光誘起キャリアとコヒーレントフォノンの超高速ダイナミクス

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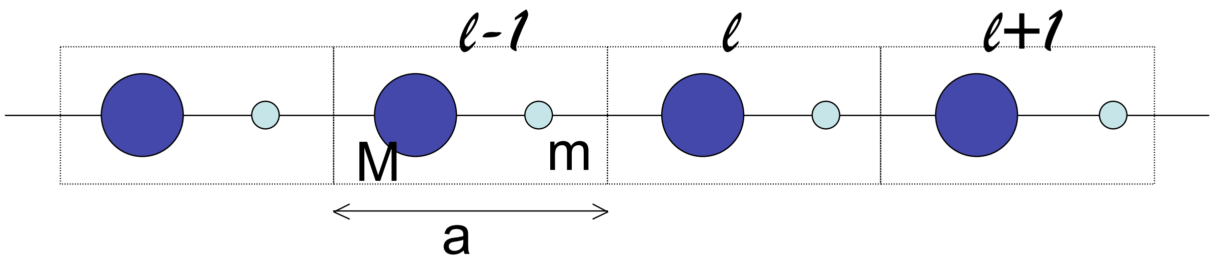
Outline

- イントロダクション（コヒーレントフォノン）
- パルスX線発生
- 時間分解X線回折測定
  - ピコ秒時間スケール：GaAs音響フォノンパルス伝搬
  - フェムト秒時間スケール：CdTe光学フォノン振動
- まとめ

フォノン（2原子直線格子）



$$M\frac{d^2U_l}{dt^2} = f(u_l + u_{l+1} - 2U_l)$$

$$m\frac{d^2u_l}{dt^2} = f(U_l + U_{l+1} - 2u_l)$$

$$\omega_{\pm}^2 = \frac{f(M+m)}{Mm}\left[1 \pm \left\{1 - \frac{4Mm}{(M+m)^2}\sin^2\left(\frac{qa}{2}\right)\right\}^{1/2}\right]$$

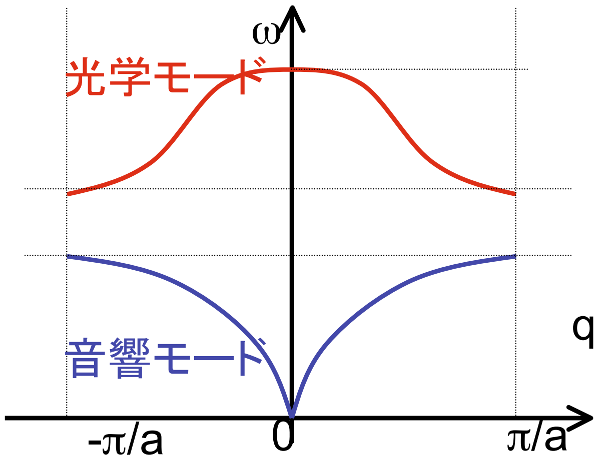
i) ω_のモードはq->0でω_>0となり、 $u_l = u_0 e^{-i\omega t}$

全ての原子は同位相かつ同振幅で振動することにある：音響的振動

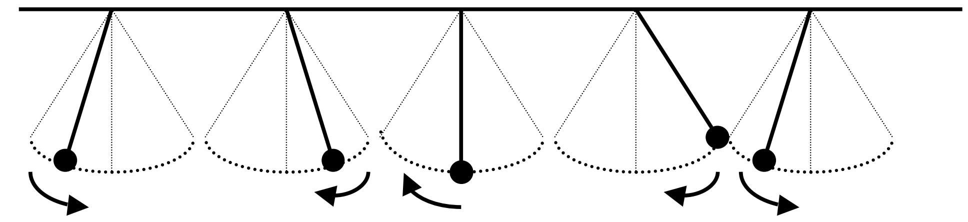
ii) ω_+のモードはq->0で、 $\omega_+^2 = \frac{2f(M+m)}{Mm}$

隣り合った異種原子がその質量に反比例して、逆位相で（反対方向に）振動している

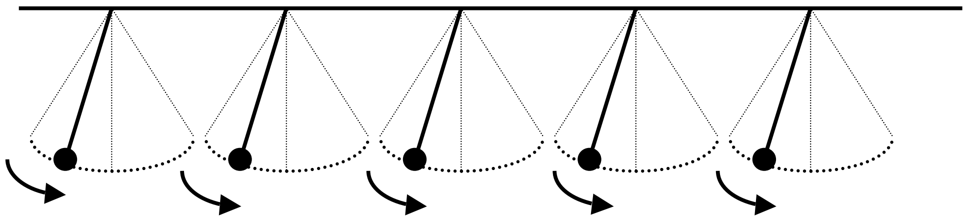
：光学モード



熱振動とコヒーレントフォノン



熱振動：各原子の振動の位相がバラバラ



コヒーレントフォノン：
各原子の振動の位相が揃った状態

格子振動の振動周期よりも十分短いパルス励起で発生することができる

コヒーレントフォノン発生メカニズム

- 発生メカニズム
- DECP(displacive excitation of coherent phonon)
  - 光吸収による励起状態での振動：発生キャリア量に比例した変位
  - 無極性な半金属や無機物質
- ISRS (Impulsive stimulated Raman scattering)
  - 光励起による基底状態での振動：励起光強度の2乗、ラマン活性なモード
  - 吸収が無い物質
- USSC(ultrafast screening of space-charge field)
  - 光吸収により発生したキャリアによる表面電荷層のスクリーニング（バンドベンディングの減少）
  - 極性のある半導体

一般的計測方法：
フェムト秒レーザーの過渡反射率・過渡透過率測定

コヒーレントフォノンコントロール（Bi）

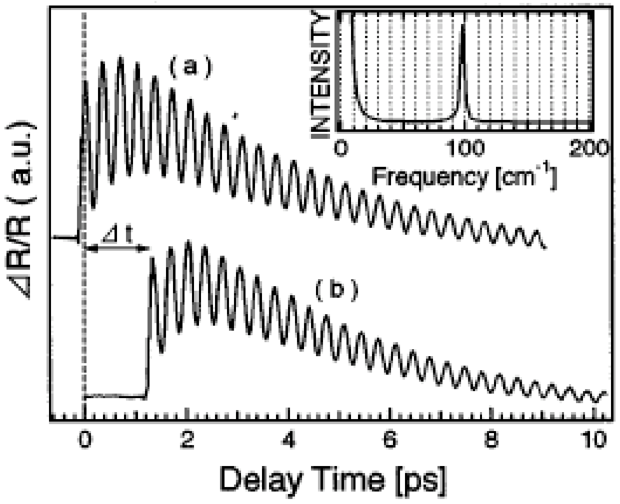


FIG. 1. Reflectivity change for Bi induced by two excitation pulses: (a) Reflectivity change induced by the first excitation pulse alone; (b) reflectivity change induced by the second excitation pulse alone. Inset: Fourier transform power spectrum of the time domain data.

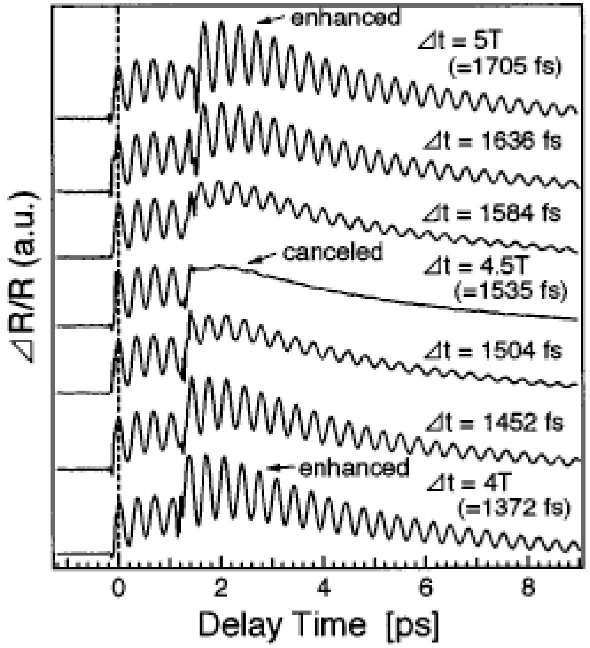
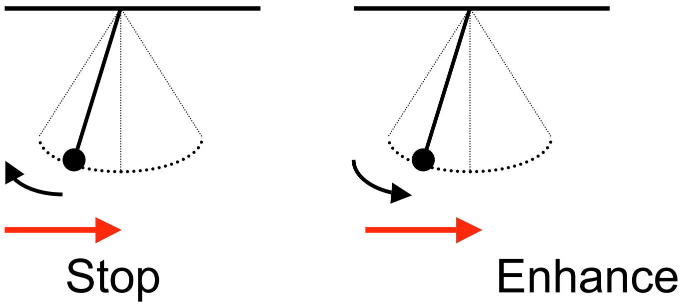


FIG. 2. Double-pulse pump–probe data observed in a Bi film at room temperature. Δt is the time difference between two pulse components of the double-pulse excitation.



Hase et al. Appl. Phys. Lett., 69 (1996) 2474

- 光学測定を用いたコヒーレントフォノンの計測でフォノンの寿命、電子フォノン相互作用が研究されている。
- パルス波形整形によりコヒーレントフォノンを制御し、光による構造制御を目指した研究行われている。

↓

光学測定では、原子変位量や構造がよく分からない

↑

超高速時間分解X線回折を用いたコヒーレントフォノン研究

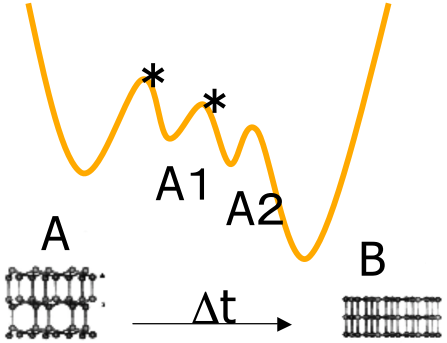
Time-resolved measurement of coherent phonon

- Coherent phonon has been studied extensively using several optical measurement techniques: time-domain spectroscopy
  - reflection or transmission, SHG, THz radiation
    - T.K. Cheng, APL 59 (1991) 1923
    - M. Hase et al. PRL 88 (2002) 067401.
- Lattice displacement has been directly studied with X-ray diffraction by several groups
  - Acoustic phonon propagation
    - A. Rousse et al. Nature 410 (2001) 65
  - Coherent optical phonon
    - K. Sokolowski-Tinten et al. Nature 420 (2003) 287.

