Structural dynamics explored by time-resolved X-ray diffraction

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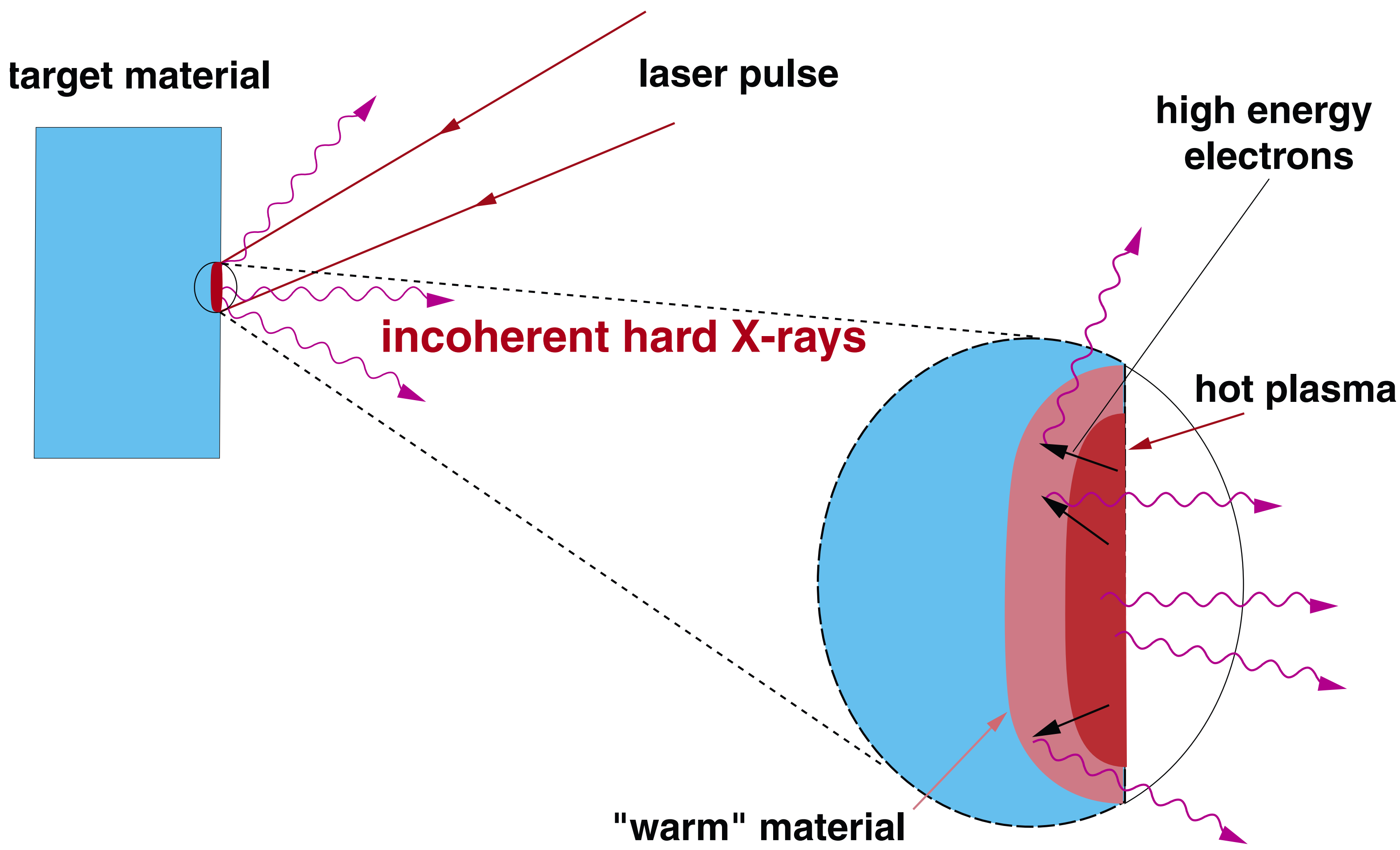
Outline

- Subpicosecond keV X-ray pulses
- Ultrafast non-thermal melting of semiconductors
- X-ray diffraction experiments on Bismuth
- Displacive excitation of coherent optical phonons
- Conclusions
- Outlook

