Coherent phonon spectroscopy

D. Boschetto[1],

C. Rischel[2], M. Merian[1], I. Uschmann[3], E. Foester[3],

O. Albert[1], J. Etchepare[1] and A. Rousse[1]

1 Laboratoire d'Optique Appliquée, ENSTA/Ecole Polytechnique, UMR 7639 CNRS,
INSERM U451, Chemin de la Hunière, 91761 Palaiseau, France

2 Niels Bohr Institute, Blegdamsvej 17, DK-2100 Copenhagen Ø, Denmark

3 X-ray Optics Group, Institute of Optics and Quantum Electronics,
Friedrich Schiller University Jena, Max-Wien-Platz 1, 07743 Jena, Germany

Outlines of the Talk

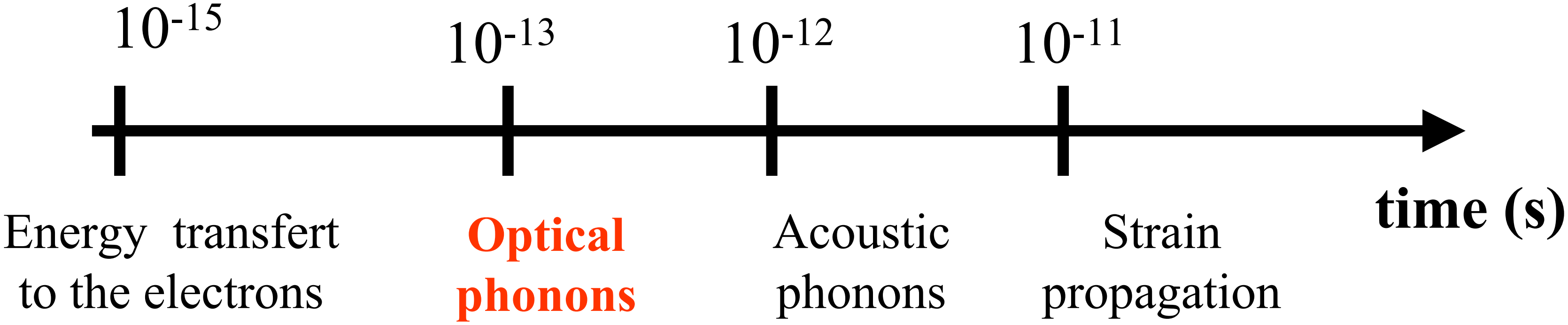
-Overview of phonon features

-Diffraction pattern perturbed by optical phonon

-Choice of the first sample to be investigated by TRXD: Bismuth

-Optical phonon in Bismuth: optical and TRXD measurements

Solid excited by a laser pulse



10^{-15}

10^{-13}

10^{-12}

10^{-11}

time (s)

Energy transfert to the electrons

Optical phonons

Acoustic phonons

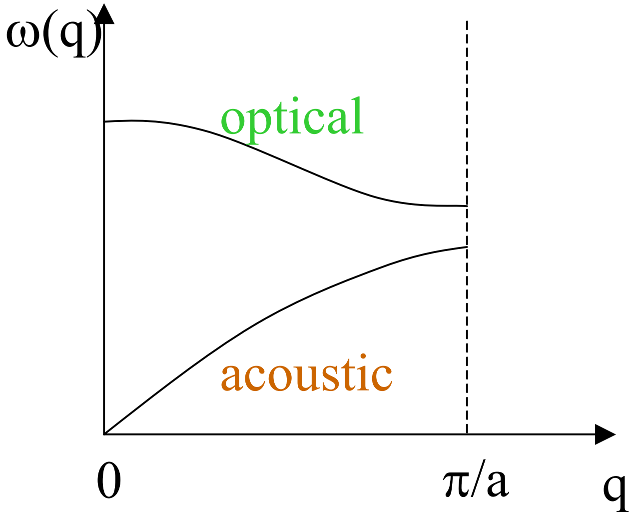
Strain propagation

Optical Phonons:

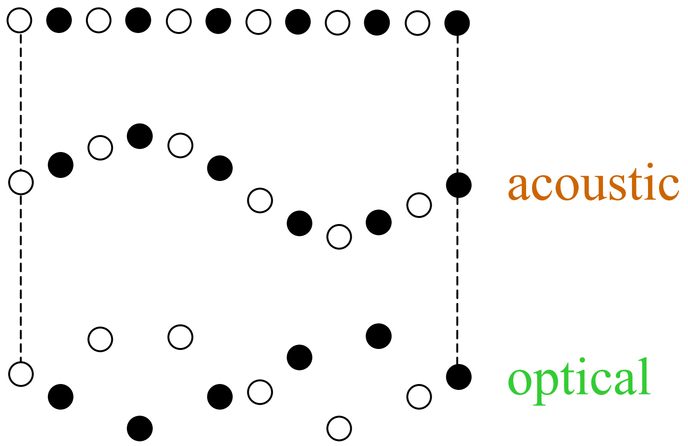
first response of the nucleus to the external excitation

Phonon

Phonon dispersion



Lattice vibration of diatomic linear chain



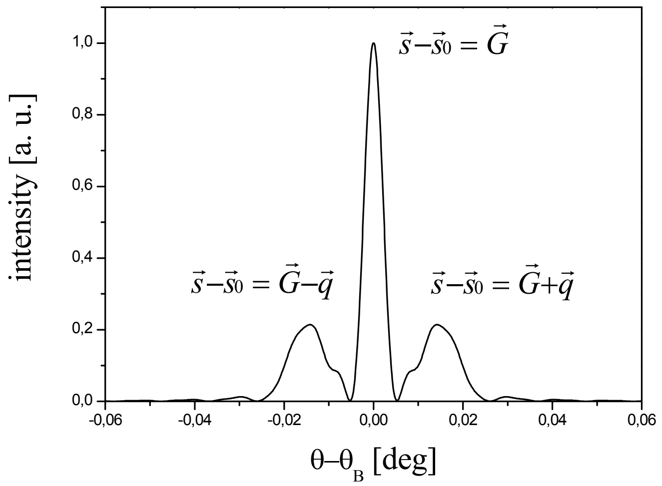
Phonon parameters:

-Wavelength : fixed by the experimental geometry

-Frequency : directly measured by optical pump-probe experiments
-Dumping time : directly measured by optical pump-probe experiments

-Displacement amplitude : no direct measurement exists !!