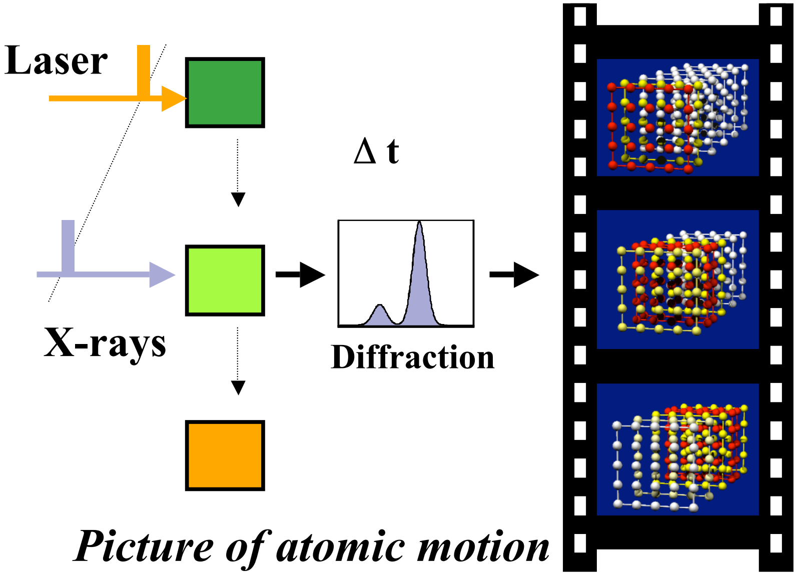
Motivation: Materials Dynamics

Elucidate dynamics of phase transition of condensed matter and control

- Transition time (Δt)?
- Pathway of structural changes (A1-A2) ?
- Transition structure (*) ?



Rate constant



Picture of atomic motion

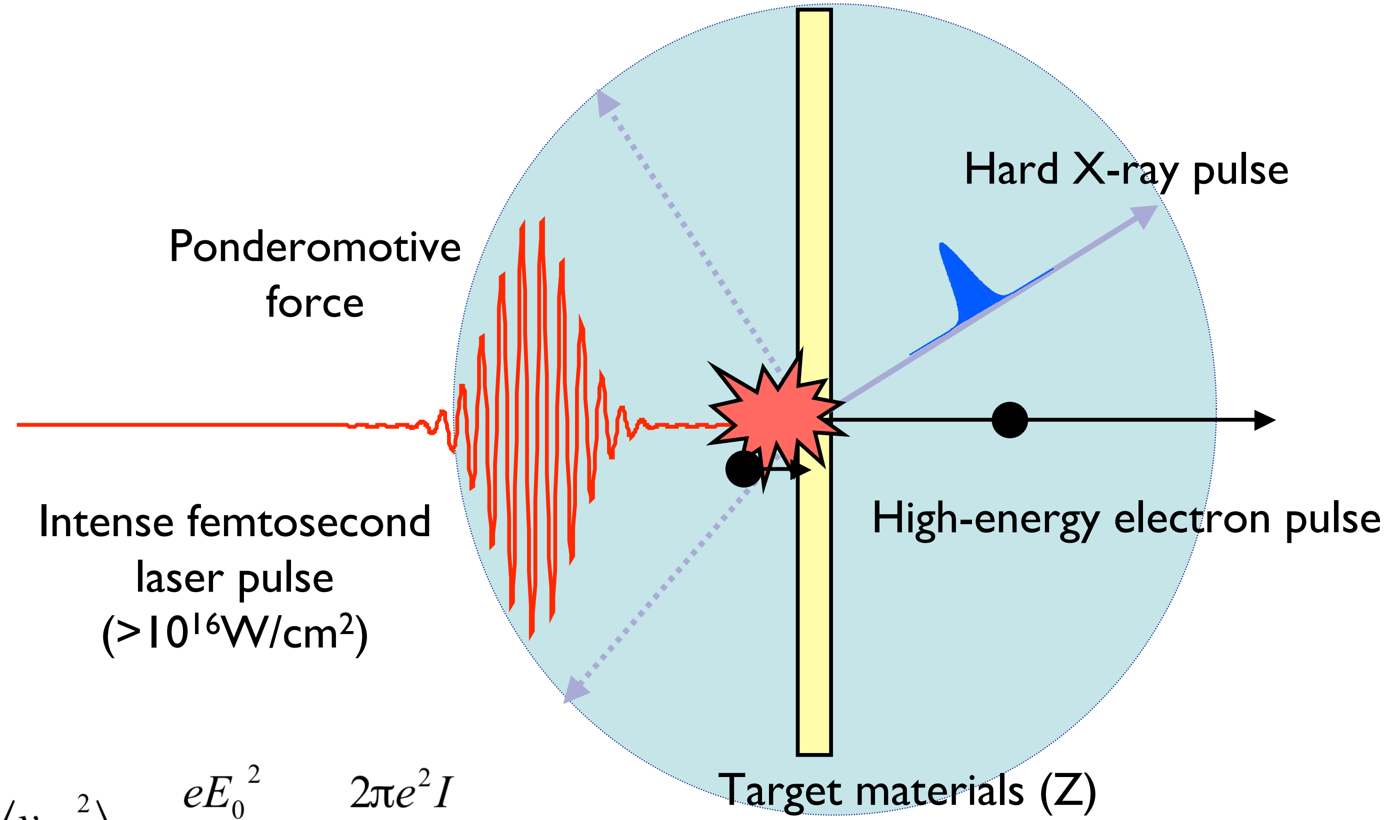
How

Structural change is directly monitored by using ultrafast X-ray diffraction with pump and probe technique

Ultrashort pulsed X-rays are required!

Quantum emissions

High-energy short-pulsed beams of electrons, ions and photons (X-rays) generated from femtosecond intense laser field (>10^{16}W/cm^2).



Ponderomotive force

Intense femtosecond laser pulse (>10^{16}W/cm^2)

Hard X-ray pulse

High-energy electron pulse

Target materials (Z)

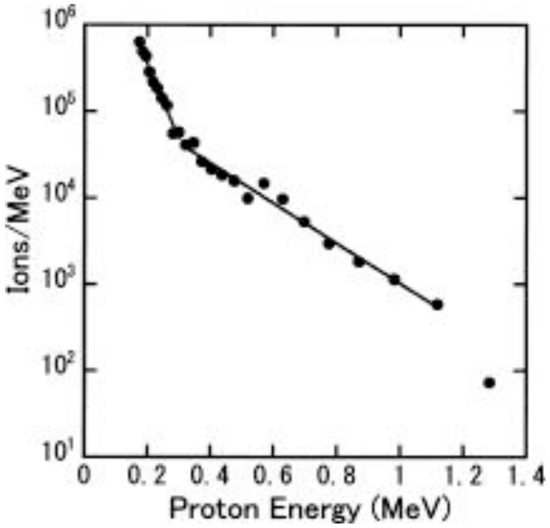
$$U_{\mathrm{p}} = \frac{1}{2} m_e \langle v_{\mathrm{osc}}{}^2 \rangle = \frac{eE_0{}^2}{4m_e\omega^2} = \frac{2\pi e^2 I}{m_e c\omega^2}$$

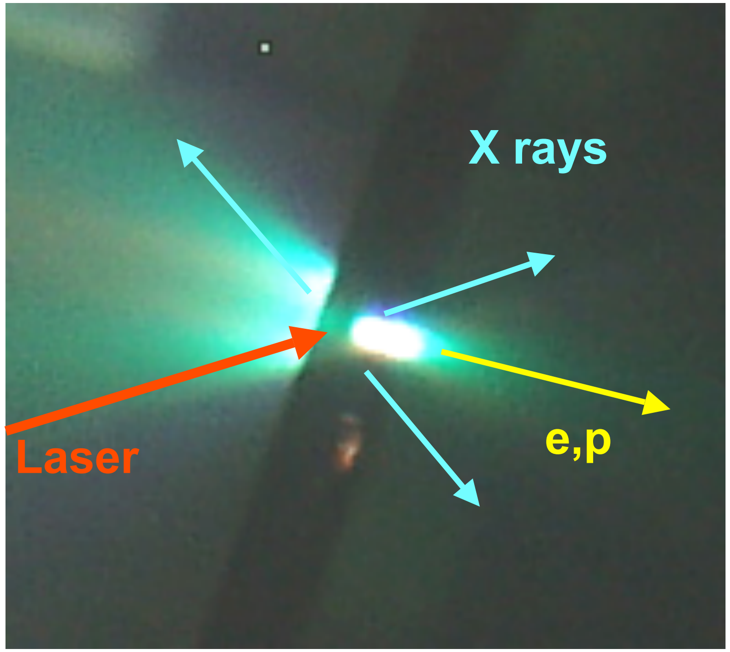
$$= 9.3\times10^{-14} I(\mathrm{W/cm}^2)\lambda(\text{ìm})^2$$

Quantum emission

High-energy short-pulsed beams of electrons, ions and photons (X-rays) generated from femtosecond intense laser field (>10^{16}W/cm^2).

