarkable oscillatory change in transmission and reflectance. This effect can be understood in terms of the generation and propagation of coherent acoustic phonons. These observations provide the basis for a new method for the study of the velocity and attenuation of high-frequency phonons in materials where the phonons' mean free path is very short.

The pump pulse was generated by a passively mode-locked dye laser which produced 1-ps pulses with 2.0-eV photon energy, 0.5-MHz repetition rate, and 1-nJ pulse energy. The probe pulse was derived from the same laser, and passed through a variable optical delay line. The intensity of the probe pulse was typically a few percent of the pump pulse. Both pulses were focused onto a 50-μm diameter spot on the sample. The experiment consisted of a measurement of the transmission and reflectance of the probe pulse as a function of time delay relative to the pump.

The As_2Te_3 samples were amorphous films dc sputtered onto sapphire substrates. The films of thickness 470, 900, and 1200 Å were prepared at substrate temperature $\theta_S = 300$ K, and the 1600-Å film was prepared with $\theta_S = 77$ K. The samples deposited at 300 K had an absorption depth ζ for 2.0-eV light of 300 Å, and the 77-K sample was slightly less absorbing. The *cis*-$(CH)_x$ film was 1000 Å thick and was deposited on a quartz substrate. The pump and probe pulses were both incident on the film from the substrate side.

The photoinduced changes in transmission in a-As_2Te_3 are shown in Fig. 1. One can see that the response is the sum of two components: (1) a step-like decrease in transmission ($\Delta T/T \approx 10^{-3}$) at time zero followed by a monotonic increase, and (2) a damped oscillation. We have previously ob-

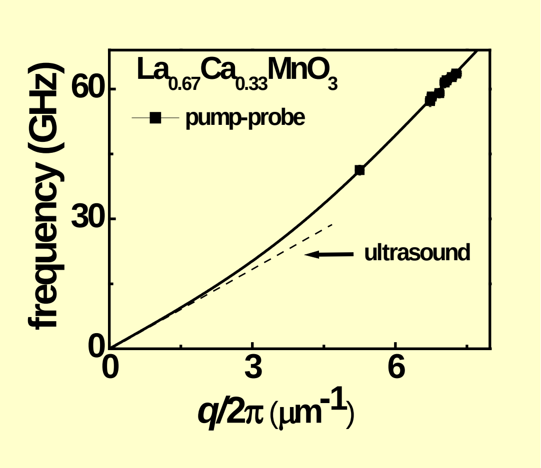
FIG. 1. Photoinduced transmission in a-As_2Te_3 for films of different thickness at room temperature.