Coherent Phonon Contribution to the Rocking Curve



- Satellite peaks : $\Delta\theta \propto |\vec{q}|$

-Time-dependent structure factor:

$$F_{(hkl)}(t) = \sum_{j=1}^{n} f_j \;\exp\left[2\pi i\,\left(hx_j(t)+ky_j(t)+lz_j(t)\right)\right]$$

Time-dependent intensity: $I(t) \propto \left|F_{(hkl)}(t)\right|^2$

Acoustic Phonons with q≠0

Time dependent intensity of the satellite peaks

When several phonons are excited, each satellite peak will oscillate with a frequency given by the phonon dispersion: ω(q) 🕑 ω(Δθ)

Optical Phonon with q≈0

All the atoms in the crystal oscillate with the same phase

↓

Time-dependent intensity of the central peak

New challenge :

The measurement of Optical Phonons (higher frequency)

(to be able to follow elementary atomic displacements)

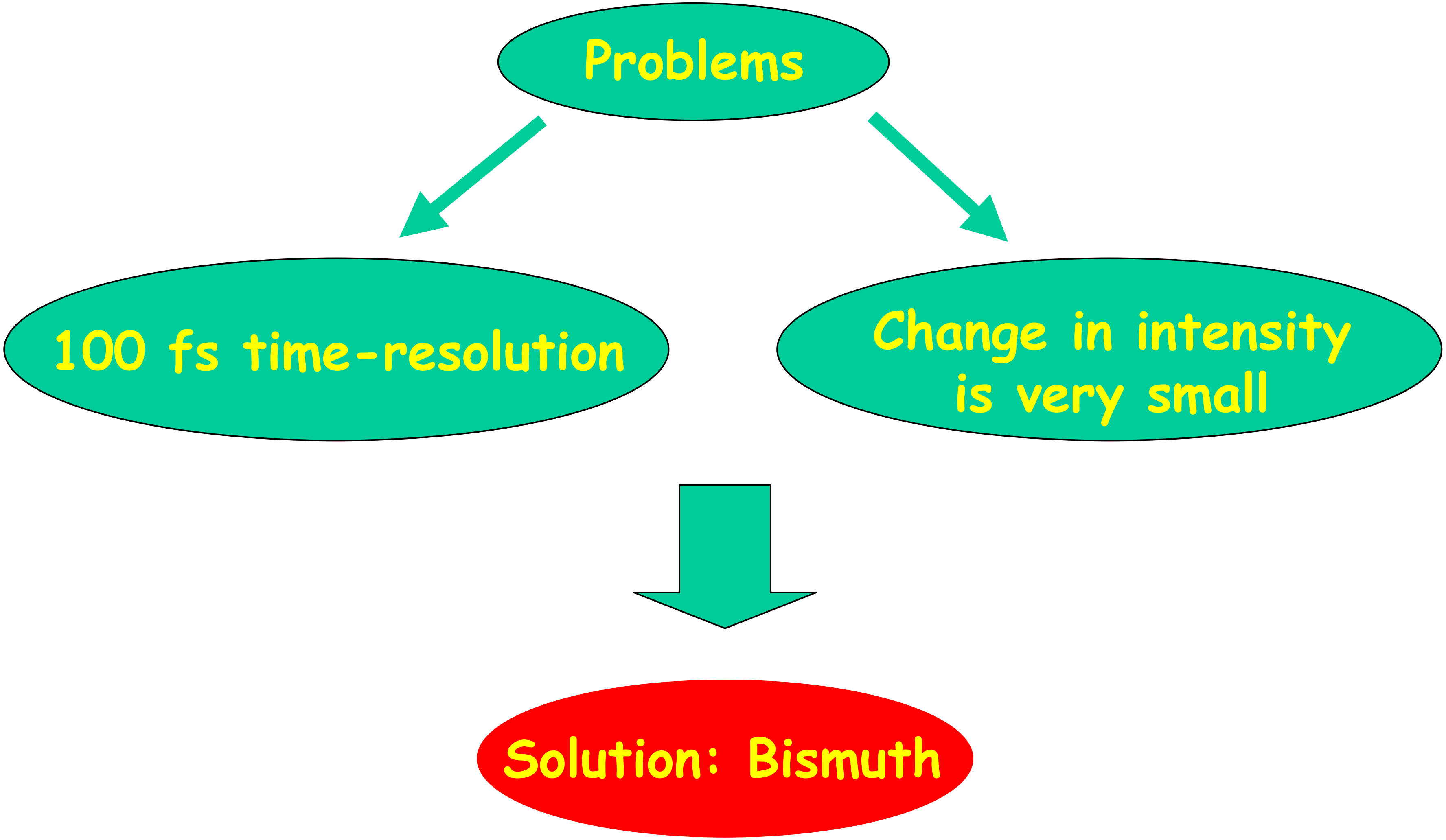
Acoustical Phonon : $T_A \sim 10$ ps

Optical Phonon : $T_O < 1$ ps

Problem :