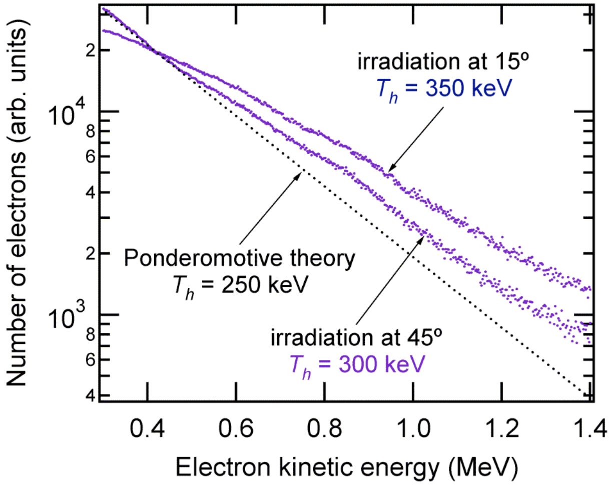
protons





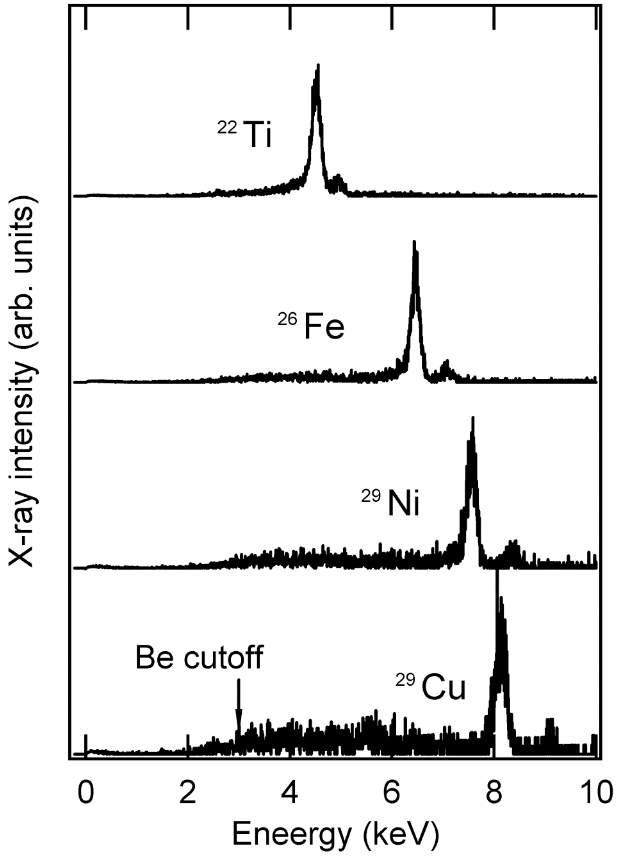
Cu-foil at I=2x10^{18}W/cm^2

electrons



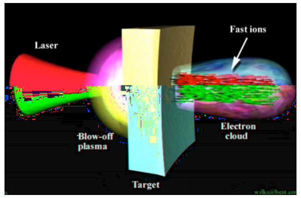
Femtosecond laser induced Hard X-ray pulse ($I = 10^{17}$ W/cm^2)

Energy spectra

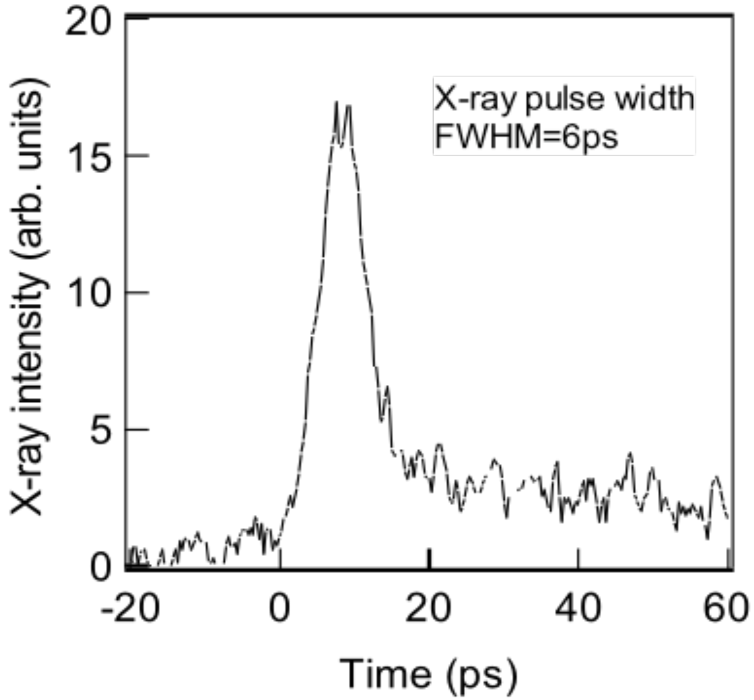


Characteristic X-rays obtained by X-ray CCD camera at power density of 3×10^{16} W/cm^2.

Pulse width of CuKa



6.5×10^{10} photons/4πsr/pulse



The temporal profiles obtained by the X-ray streak camera. The energy extends from 1 to 10 keV are accumulated. $I \sim 10^{17}$ W/cm^2

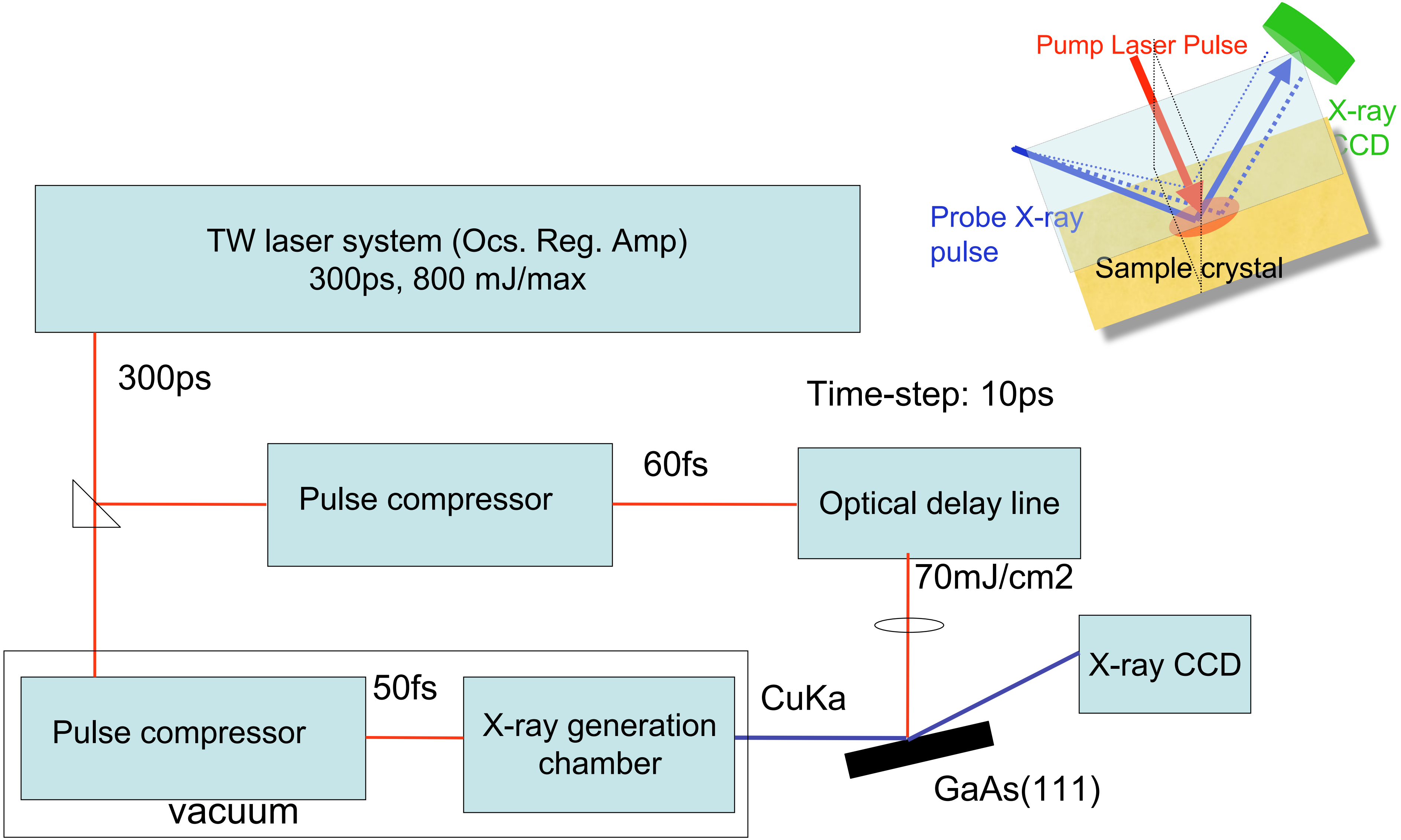
M. Yoshida *et al.* APL 73 (1998) 2393; Y. Fujimoto *et al.* JJAP 38 (1999) 6754

Picosecond time-resolved X-ray diffraction from femtosecond laser-excited GaAs(111):

Acoustic phonon propagation

Ref: C. Rose-Petruck et al., Nature 398 (1999) 310
H. Kishimura et al., JCP 117 (2002)10239

Schematic of experimental setup



TW laser system (Ocs. Reg. Amp)
300ps, 800 mJ/max

300ps

Pulse compressor

60fs

Optical delay line

Time-step: 10ps

70mJ/cm2

X-ray CCD

CuKa

GaAs(111)

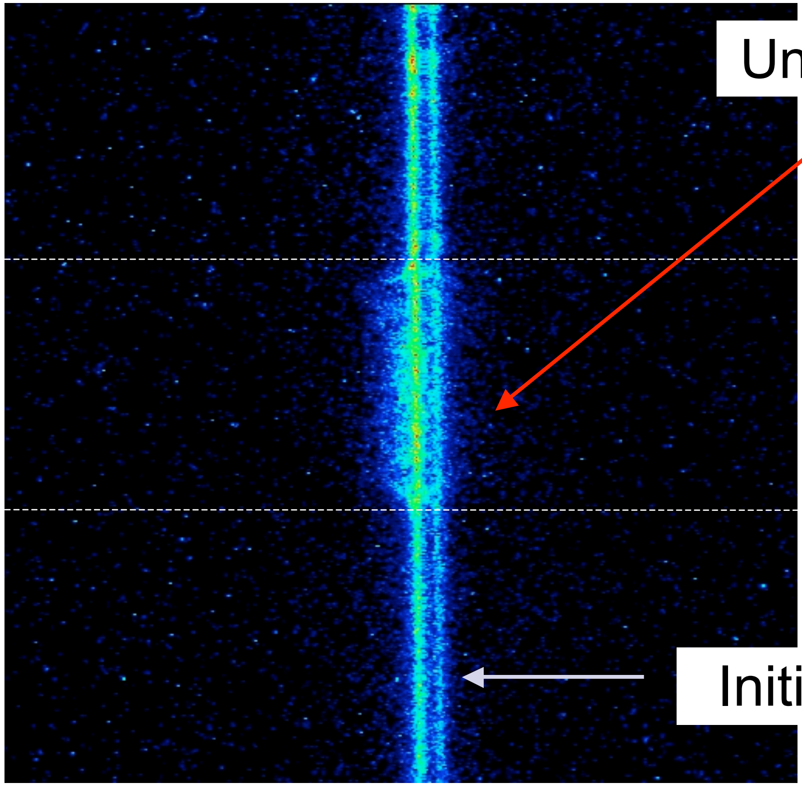
Pulse compressor

50fs

X-ray generation chamber

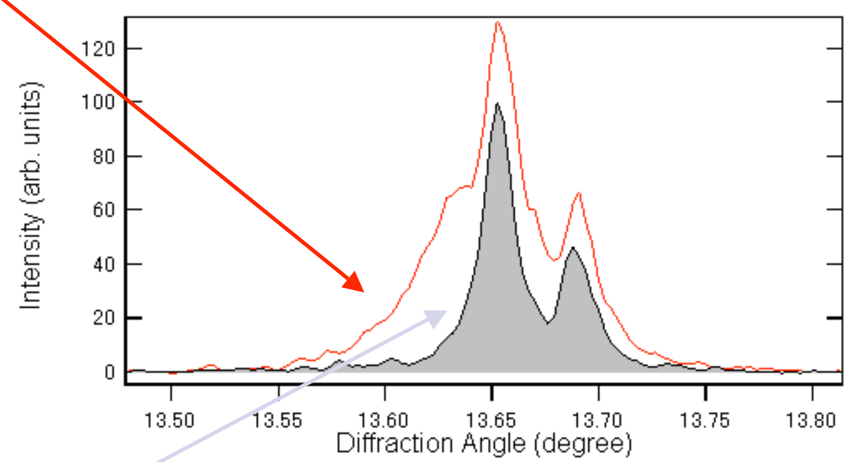
vacuum

CCD image and rocking curve of X-ray diffraction from laser irradiated GaAs(111)