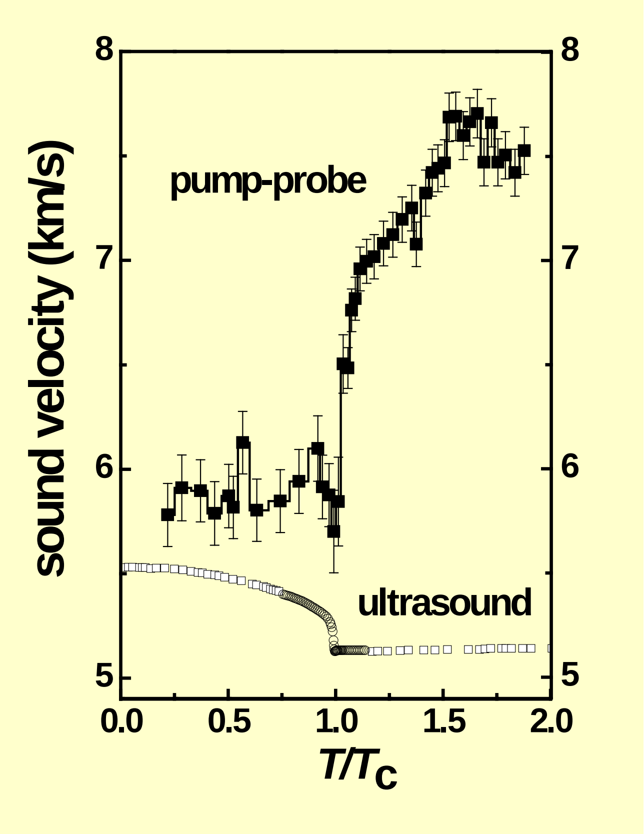


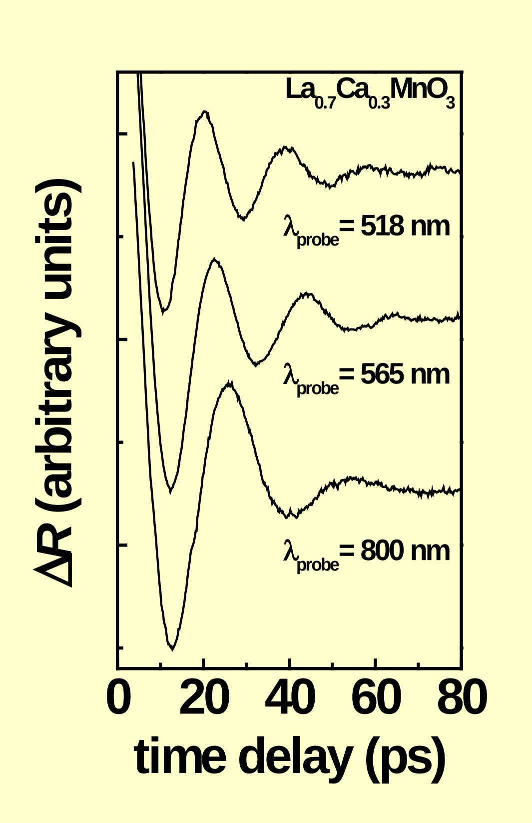
C. Thomsen et al., Phys. Rev. Lett. **53**, 989 (1984)

ACOUSTIC PHONONS

Anomalous First-to-Zero Sound Crossover
in $La_{1-x}Ca_xMnO_3$







Y. Ren et al., Phys. Rev. B **74**, 012405 (2006)

ACOUSTIC PHONONS

PRL 101, 025505 (2008) PHYSICAL REVIEW LETTERS week ending 11 JULY 2008

Probing Unfolded Acoustic Phonons with X Rays

M. Trigo,[1,*] Y. M. Sheu,[1] D. A. Arms,[2] J. Chen,[1] S. Ghimire,[1] R. S. Goldman,[3] E. Landahl,[2,†] R. Merlin,[1] E. Peterson,[1] M. Reason,[3] and D. A. Reis[1]

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(Received 6 May 2008; published 11 July 2008)

Ultrafast laser excitation of an InGaAs/InAlAs superlattice (SL) creates coherent folded acoustic phonons that subsequently leak into the bulk (InP) substrate. Upon transmission, the phonons become "unfolded" into bulk modes and acquire a wave vector much larger than that of the light. We show that time-resolved x-ray diffraction is sensitive to this large-wave vector excitation in the substrate. Comparison with dynamical diffraction simulations of propagating strain supports our interpretation.

DOI: 10.1103/PhysRevLett.101.025505 PACS numbers: 63.22.−m, 78.47.−p

High-frequency acoustic phonons with large wave vectors and short mean free paths play a fundamental role in heat transport and energy relaxation in solids [1]. In particular, in insulators these modes are the dominant carriers of heat. The combination of their short wavelengths and relatively low energies make them a unique probe of interfaces and nanoscale structures [2]. Thus, there is great interest in the generation and detection of coherent acoustic modes with such characteristics. Ultrafast laser pulses, together with the branch folding that occurs in a superlat-

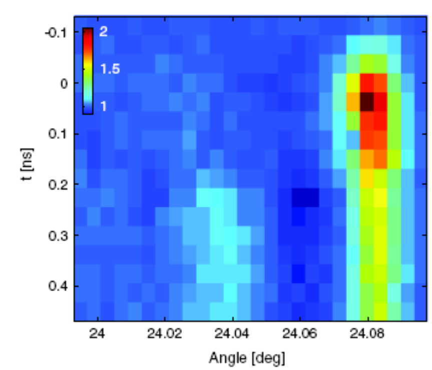
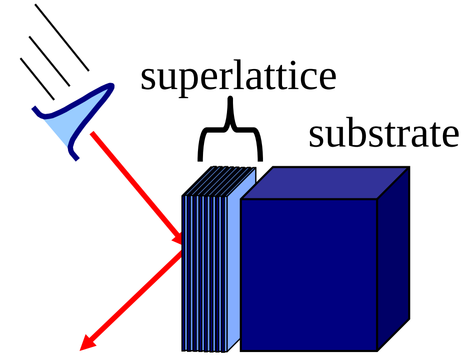


FIG. 2 (color online). Ratio I_{on}/I_{off} as a function of time delay at the expected angular position for the folded phonons below the (-1) sideband. The feature at 24.08° is the shift of the (-1) peak due to thermal expansion induced by the laser pulse. At $t = 0.2$ ns, the signature from unfolded phonons in the substrate appears at $\theta = 24.04°$, which corresponds to a wave vector transfer $q^* = 2\pi/D^*$.