The choice of the first material to start with

Difficulties in Optical Phonon Detection



Problems

100 fs time-resolution

Change in intensity
is very small

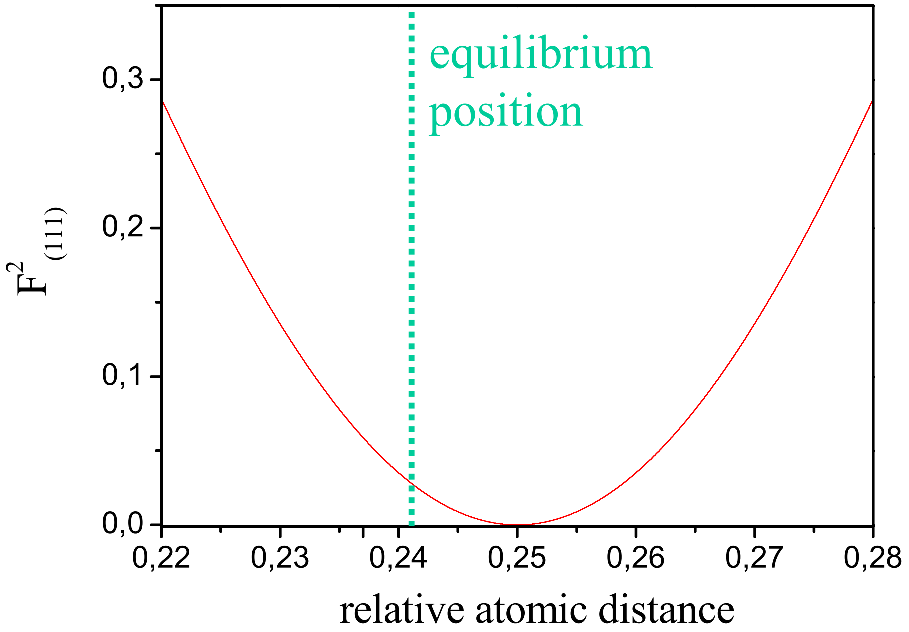
Solution: Bismuth

Bismuth structure

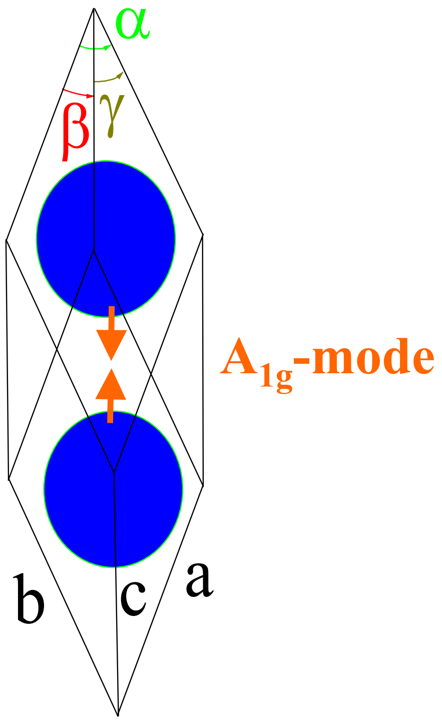
Trigonal primitive unit cell with a=b=c=0.4745 nm and α=β=γ=57.23 deg, containing two atoms at: $(\pm u, \pm u, \pm u)$ $u = 0.237$

Structure factor :

$$F_{(111)} = 2 f_{Bi} \cos[6\pi \cdot (u + \Delta u)]$$



J. Wark, SPIE proceeding (2000)



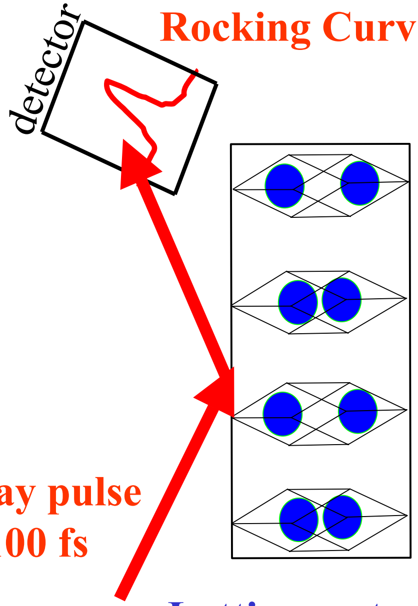
ω_{A1}= 2.92 THz

⇓

T_{A1}= 342 fs

Simulation of Rocking Curve perturbed by Optical Phonon

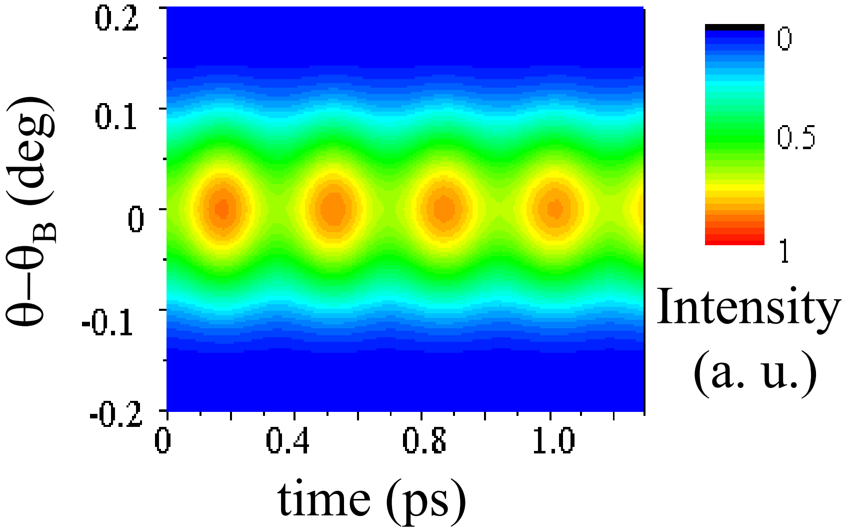
Simulation of time-dependent rocking curve from the *(1 1 1)*-plane in presence of coherent optical phonon of amplitude 0.013 Å in 12.5 nm Bismuth film