Under laser irradiated

Initial state



Diffraction Angle

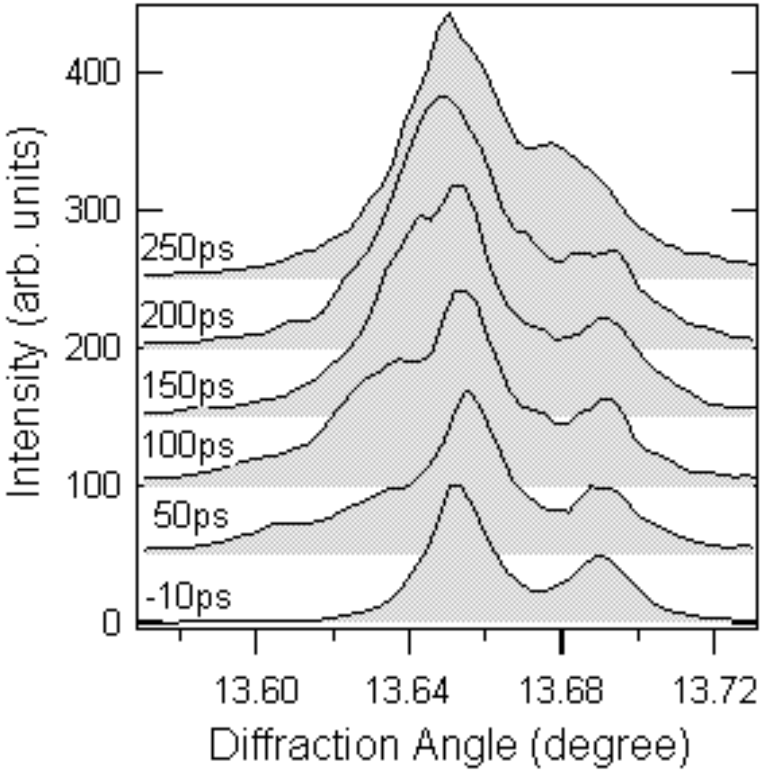
CCD camera image

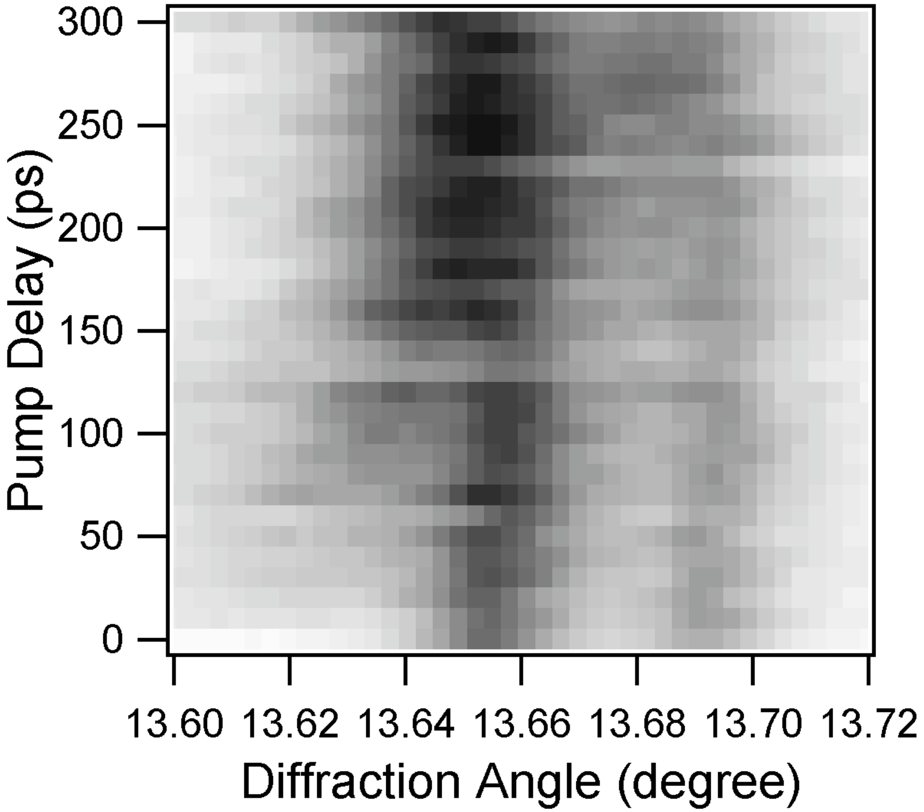
(300shot integrated)

2d sinθ = λ

A low angle shift→Expansion
A high angle shifts→Compression

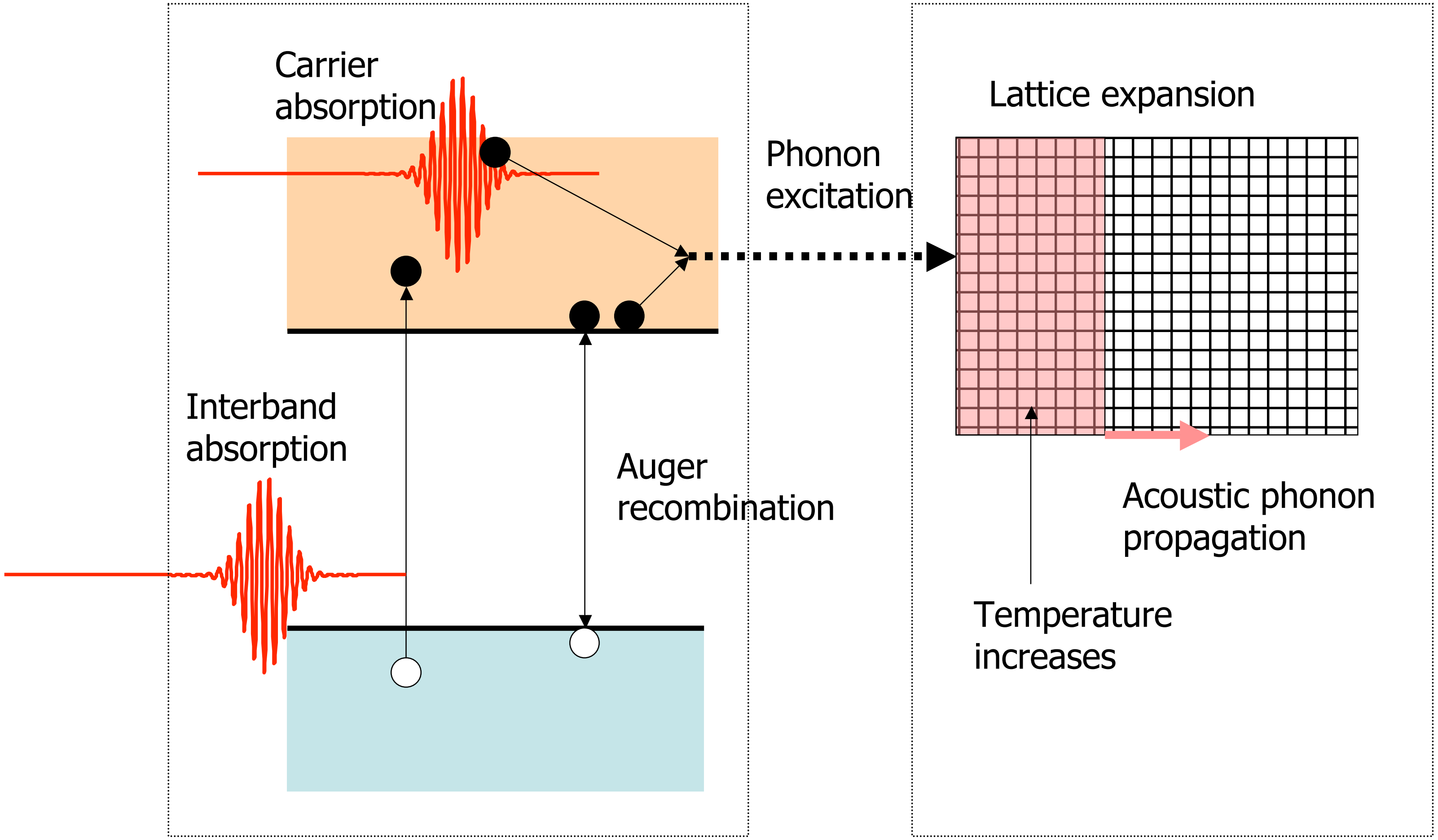
Picosecond time-resolved X-ray diffraction from 60-fs laser irradiated GaAs(111)