InGaAs/InAlAs on InP

PUMP = LIGHT

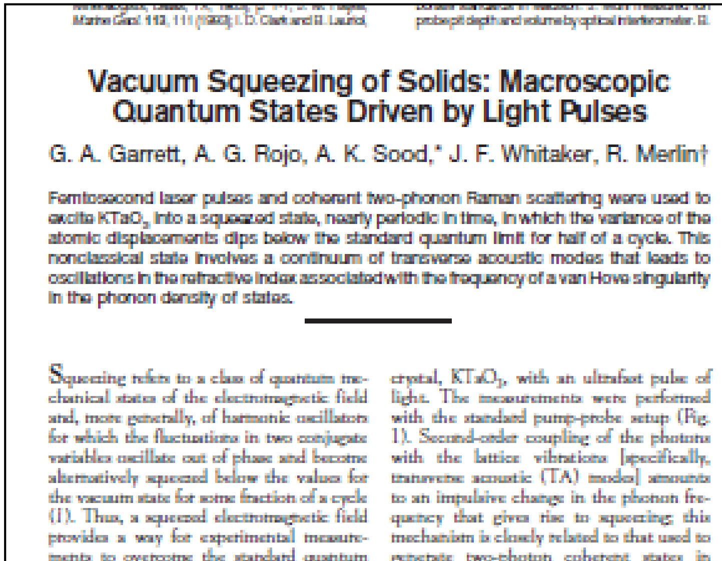
PROBE = X-RAYS

M. Trigo et al., Phys. Rev. Lett. **101**, 025505 (2008)

- Optical Phonons
- Acoustic Phonons
- Squeezed Phonons
- Polaritons

SQUEEZED PHONONS

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



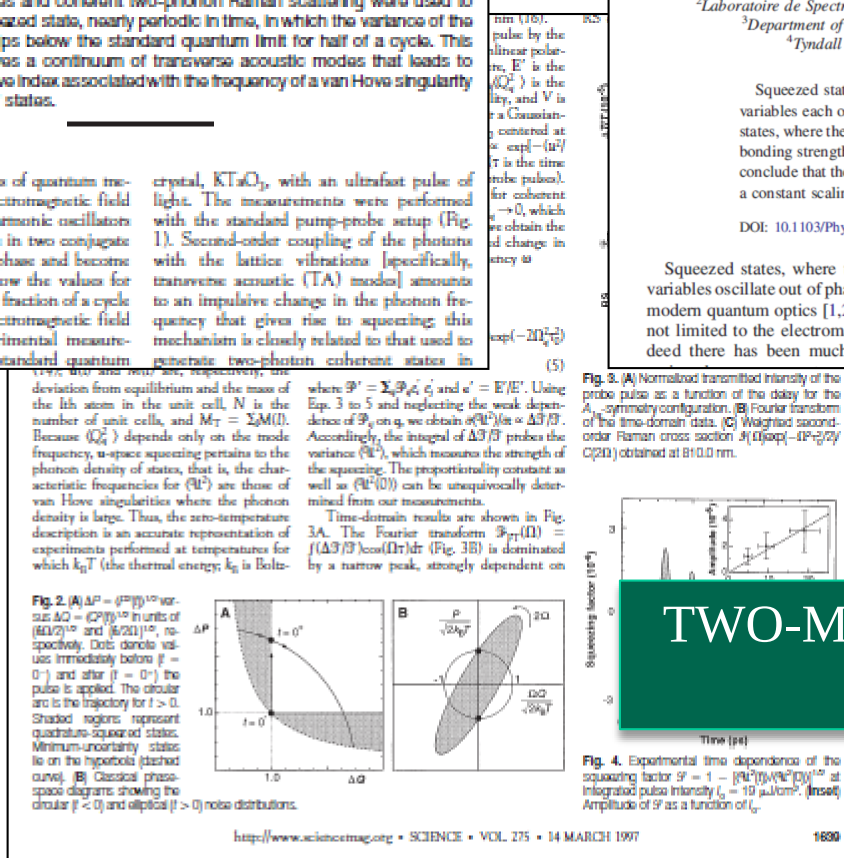
Vacuum Squeezing of Solids: Macroscopic Quantum States Driven by Light Pulses