Rocking Curve

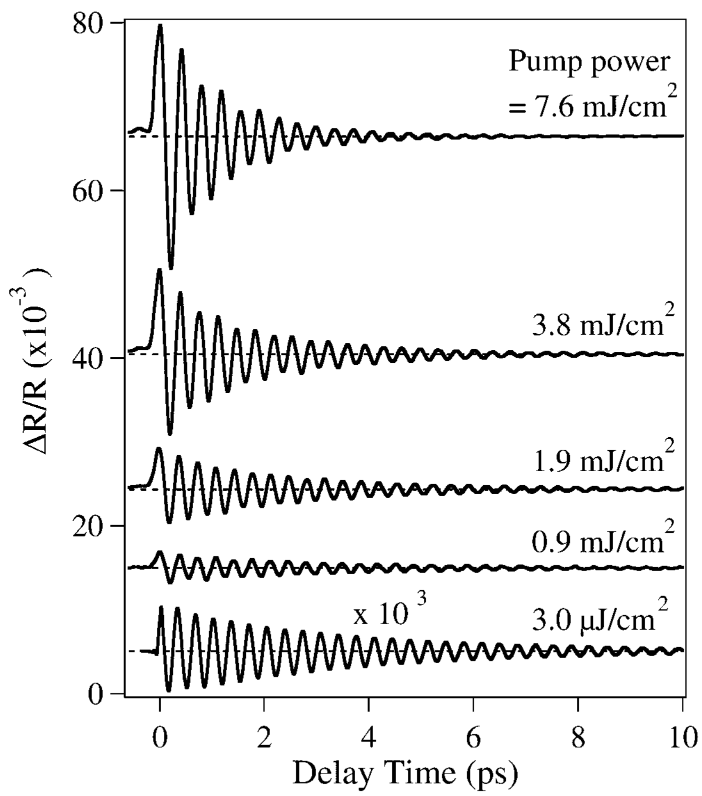
X-ray pulse
100 fs

Lattice perturbed
By optical phonon

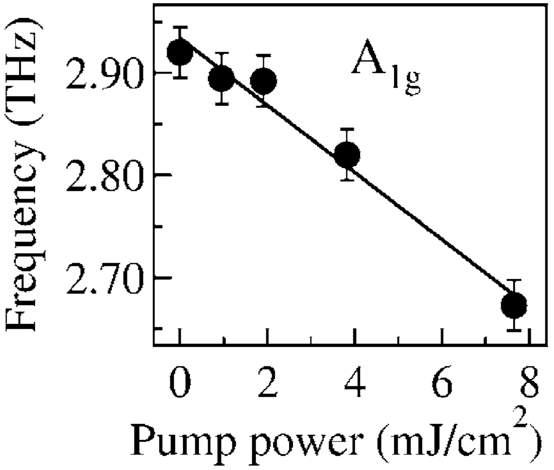


The change in integrated intensity is about 15 %

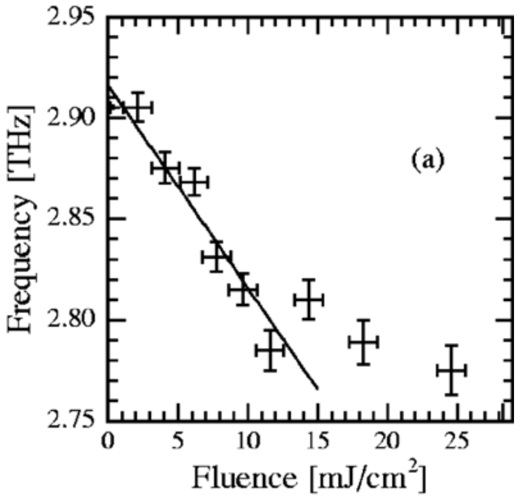
Coherent Optical Phonon in Bismuth: Optical Experiments



Hase et al., PRL **88**, 67401 (2002)



- *softening*



DeCamp et al.,
PRB **64**, 92301 (2002)