Electronic system

Lattice system



Carrier absorption

Phonon excitation

Lattice expansion

Interband absorption

Auger recombination

Acoustic phonon propagation

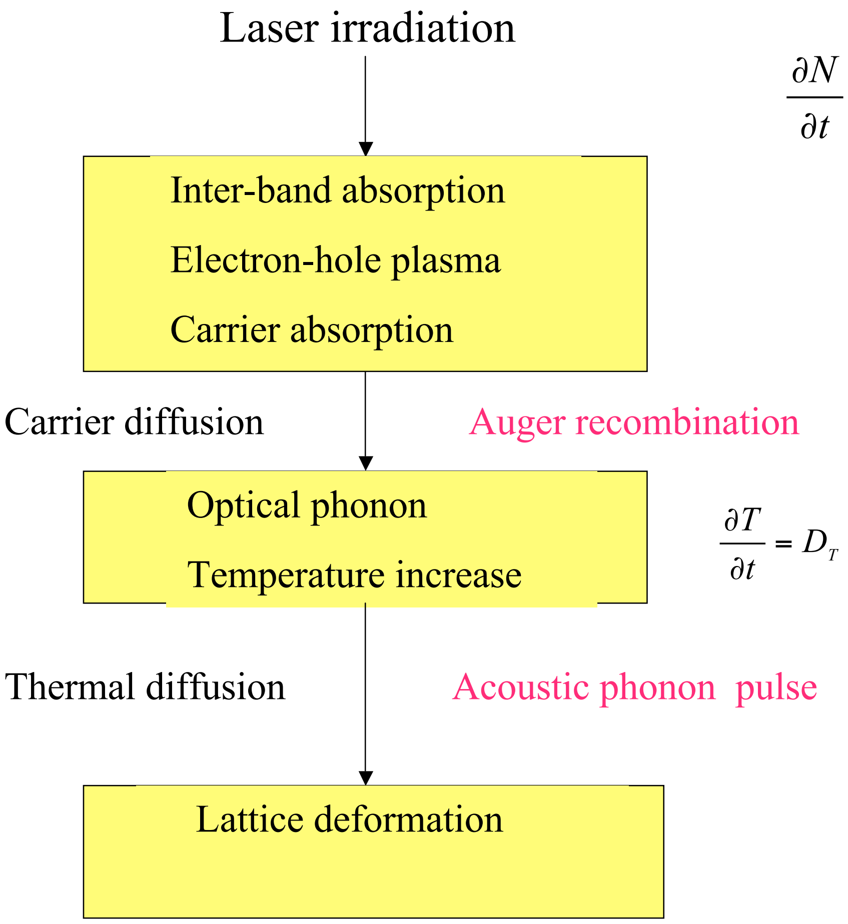
Temperature increases

Rate equations: N(x,t), T(x,t)

Diffusion equations: T(x,t)

Hydro-dynamic equations: u(x,t)

Model of laser heating and phonon generation



Laser irradiation

Inter-band absorption

Electron-hole plasma

Carrier absorption

Carrier diffusion

Auger recombination

Optical phonon

Temperature increase

Thermal diffusion

Acoustic phonon pulse

Lattice deformation

$$\frac{\partial N}{\partial t} + \frac{N}{\tau_R} = D\frac{\partial^2 N}{\partial x^2} + \frac{\alpha(1-R)}{E} I(x,t) + \frac{\beta(1-R)^2}{2E} I(x,t)^2$$

$$\frac{\partial}{\partial t} I(x,t) = -\left[\alpha + \Theta N + \beta I(x,t)\right] I(x,t)$$

Rate eq., diffusion eq.

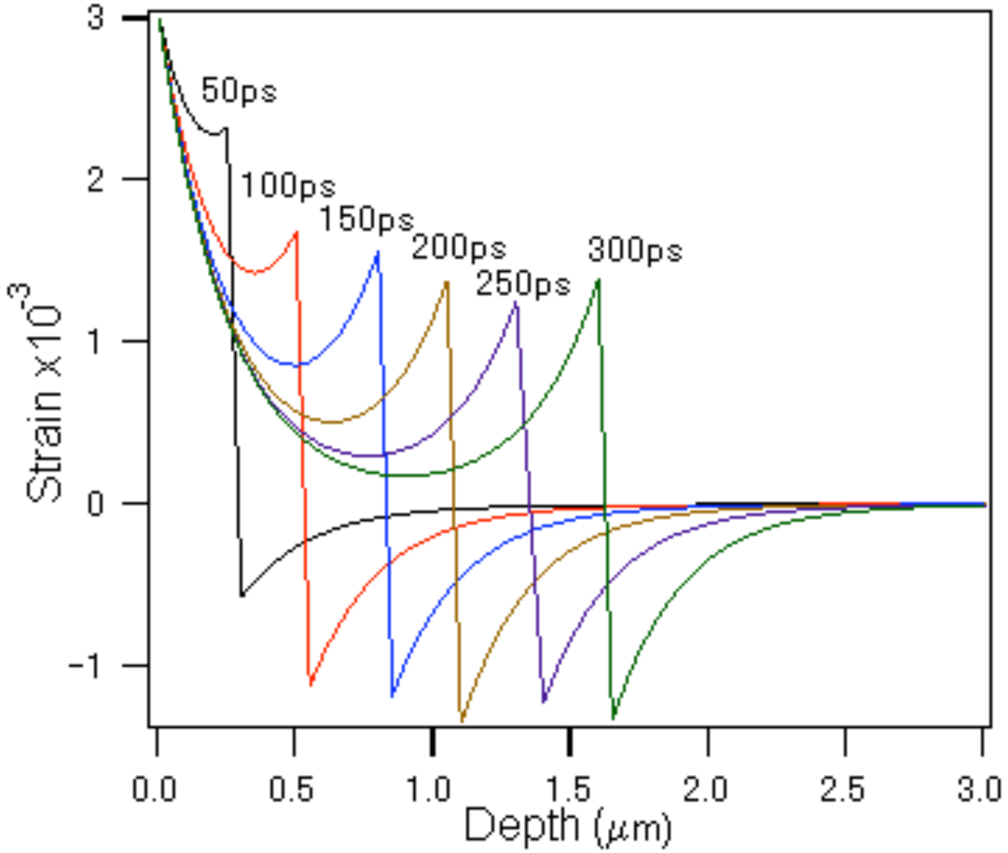
$$\tau_R = \frac{1}{\gamma N^2}$$

$$\frac{\partial T}{\partial t} = D_T \frac{\partial^2 T}{\partial x^2} + \frac{E_g N}{C_v \tau_R} + \left(\frac{E - E_g}{C_v}\right)\left[\left(\frac{\alpha(1-R)}{E} + \Theta N\right) + \frac{\beta(1-R)^2}{2E} I(x,t)\right] I(x,t)$$

Hydro-dynamics

$$\rho \frac{\partial^2 u}{\partial t^2} = \rho v^2 \frac{\partial^2 u}{\partial z^2} + B \frac{\partial E_g}{\partial P}\frac{\partial N}{\partial z} + 3B\beta \frac{\partial T}{\partial z}$$

Strain generated by impulsive laser excitation



$$\eta = \eta_0 \left[e^{-\frac{z}{\xi}} \left(1 - \frac{1}{2} e^{-\frac{vt}{\xi}} \right) - \frac{1}{2} e^{-\frac{|z-vt|}{\xi}} sgn(z - vt) \right]$$

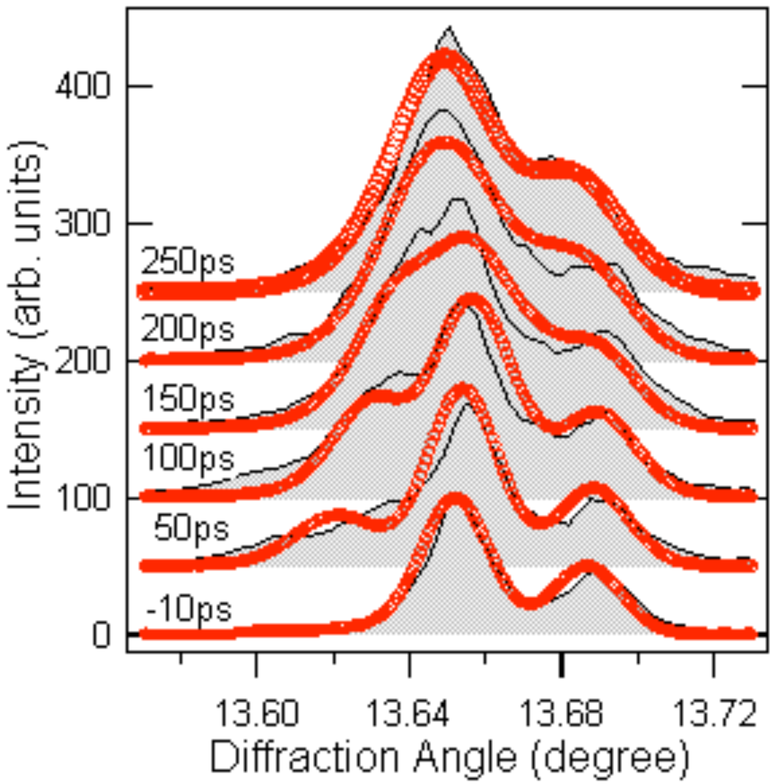
η: strain

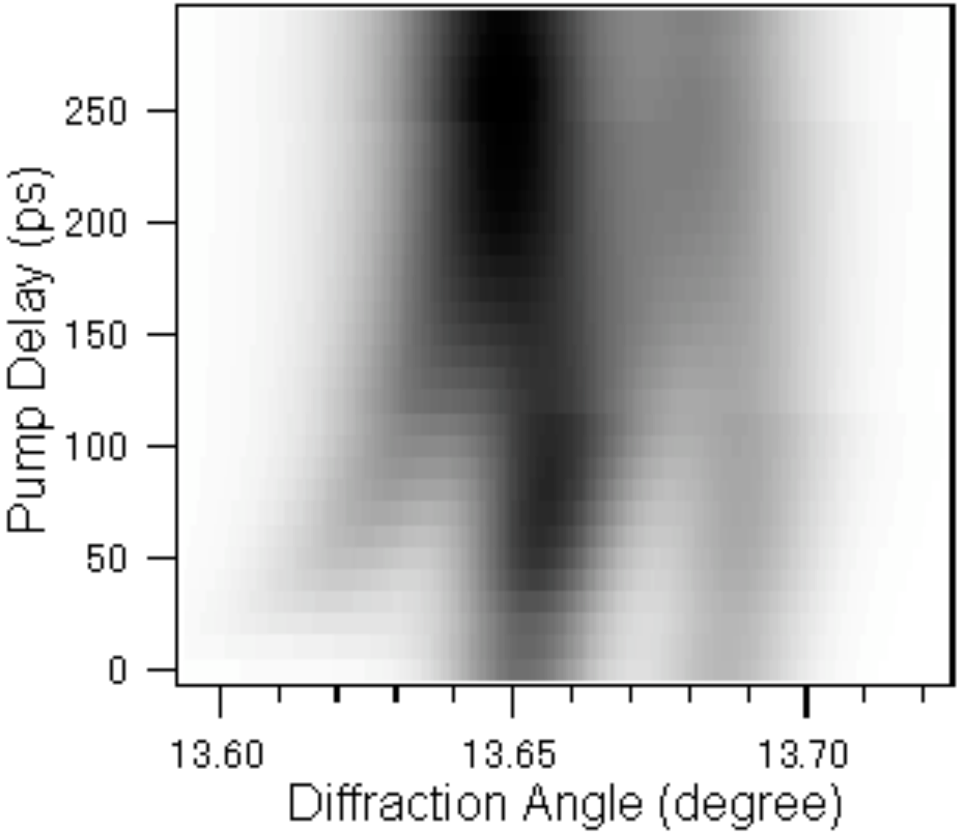
V: sound velocity

ξ: absorption length

C.Thomsen, *et al.*, Phys. Rev. B **34**, 4129 (1986)