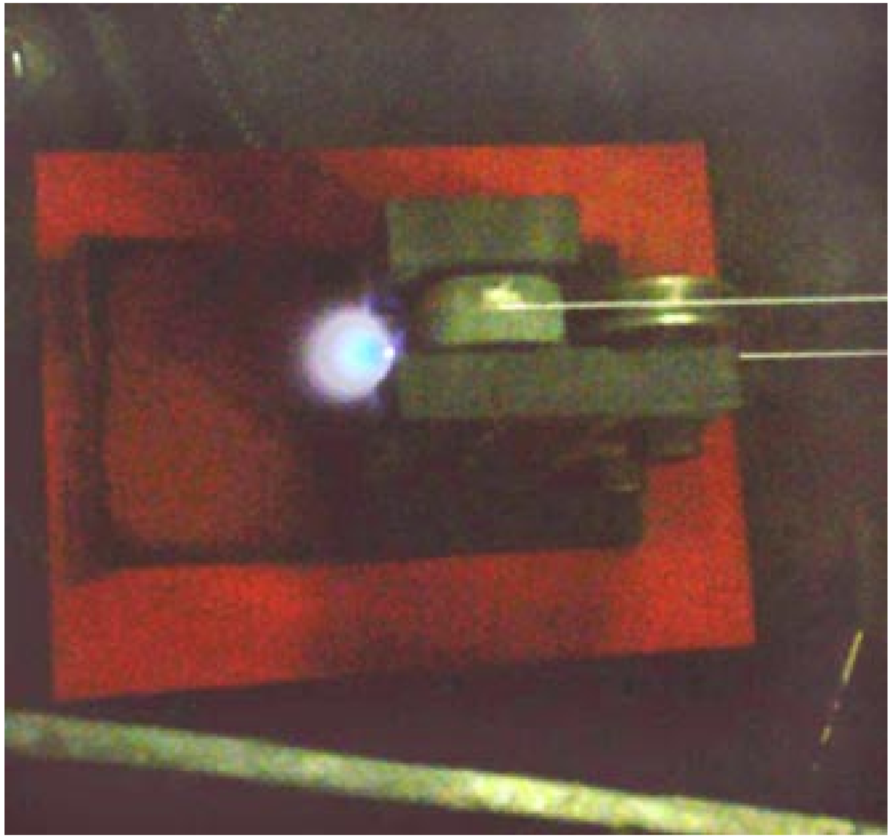
Generation of subpicosecond keV X-ray pulses