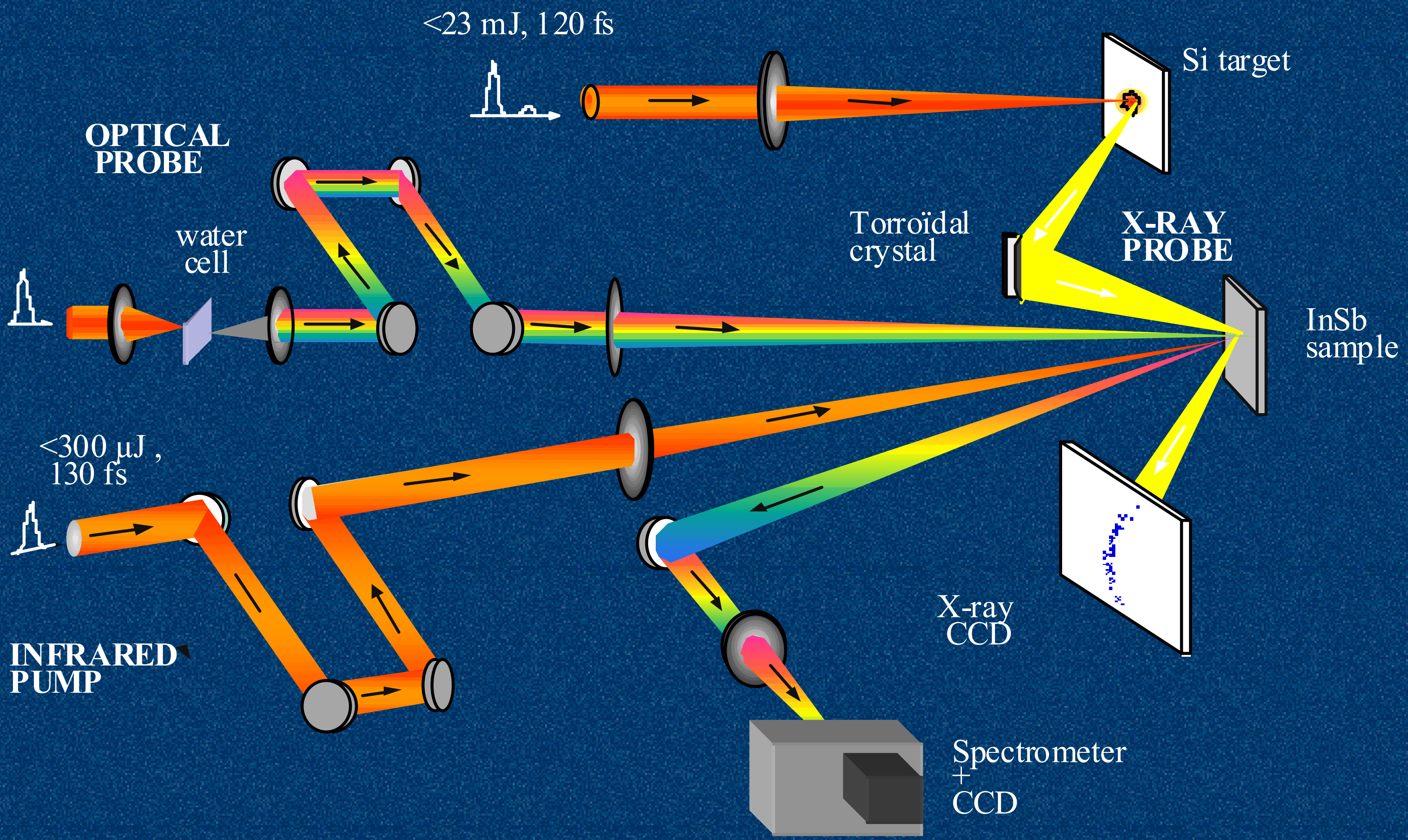
Problem: <u>no quantitative</u> measure of phonon amplitude

slide from K. Sokolowski-Tinten

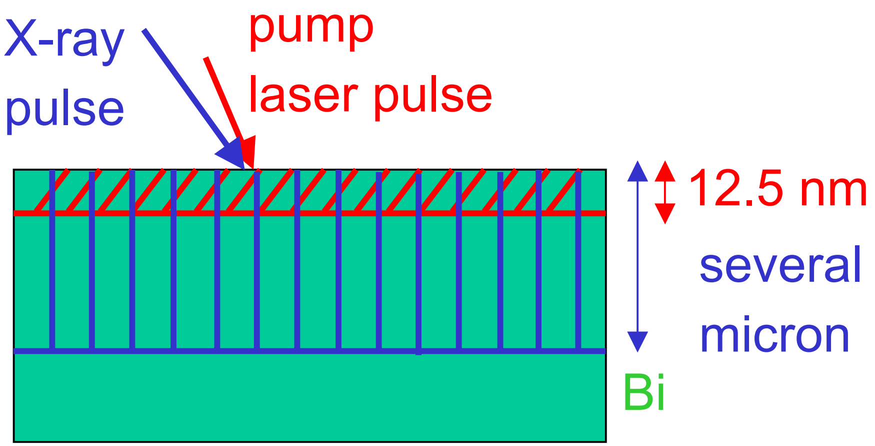
Femtosecond X-Ray Diffraction on Bi:
Optical Phonon Measurement

Schematic description of the experimental set-up

L.O.A



Penetration depth mismatch in Bi



Problem: penetration depth mismatch

Laser pulse: ~0.01 μm X-ray: ~1 μm

Solutions

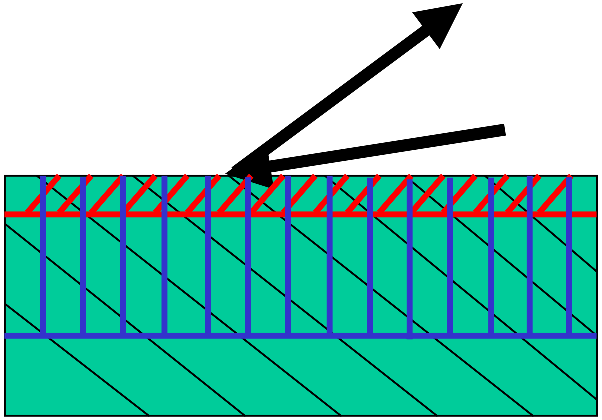
Low energy X-ray and asymmetric reflection on massif sample