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

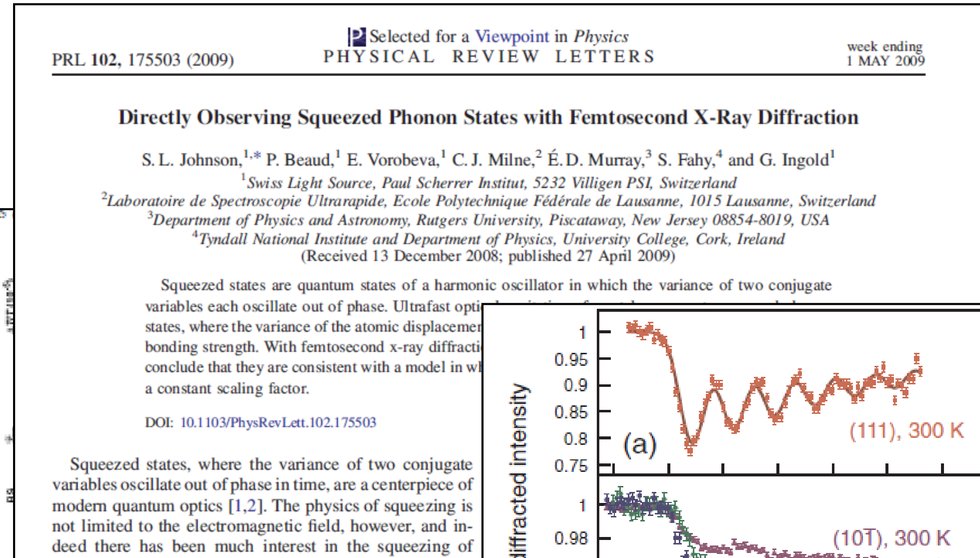
Femtosecond laser pulses and coherent two-phonon Raman scattering were used to excite $KTaO_3$ into a squeezed state, nearly periodic in time, in which the variance of the atomic displacements dips below the standard quantum limit for half of a cycle. This nonclassical state involves a continuum of transverse 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 photon frequency that gives rise to squeezing; this mechanism is closely related to that used to generate two-photon coherent states in



KTaO$_3$



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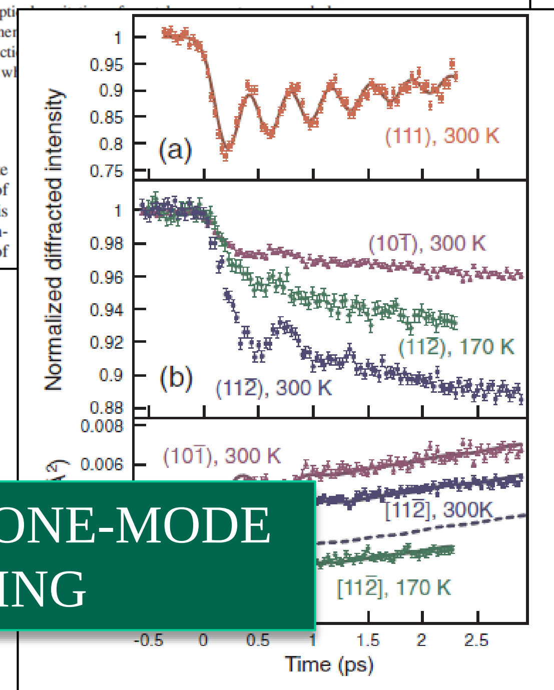
Squeezed states are quantum states of a harmonic oscillator in which the variance of two conjugate variables each oscillate out of phase. Ultrafast opti... states, where the variance of the atomic displacemen... bonding strength. With femtosecond x-ray diffracti... conclude that they are consistent with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