Simulation of strain profile inside the GaAs(111) crystal





Maximum strain	**0.30%**
Absorption length	**255nm**

Obtained effective absorption length in this study: ξ= 255 nm

c.f. linear absorption length of GaAs at 800nm: ξ= 900 nm

$$\xi = \frac{1}{\alpha}$$

$$\alpha = \alpha_0 + I\beta$$

I: Intensity
α: An absorption coefficient
α_0: A linear absorption coefficient
β: A two-photon absorption coefficient

Intensity I = 1.17 GW/cm^2

→ A two-photon absorption coefficient β= 27±11 cm/GW

c.f. β= 26±8 cm/GW (using ps laser pulse)

(T.F.Boggess, *et al.*, IEEE J. Quantum Electron. **21**, 5 (1985))

Femtosecond time-resolved X-ray diffraction from femtosecond laser-excited CdTe:

Coherent optical phonon

Ref: K. Sokolowski-Tinten et al., Nature 422 (2003) 287 and (2006): Bi-film
Y. Hironka et al., Ultrafast Phenomena (2006)

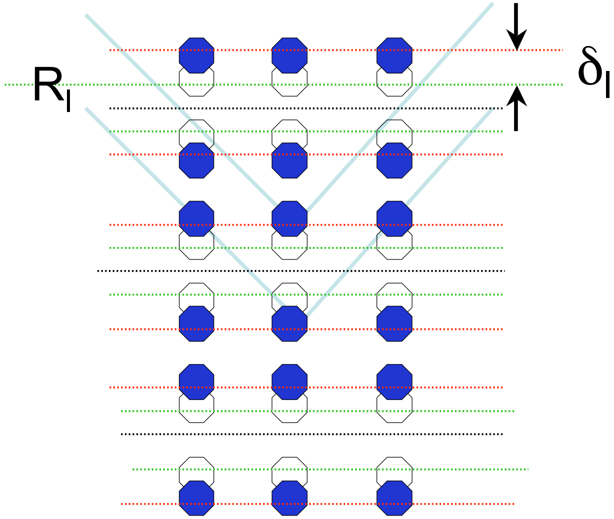
Effect of coherent optical phonons in ultrafast time-resolved X-ray diffraction

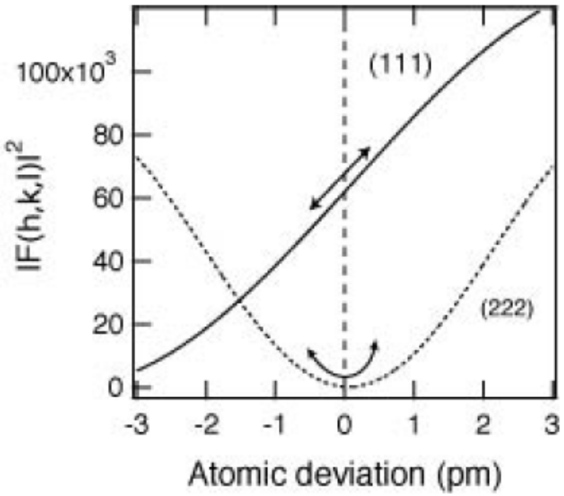
- No change in diffraction angle
- Diffraction intensity changes as a change of structural factor: F

$$|F|^2 = FF^* = I\sum_l f_l f_{l'} \exp\left(\frac{2\pi i}{\lambda}(S-S_0)(R_l - R_{l'})\right)\exp\left(\frac{2\pi i}{\lambda}(S-S_0)(\delta_l - \delta_{l'})\right)$$

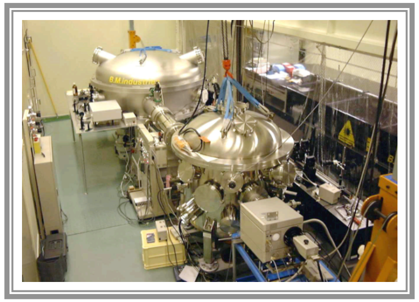
$$\approx |F_0|^2 + \sum_l iQ\exp(i\omega t + \theta_l)$$

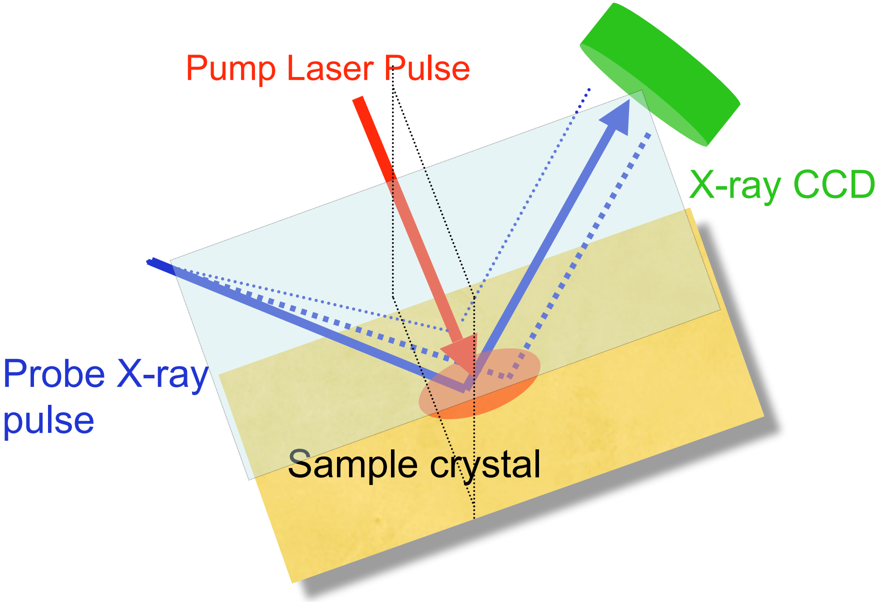
(Thermal: Debye-Waller factor)