Thin film

Experimental conditions at LOA

Si-K_α = 7.13 Å (111)-reflection : θ_B=64.39 deg

We choose a miscut of –55 deg



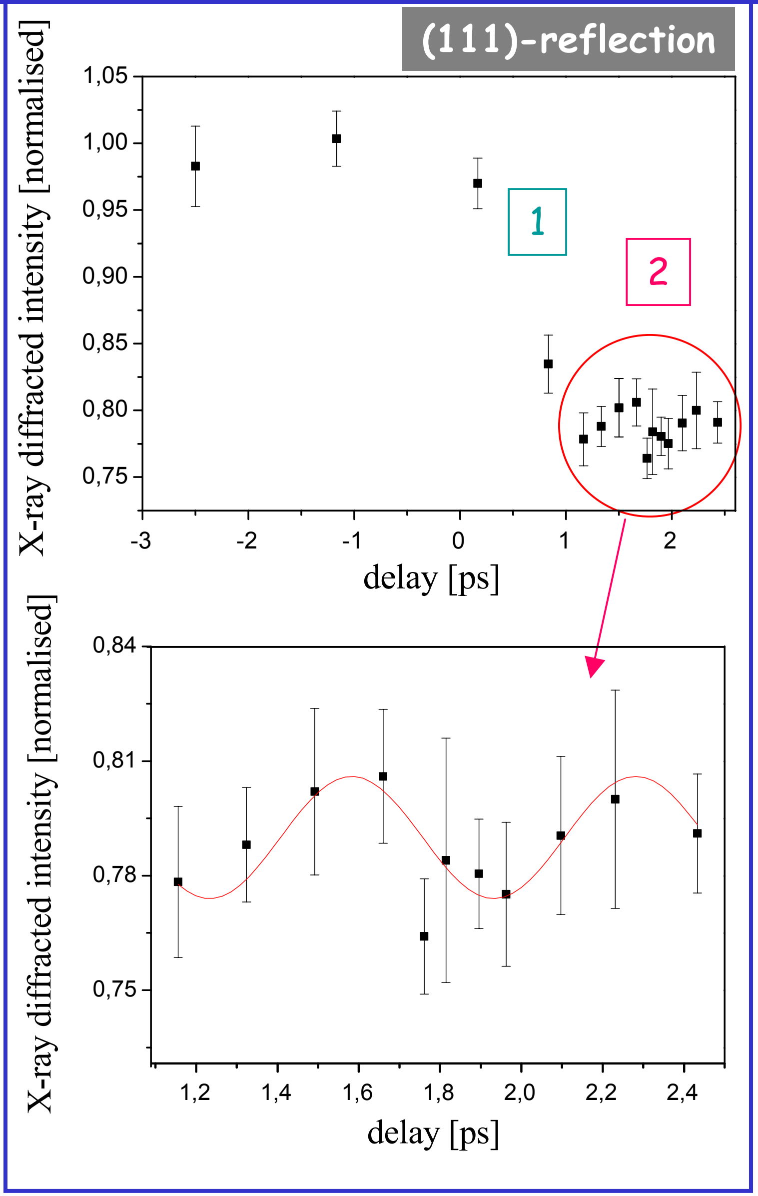
Grazing incidence angle ~ 10 deg

Laser attenuation length : 0.012 µm

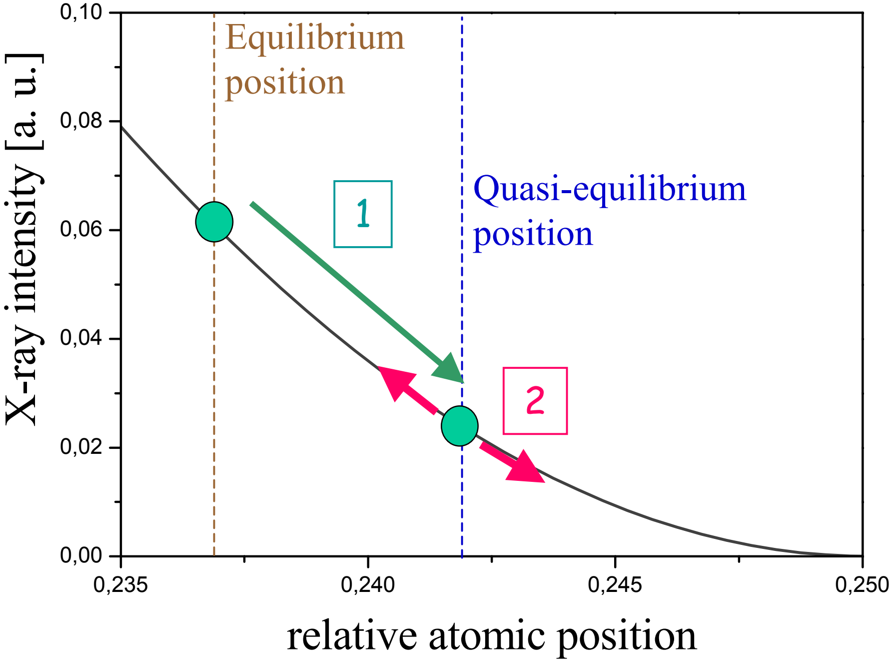
X-ray attenuation length : 0.09 µm

Bi [111] miscut of -55 deg

Femtosecond X-ray Diffraction detection of Coherent Optical Phonon in Massif Bi



Laser fluence: 6 mJ/cm^2



Displacement amplitude ~ 0.2 Å

Softening: **ν_{obs}~ 1.54 THz (T_{obs} ~ 650 fs)**
ν_{A1}= 2.92 THz (T_{A1} = 342 fs)

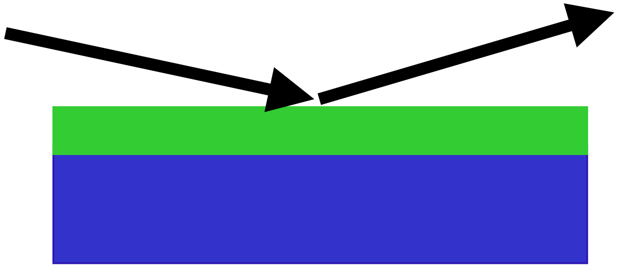
K. Sokolowski-Tinten, C. Blome, J. Blums, C. Dietrich,
A. Tarasevitch, M. Horn von Hoegen, D. von der Linde
Institut für Experimentelle Physik, Universität Duisburg-Essen