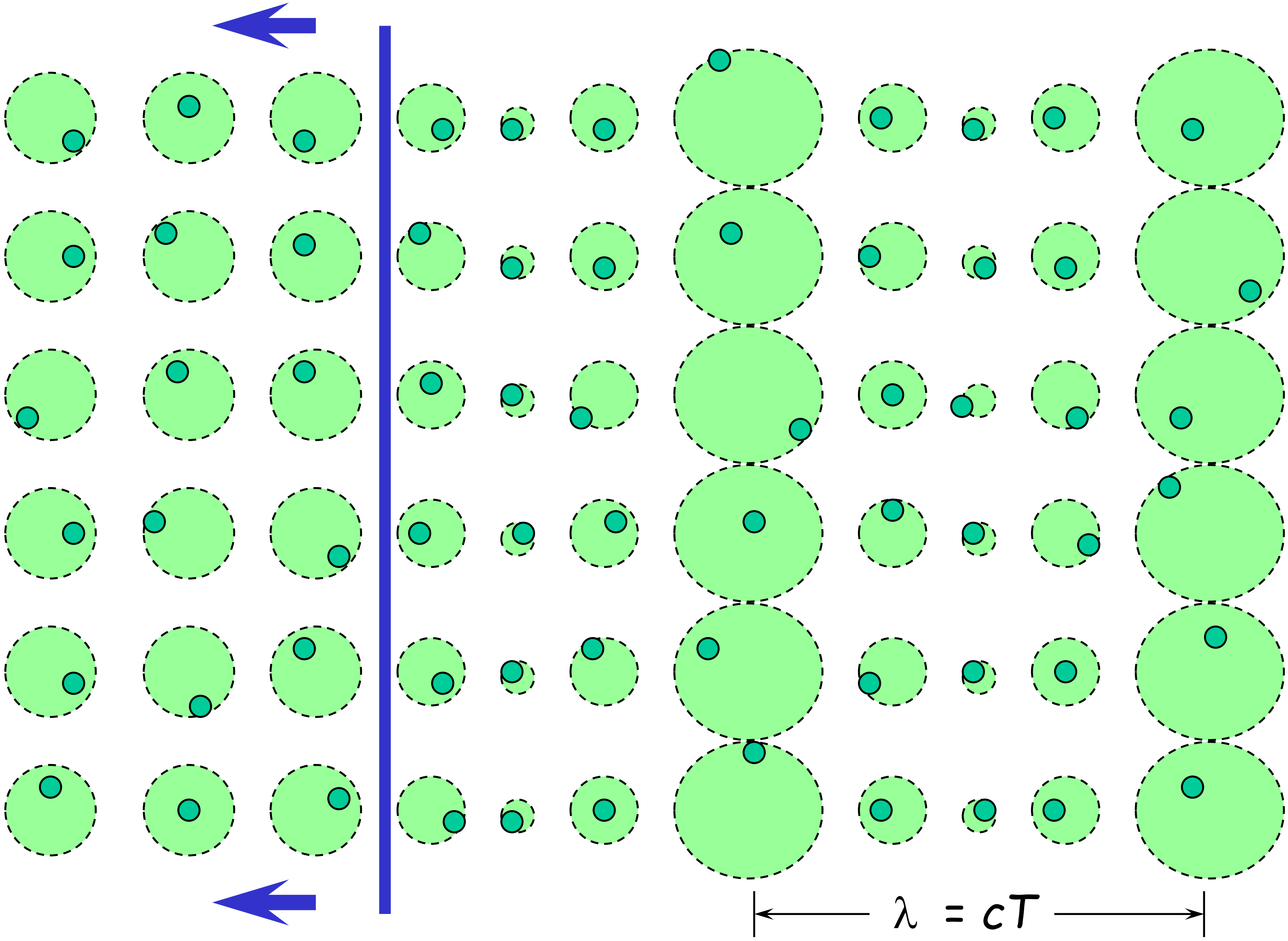


TWO-MODE vs. ONE-MODE SQUEEZING

Bi

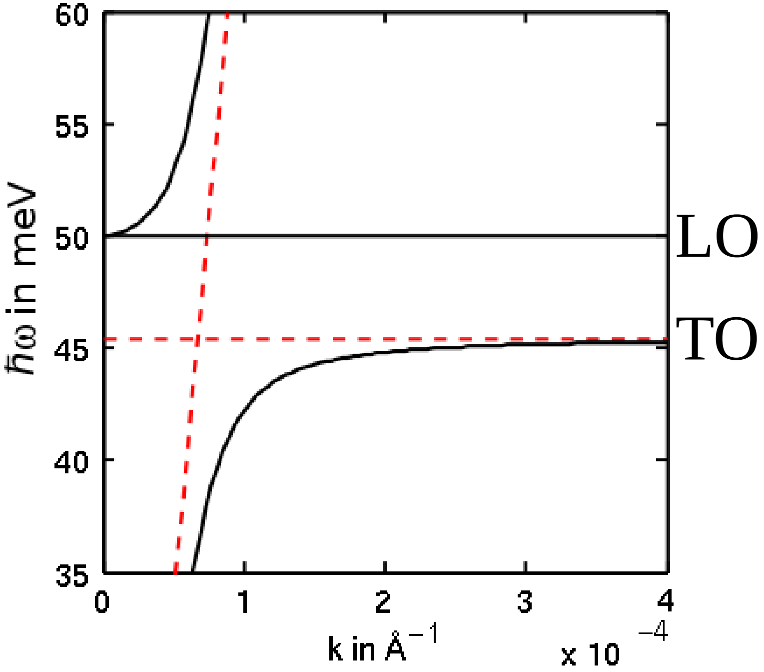
G. A. Garrett, A. G. Rojo, A. K. Sood, J. F. Whitaker and RM, Science **275**, 1638 (1997)