Experimental geometry and diffraction image

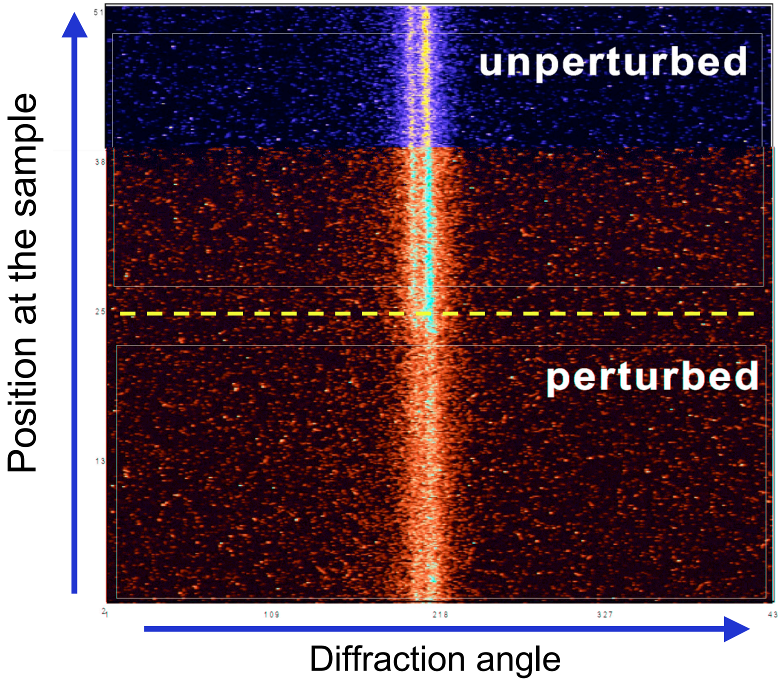




Symmetric Bragg diffraction geometry

2dsinθ=λ

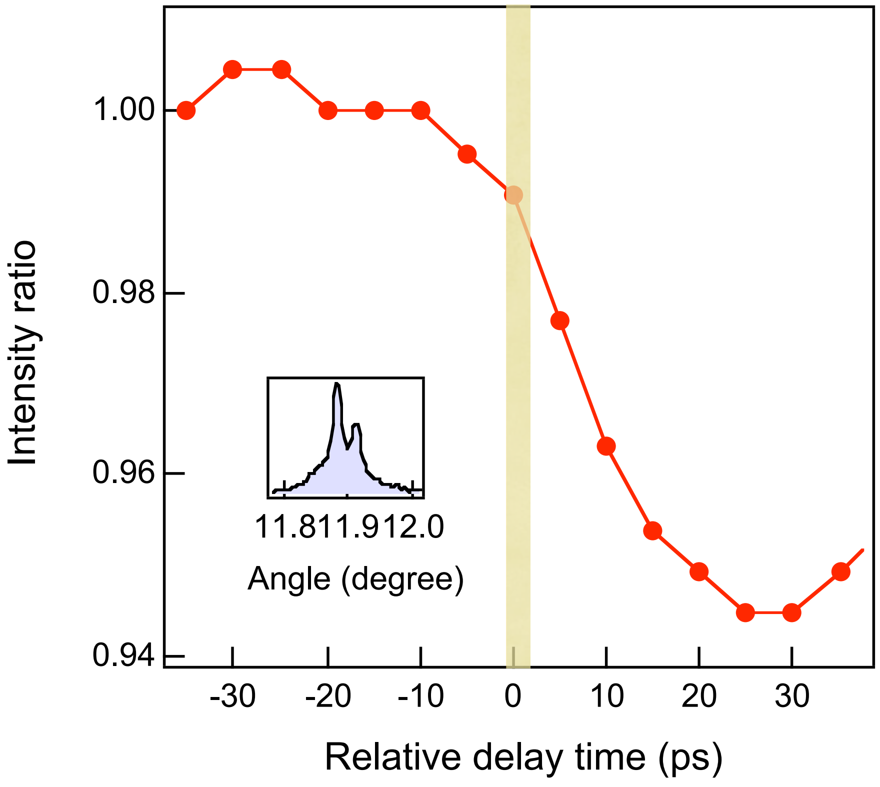
CCD image of X-ray diffraction



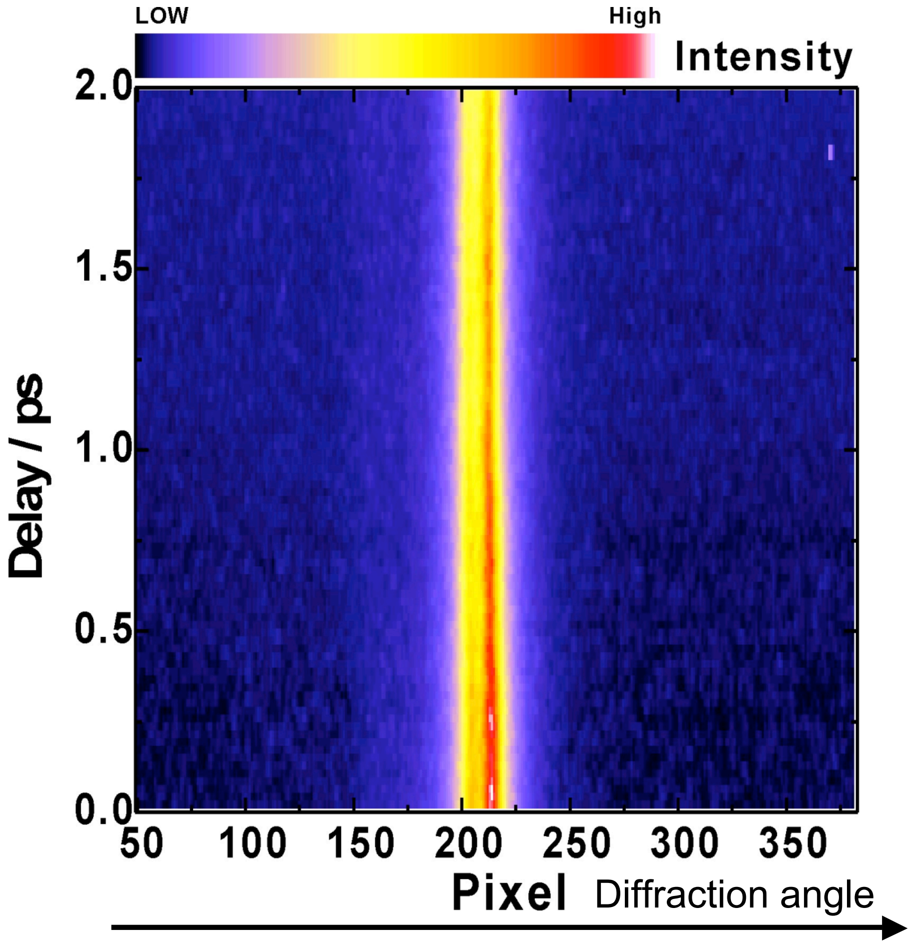
Reflecting Plane : CdTe(111)

θ_B=11.88384 deg. θ_B=11.9138 deg

fs-TRXD from 50-fs laser irradiated CdTe(111)

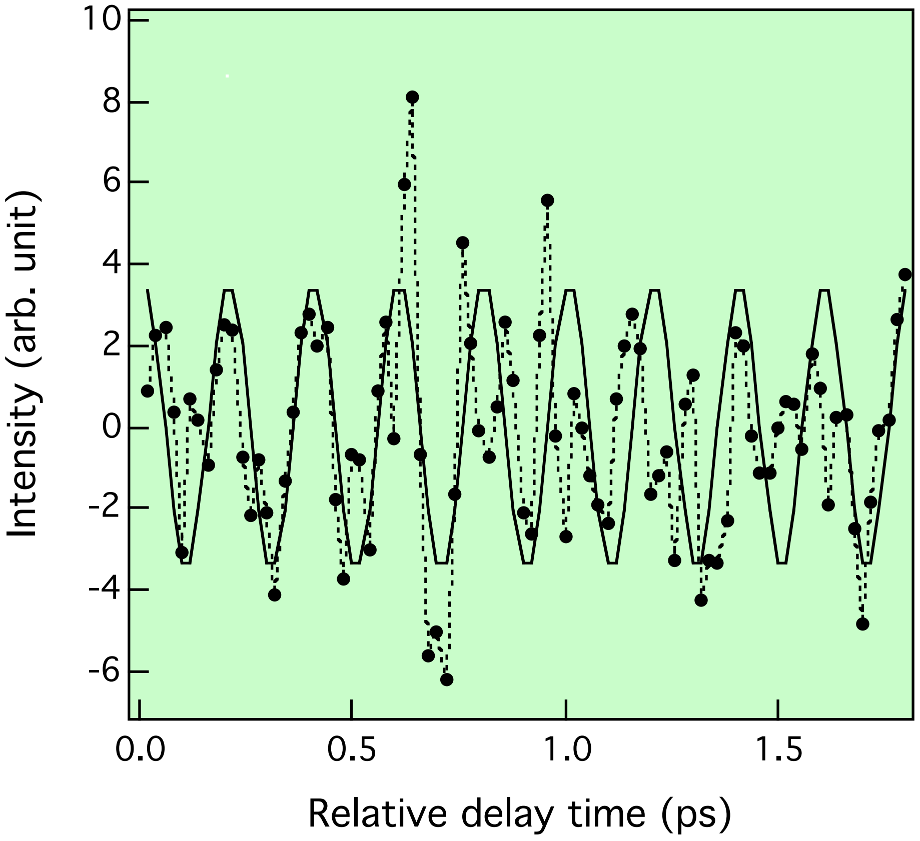


Temporal profile of diffracted X-ray intensity

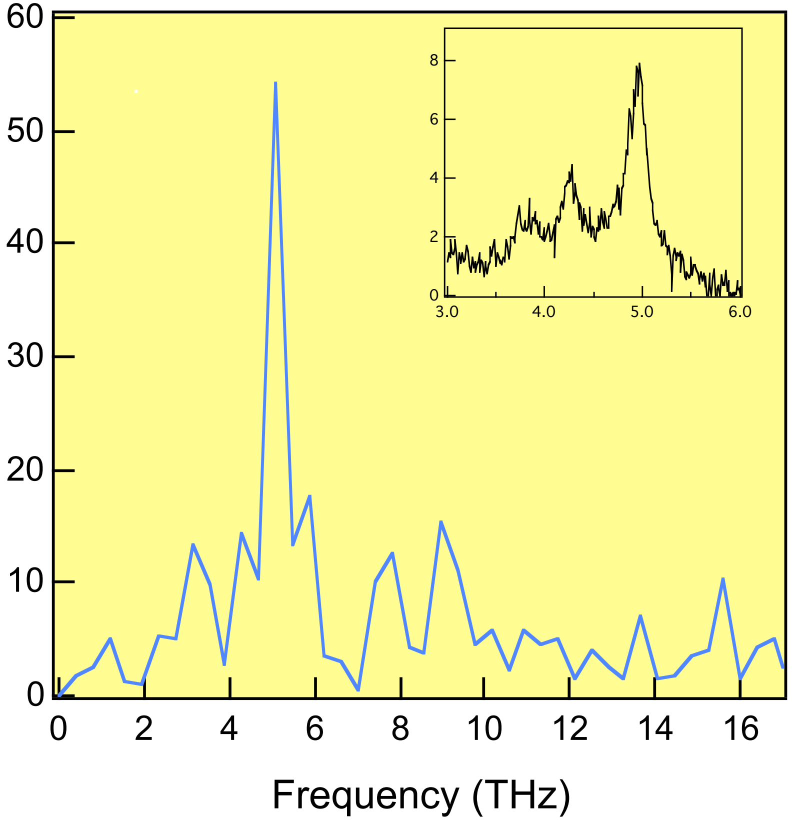


No shift of the diffraction peak but modulation of diffracted X-ray intensity

Coherent optical phonons in CdTe(111):fs-TRXRD



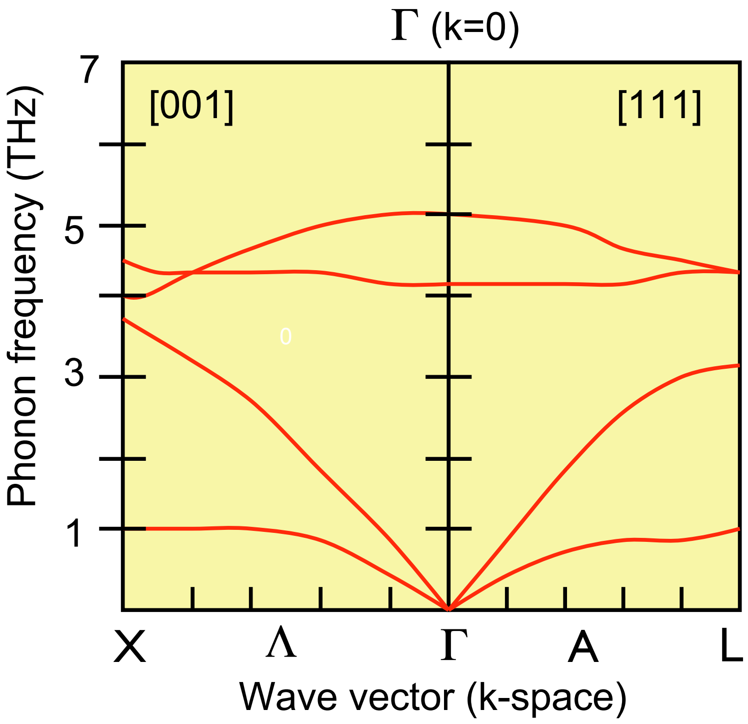
Modulation in X-ray diffraction intensity



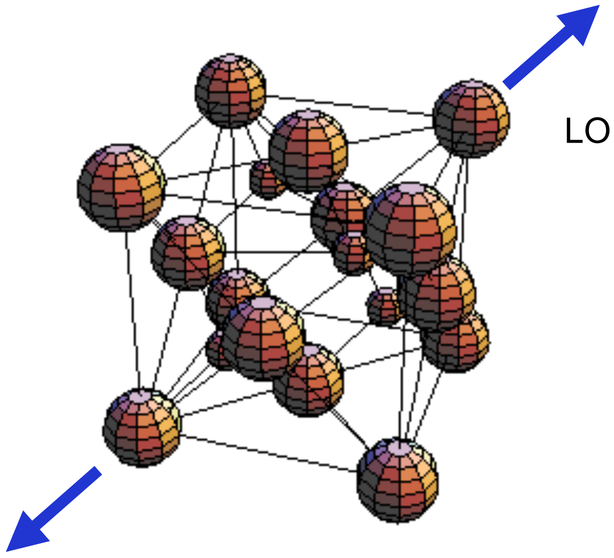
Fourier power spectrum of modulation in X-ray diffraction intensities and normal Raman spectrum of CdTe (inset).