A. Cavalleri
Material Science Div., Lawrence Berkeley National Laboratory

I. Uschmann, E. Förster
AG Röntgenoptik, Friedrich-Schiller-Universität Jena

M. Kammler
Institut für Halbleitertechnologie, Universität Hannover

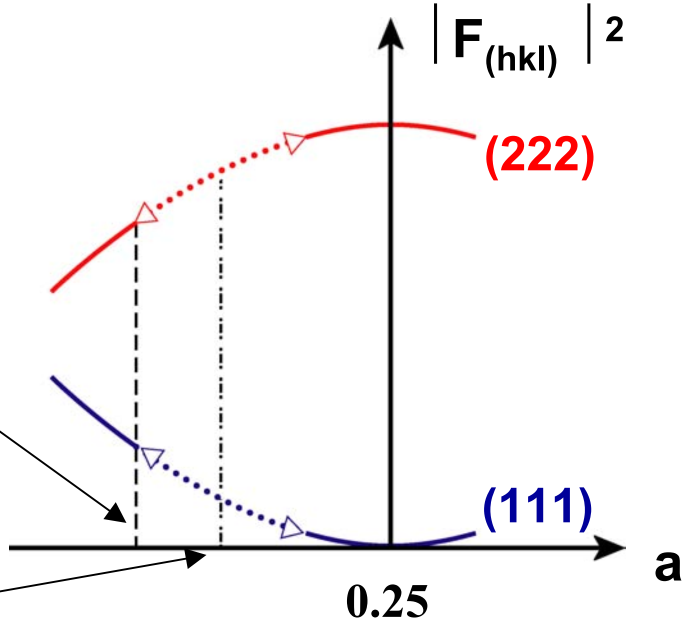
Experimental conditions in Essen



Ti-K_α = 2.75 Å

(111)-reflection : θ_B= 20.35 deg

(222)-reflection : θ_B= 44.08 deg



initial equilibrium position

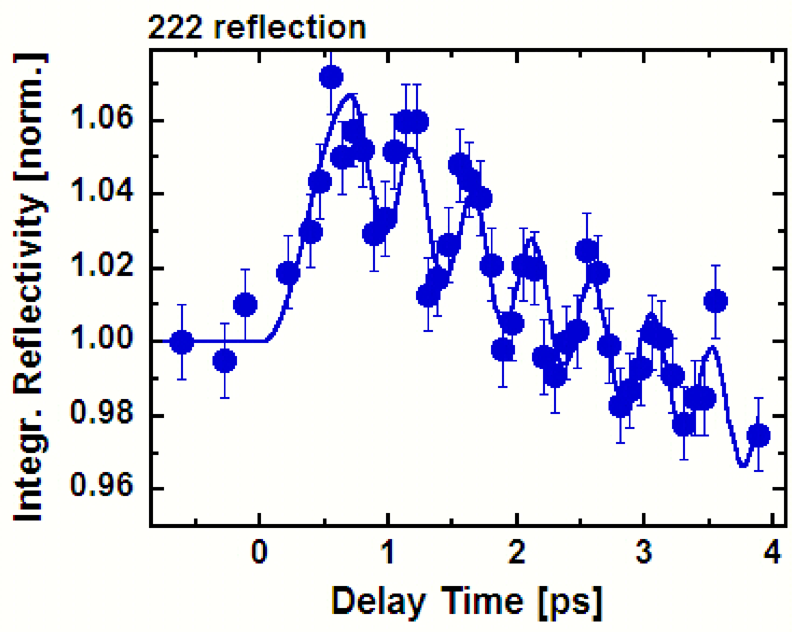
***displaced* quasi-equilibrium position**

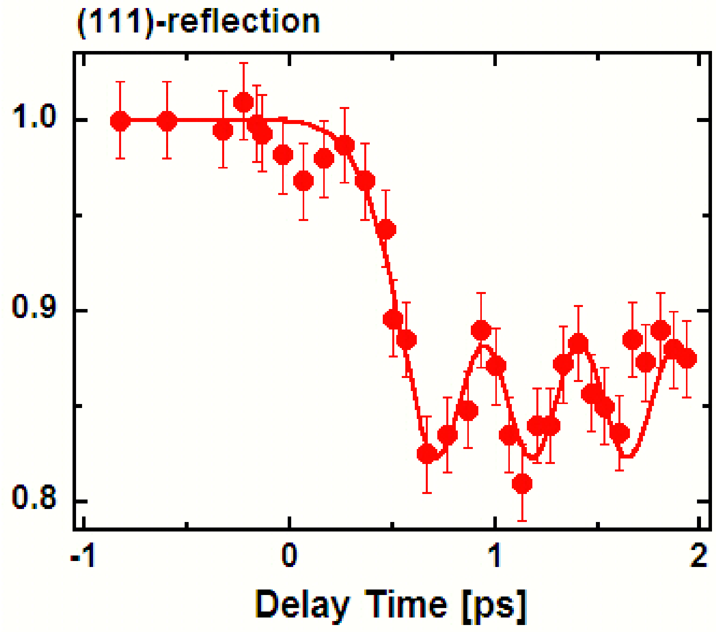
→ **increase** and oscillation of the **(222)** reflection

→ **decrease** and oscillation of the **(111)** reflection

Coherent Optical Phonons

Bi 50nm on Si, F ≈ 6 mJ/cm^2





A_{1g} optical mode:

ν_{obs} = **2.14 THz** (470 fs)