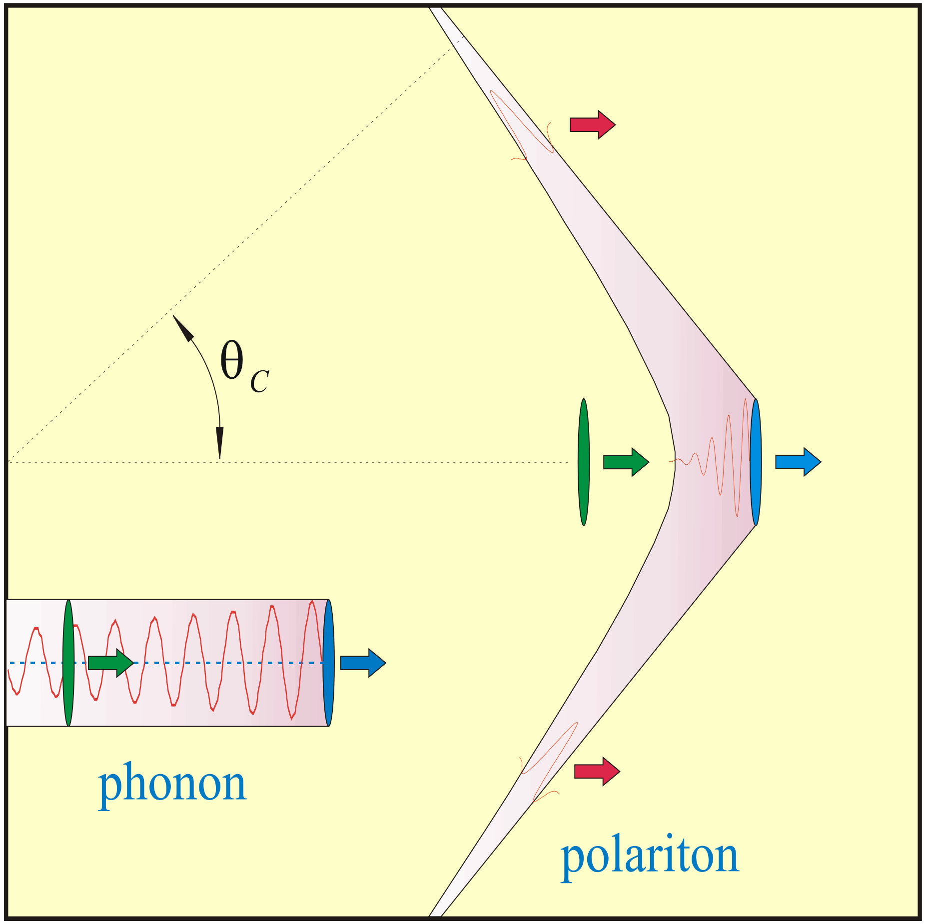
PHONON SQUEEZING



- Optical Phonons
- Acoustic Phonons
- Squeezed Phonons
- Polaritons

PHONON POLARITONS





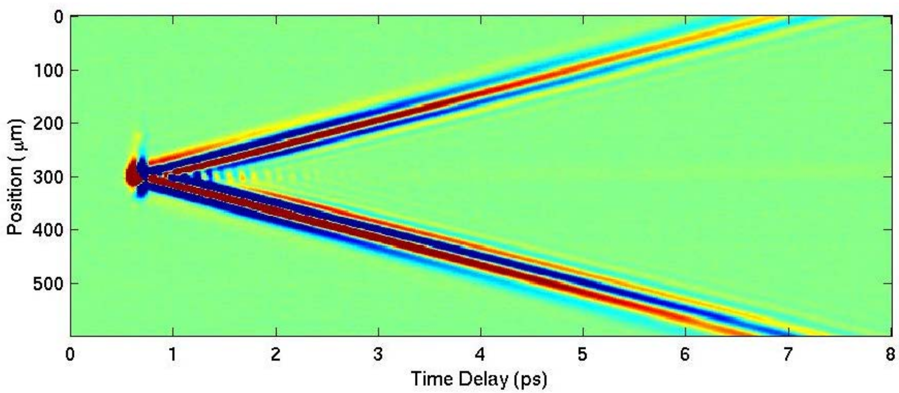
PROPAGATION EFFECTS:

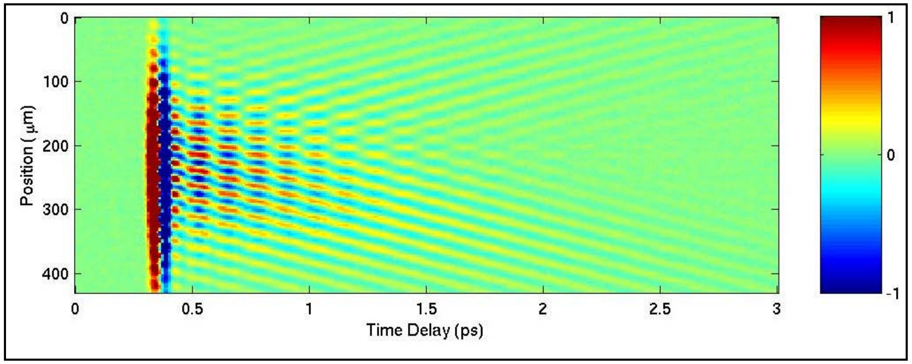
GROUP VELOCITY IN THE VISIBLE IS LARGER THAN PHASE VELOCITY IN THE INFRARED

THE NONLINEAR POLARIZATION GENERATES CHERENKOV RADIATION

PHONON POLARITONS

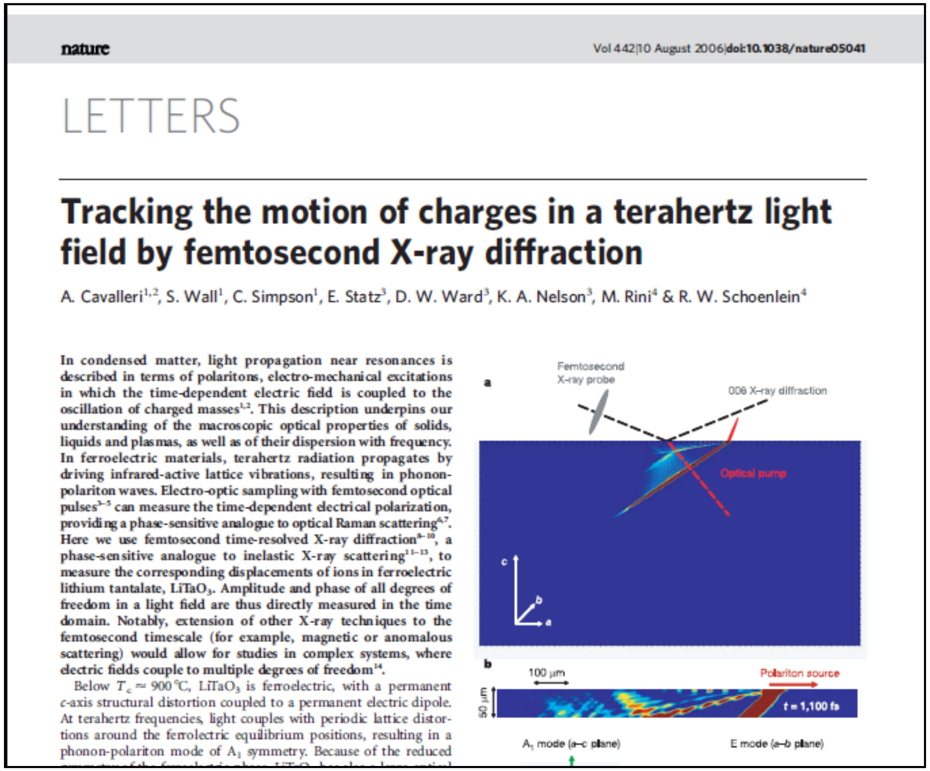
$LiTaO_3$ (superluminal)





J. Wahlstrand and RM, Phys. Rev. B **68**, 054301 (2003)

$LiTaO_3$



nature Vol 442|10 August 2006|doi:10.1038/nature05041

LETTERS

Tracking the motion of charges in a terahertz light field by femtosecond X-ray diffraction

A. Cavalleri[1,2], S. Wall[1], C. Simpson[1], E. Statz[3], D. W. Ward[3], K. A. Nelson[3], M. Rini[4] & R. W. Schoenlein[4]

In condensed matter, light propagation near resonances is described in terms of polaritons, electro-mechanical excitations in which the time-dependent electric field is coupled to the oscillation of charged masses[1,2]. This description underpins our understanding of the macroscopic optical properties of solids, liquids and plasmas, as well as of their dispersion with frequency. In ferroelectric materials, terahertz radiation propagates by driving infrared-active lattice vibrations, resulting in phonon-polariton waves. Electro-optic sampling with femtosecond optical pulses[3–5] can measure the time-dependent electrical polarization, providing a phase-sensitive analogue to optical Raman scattering[6,7]. Here we use femtosecond time-resolved X-ray diffraction[8–10], a phase-sensitive analogue to inelastic X-ray scattering[11–13], to measure the corresponding displacements of ions in ferroelectric lithium tantalate, $LiTaO_3$. Amplitude and phase of all degrees of freedom in a light field are thus directly measured in the time domain. Notably, extension of other X-ray techniques to the femtosecond timescale (for example, magnetic or anomalous scattering) would allow for studies in complex systems, where electric fields couple to multiple degrees of freedom[14].