Phonon dispersion of CdTe

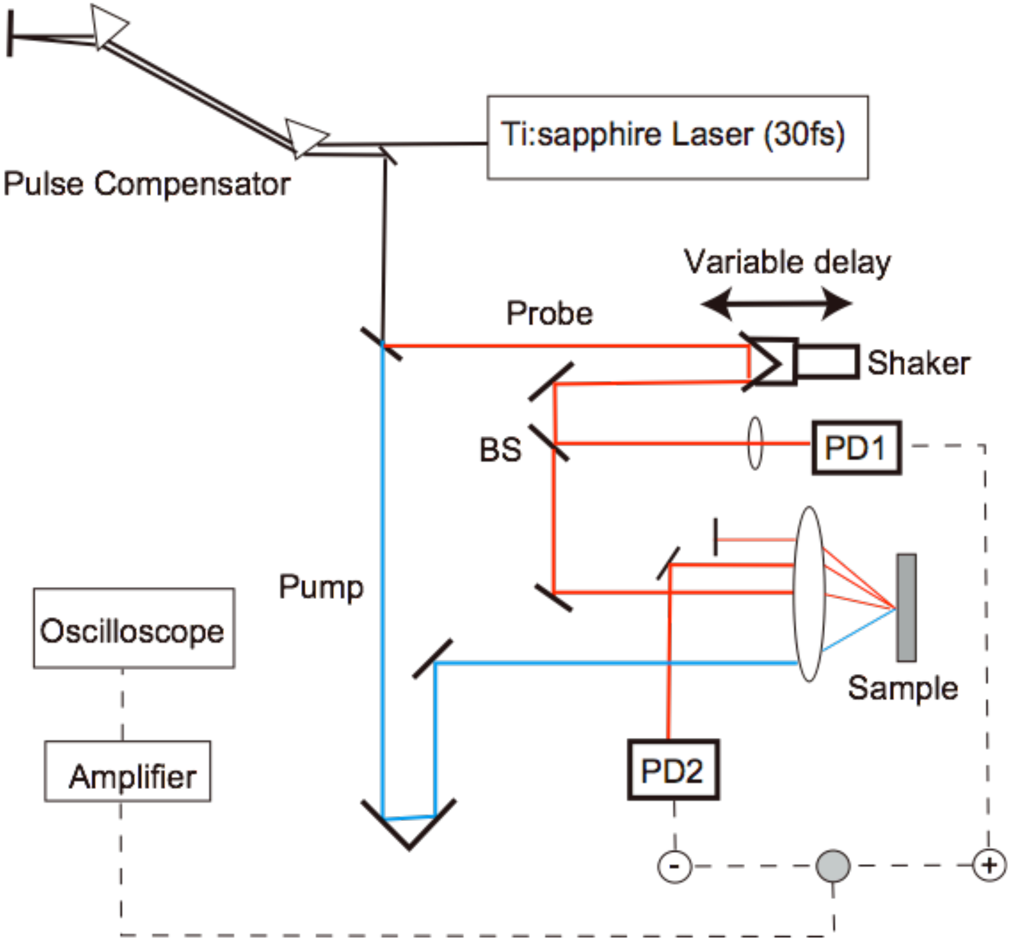
Phonon dispersion curve of CdTe

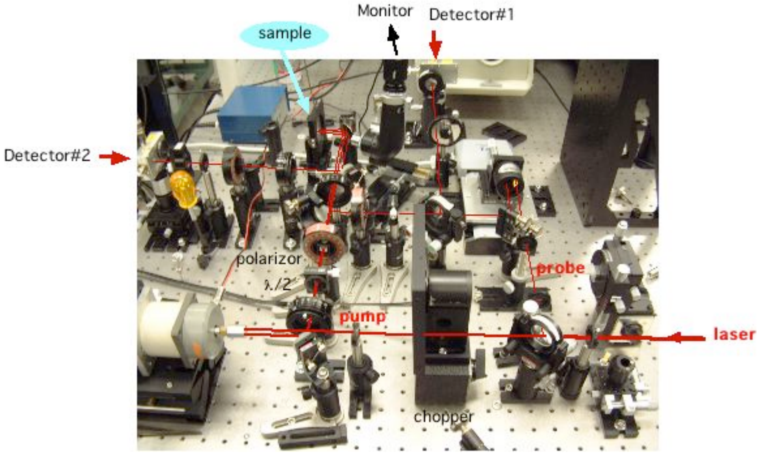


From J.M. Rowe et al. PRB10(1974)



Experimental setup for fs time-resolved optical reflection @NIMS

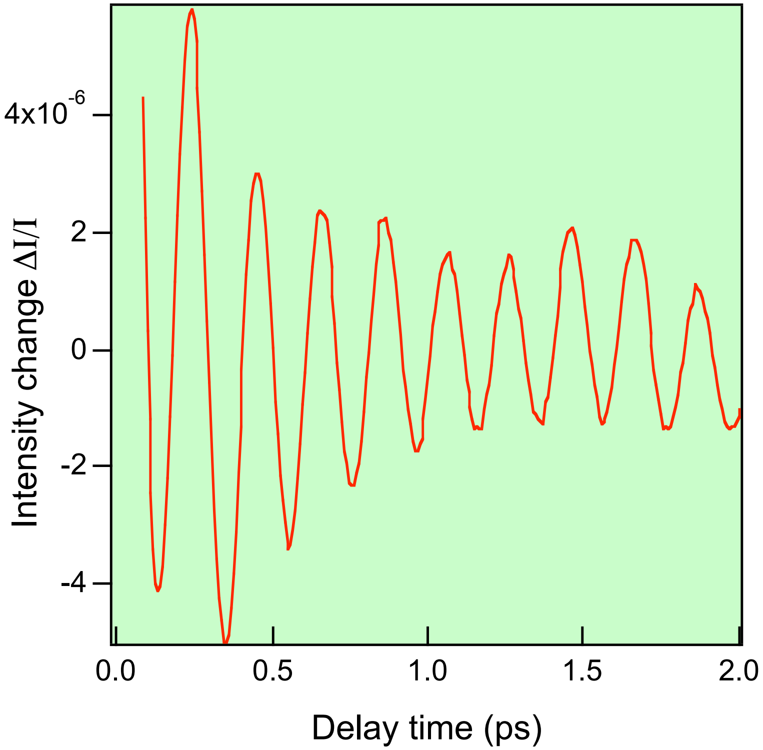


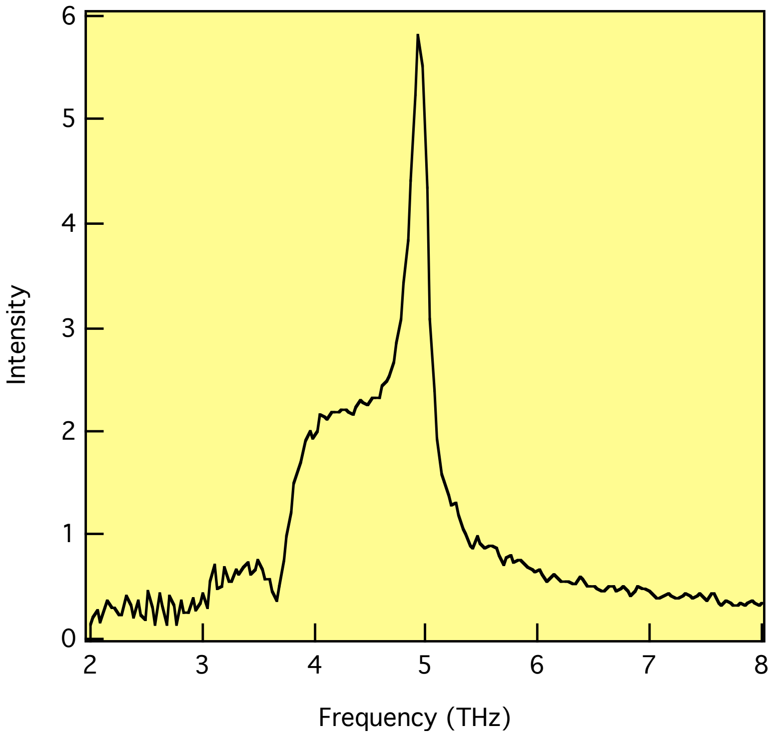


http://www.nims.go.jp/ldynamics/Femto/SU.html

Fs-time resolved optical reflection from CdTe(111)

K. Ishioka et al., Jpn. J. Appl. Phys.45 (2006) 9111





modulation in phonons (5mW):

LO(CdTe): 4.97 THz, τ =2.39 ps

A(Te): 3.82 THz, τ =1.49 ps

Fourier spectrum

Summary

- Transient lattice deformation due to propagation of acoustic phonon and shock wave inside semiconductor crystals has been observed by picosecond TR-XRD
- Coherent optical phonons of CdTe (5THz) has been observed by femtoseconf TR-XRD
- Time-resolution of 200fs in TR-XRD has been achieved

Acknowledgement

- TRXRD
  - TokyoTech: Prof. K. Kondo, Dr. Y. Hironaka, Dr. H. Kishimura, Dr. Y. Okano
- CP by optical reflection
  - NIMS: Prof. M. Kitajima, Prof. K. Ishioka, Dr. M. Hase

- Coherent control in condensed matter
  - Ohmori group, Matsumoto group (IMS), Kitajima group (NIMS)

  - Funds: MEXT, IMS