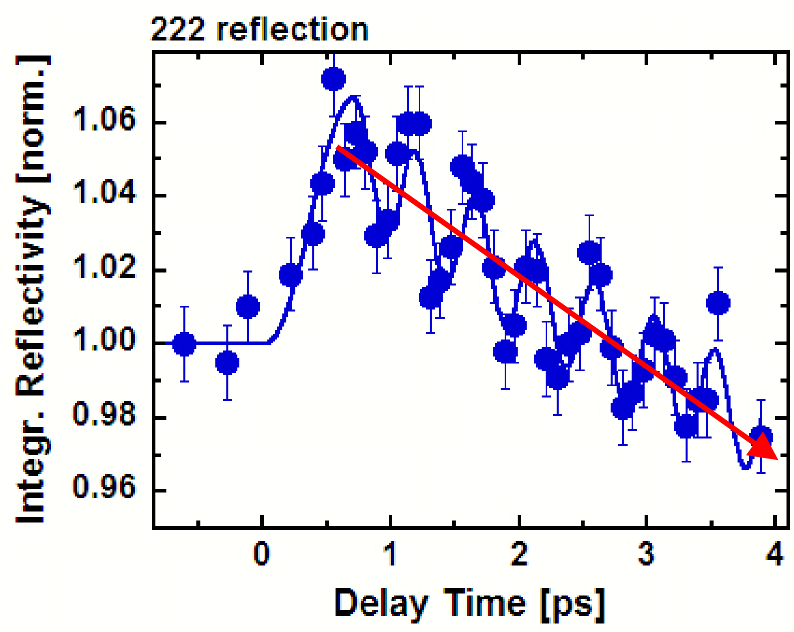
ν_0 = 2.92 THz (342 fs)

← *softening*

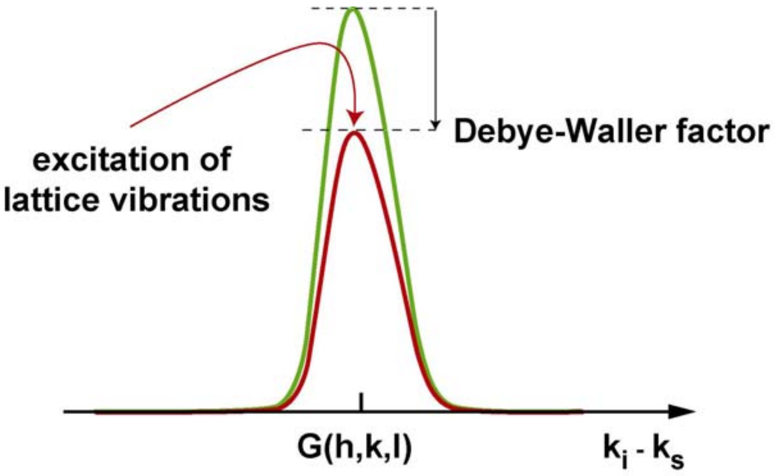
Displacement amplitude:
0,15 – 0,25 Å

K. Sokolowski-Tinten et al., Nature **422**, 287 (2003)

Phonon Dynamics



Debye-Waller effect:

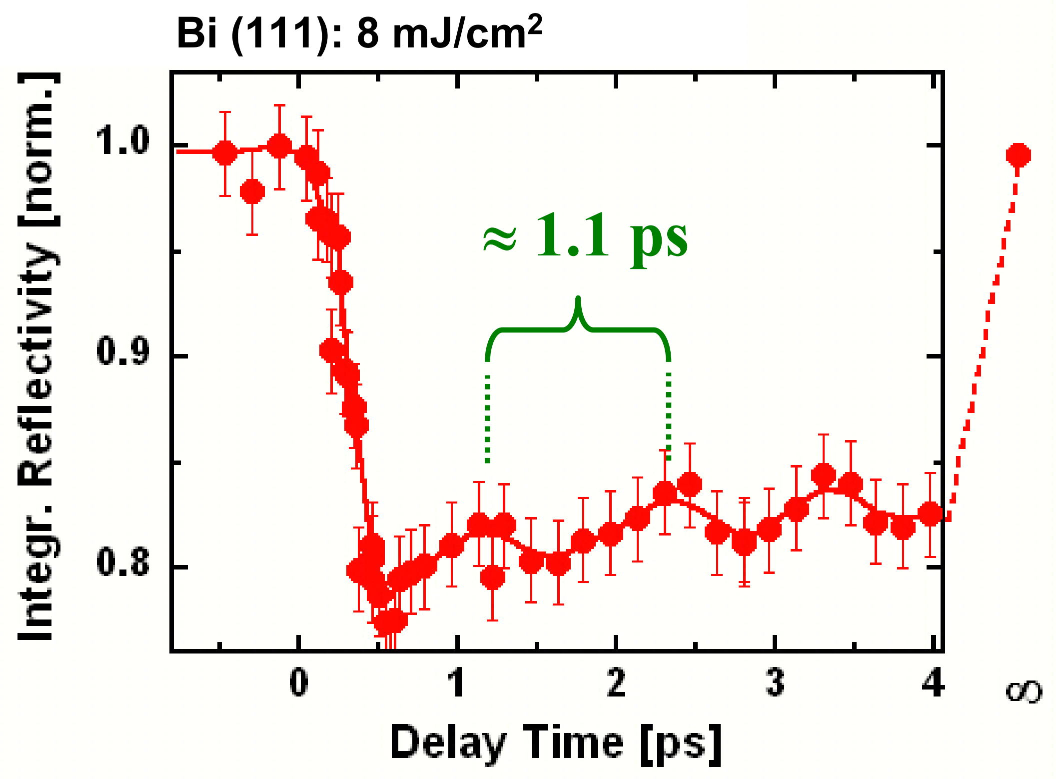


coh. optical phonons ⇨ **incoh. phonons: *heat***

K. Sokolowski-Tinten et al., Nature **422**, 287 (2003)

Coherent Phonon ⇔ Phase Transition

Change of atomic motion: ***periodic*** ⇒ ***a-periodic*** *?*



$\underline{A_{1g}\text{ optical mode:}}$

ν_{obs} = **0.9 THz** (1.1 ps)

ν_0 = 2.92 THz (342 fs)

slide from K. Sokolowski-Tinten

In both laboratory (LOA and Essen)

a big frequency shift was measured

Conclusions

-We are able to detect elementary atomic movements

-Acoustic and Optical Phonons have been seen

-Big frequency shift observed by TRXD

Perspectives:

Study the contribution of optical phonons
to ultrafast dynamics, such as
non-thermal melting and
charge-transfert dynamics