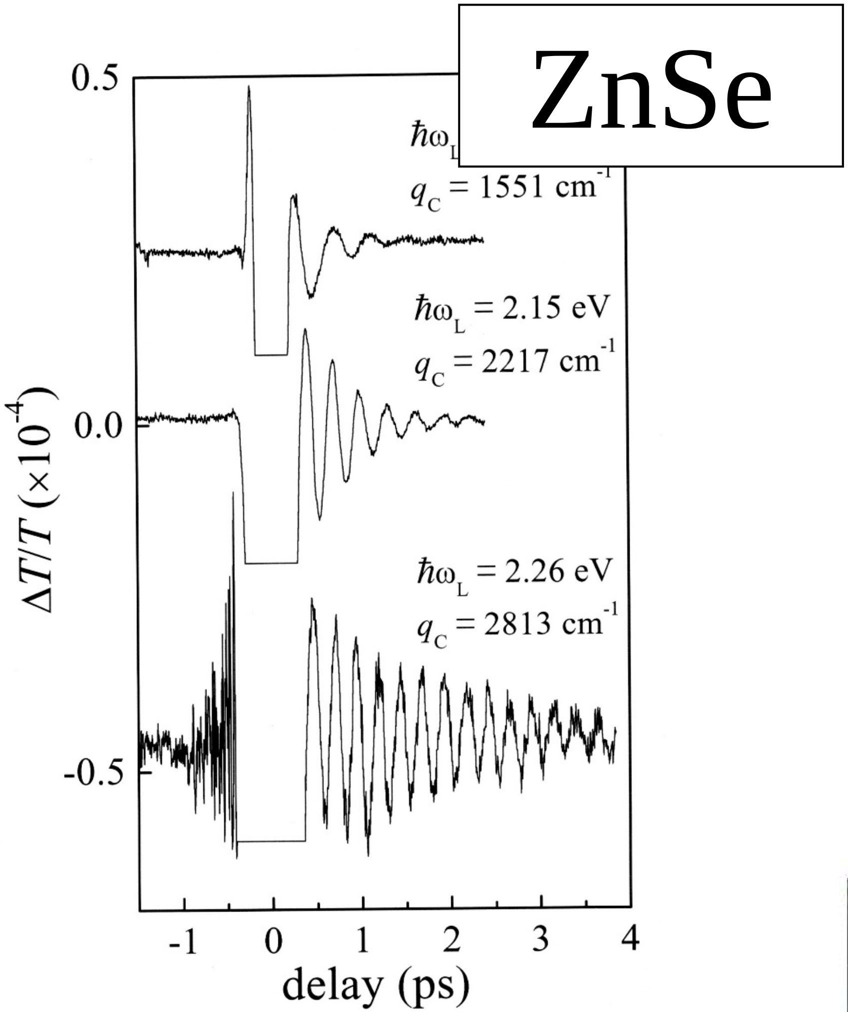
Below $T_c \approx 900\,°C$, $LiTaO_3$ is ferroelectric, with a permanent *c*-axis structural distortion coupled to a permanent electric dipole. At terahertz frequencies, light couples with periodic lattice distortions around the ferrolectric equilibrium positions, resulting in a phonon-polariton mode of A_1 symmetry. Because of the reduced

A. Cavalleri et al., Nature **442**, 664 (2006)

PUMP = LIGHT

PROBE = X-RAYS

PHONON POLARITONS



Cherenkov Radiation at Speeds Below the Light Threshold: Phonon-Assisted Phase Matching

T. E. Stevens,[1,2]* J. K. Wahlstrand,[1] J. Kuhl,[2] R. Merlin[1]†

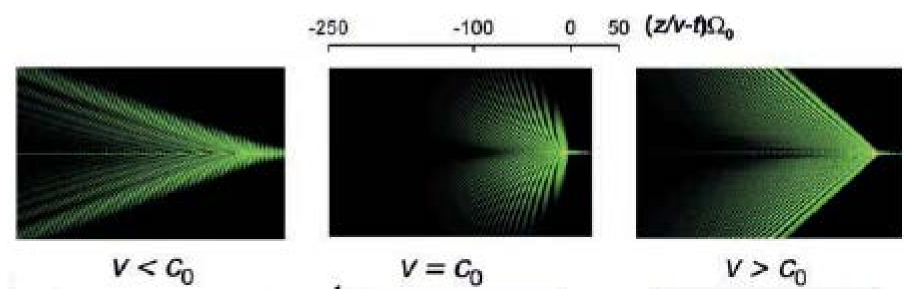
Charged particles traveling through matter at speeds larger than the phase velocity of light in the medium emit Cherenkov radiation. Calculations reveal that a given angle of the radiation conical wavefront is associated with two velocities, one above and one below a certain speed threshold. Emission at subluminal but not superluminal speeds is predicted and verified experimentally for relativistic dipoles generated with an optical method based on subpicosecond pulses moving in a nonlinear medium. The dipolar Cherenkov field, in the range of infrared-active phonons, is identical to that of phonon polaritons produced by impulsive laser excitation.

Cherenkov radiation (CR) is extensively used in experiments for counting and identifying relativistic particles (*1*, *2*). The effect derives its name from Pavel Cherenkov (*3*), who, following a suggestion by Vavilov (*4*), discovered in 1934 that substances exposed to fast electrons emit coherent light (*5*). The theory of CR was developed by Frank and Tamm in 1937 (*6*). They showed that charges traveling faster than the speed of light in a substance with a frequency-independent re-

of a resonance at frequency Ω_0 can be approximated by the nondissipative Lorentz form

$$\varepsilon(\Omega) = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{1 - (\Omega/\Omega_0)^2} \qquad (1)$$

ε_0 is the dielectric constant at $\Omega = 0$, and ε_∞ accounts for the contribution of higher lying excitations. Although analytical expressions for the Cherenkov field of a generic dielectric have been available for a



T. Stevens, J. Wahlstrand, J. Kuhl and RM, Science **291**, 627 (2001)