Laser-driven plasma X-ray source

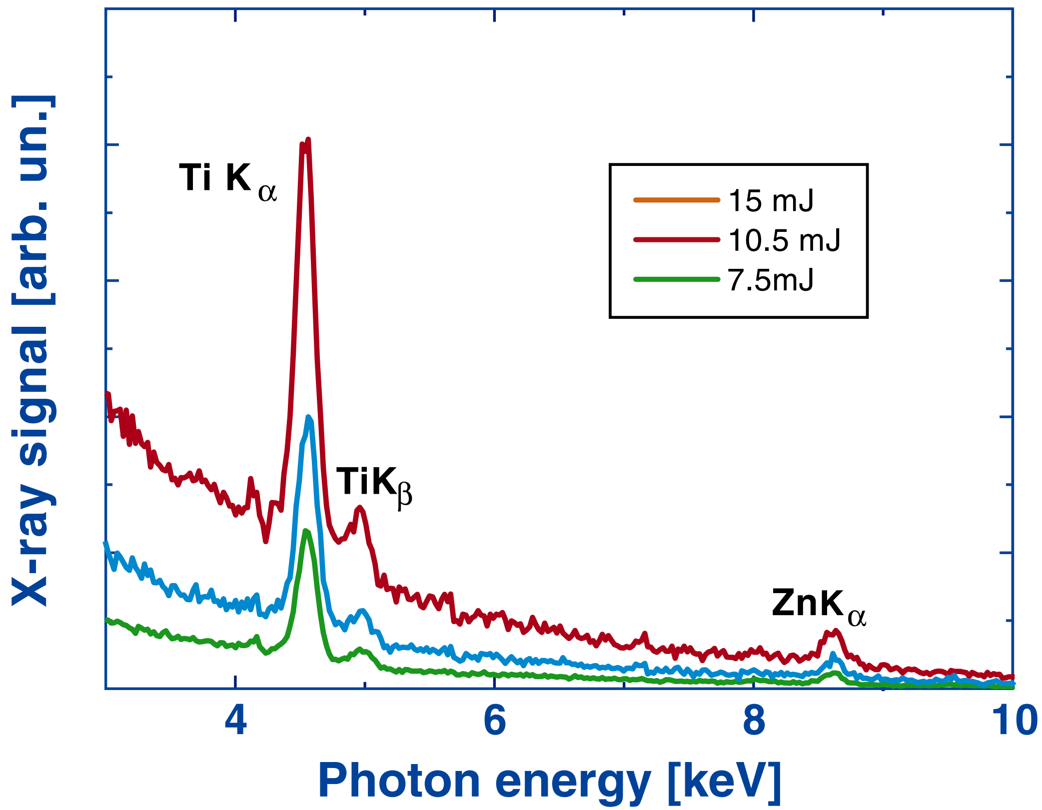


Titanium laser X-ray source

Wire target

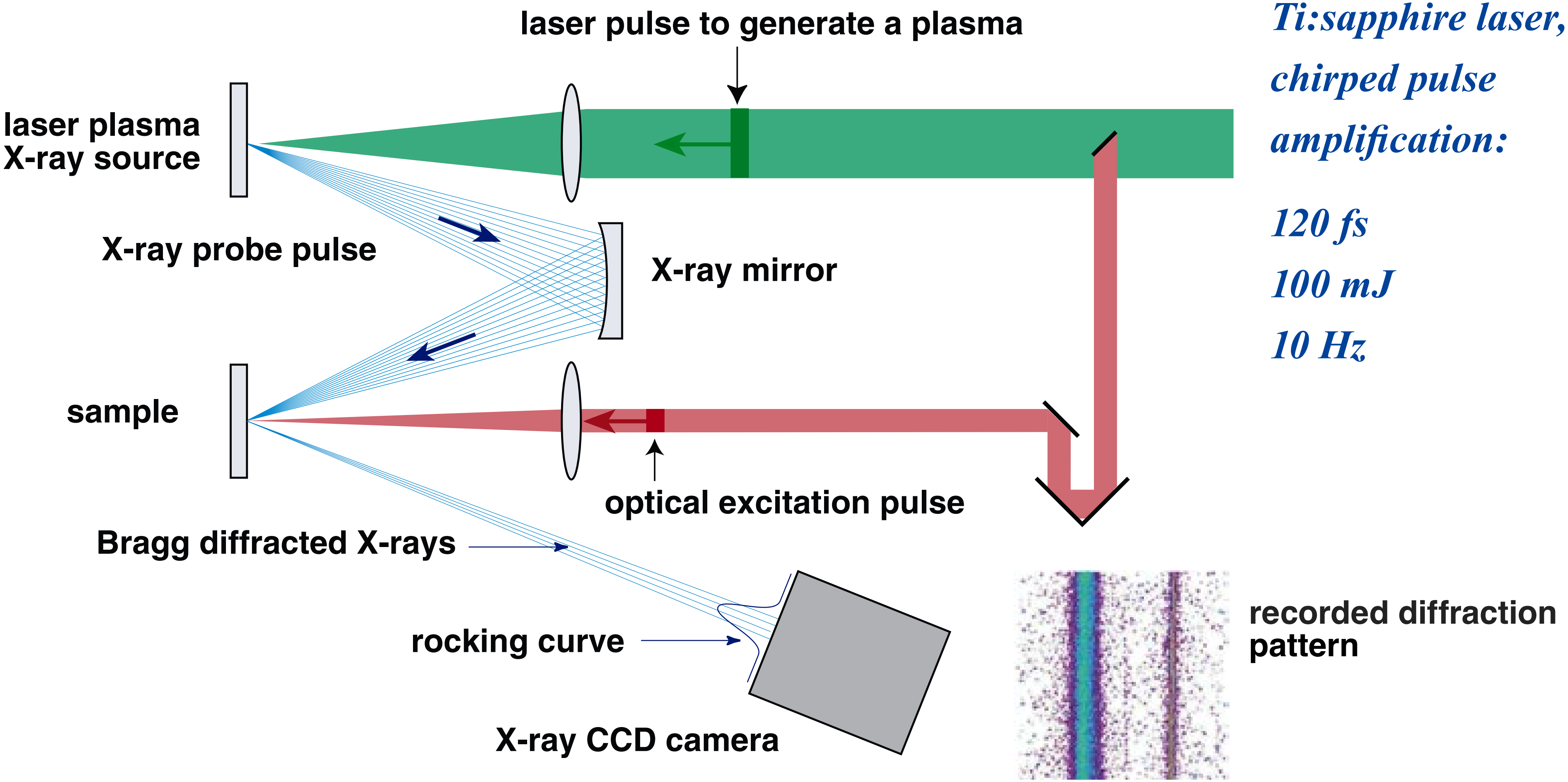


X-ray spectra



Time-resolved X-ray diffraction

Optical pump/X-ray probe



Ti:sapphire laser, chirped pulse amplification:

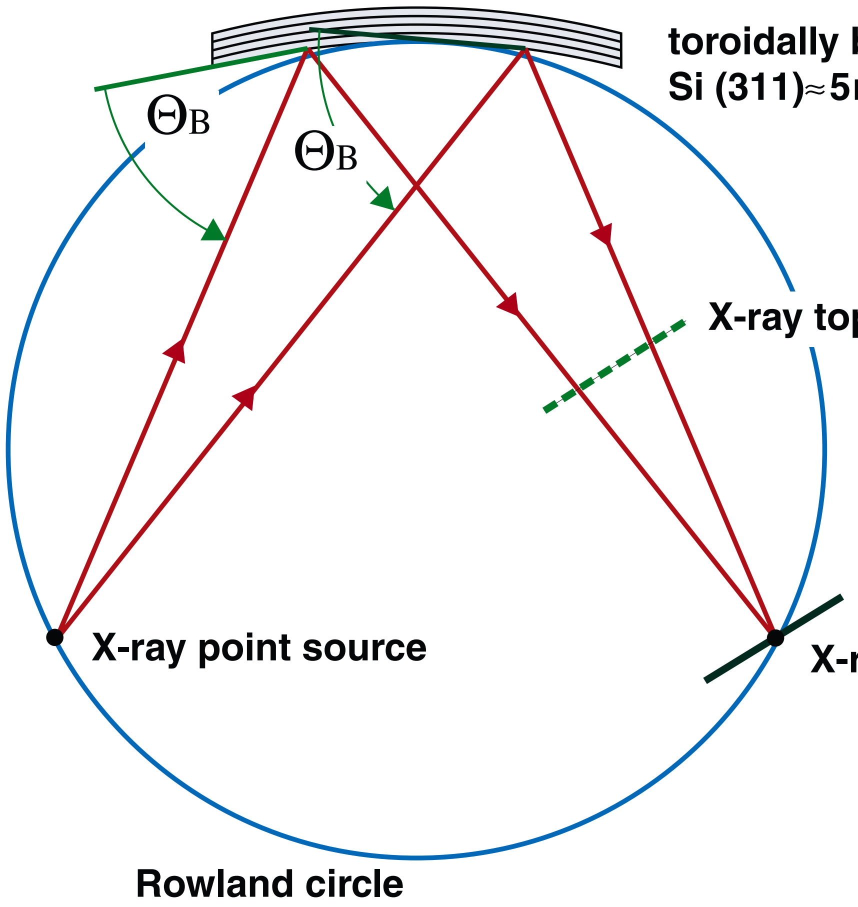
120 fs

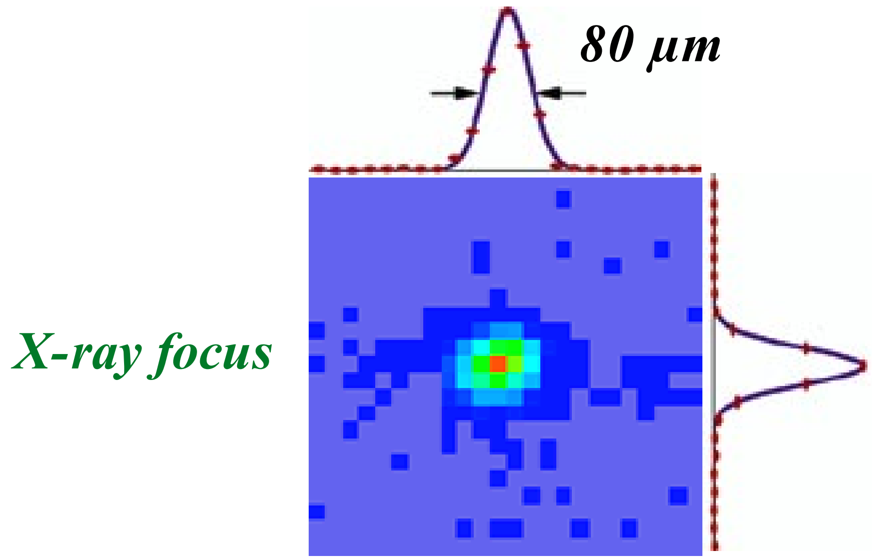
100 mJ

10 Hz

Focusing of keV X-rays

Credit: Ingo Uschmann (FSU Jena)



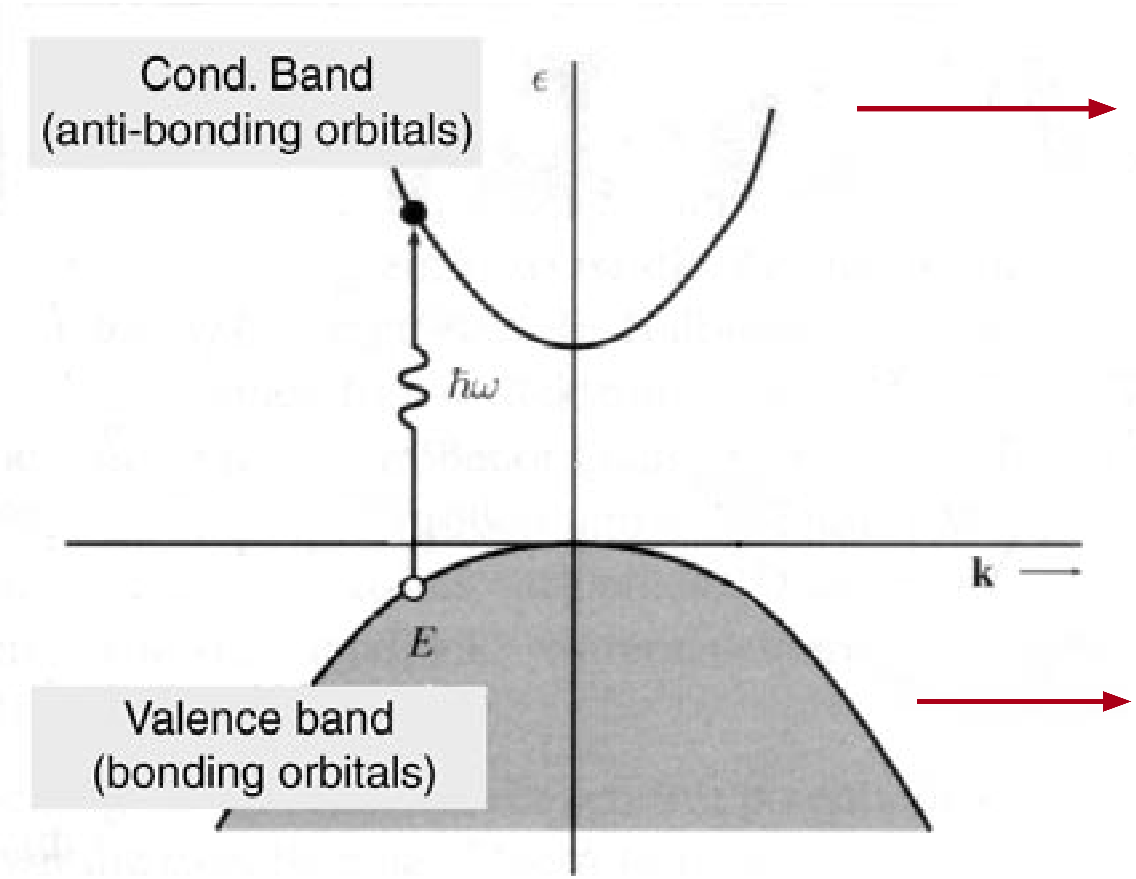


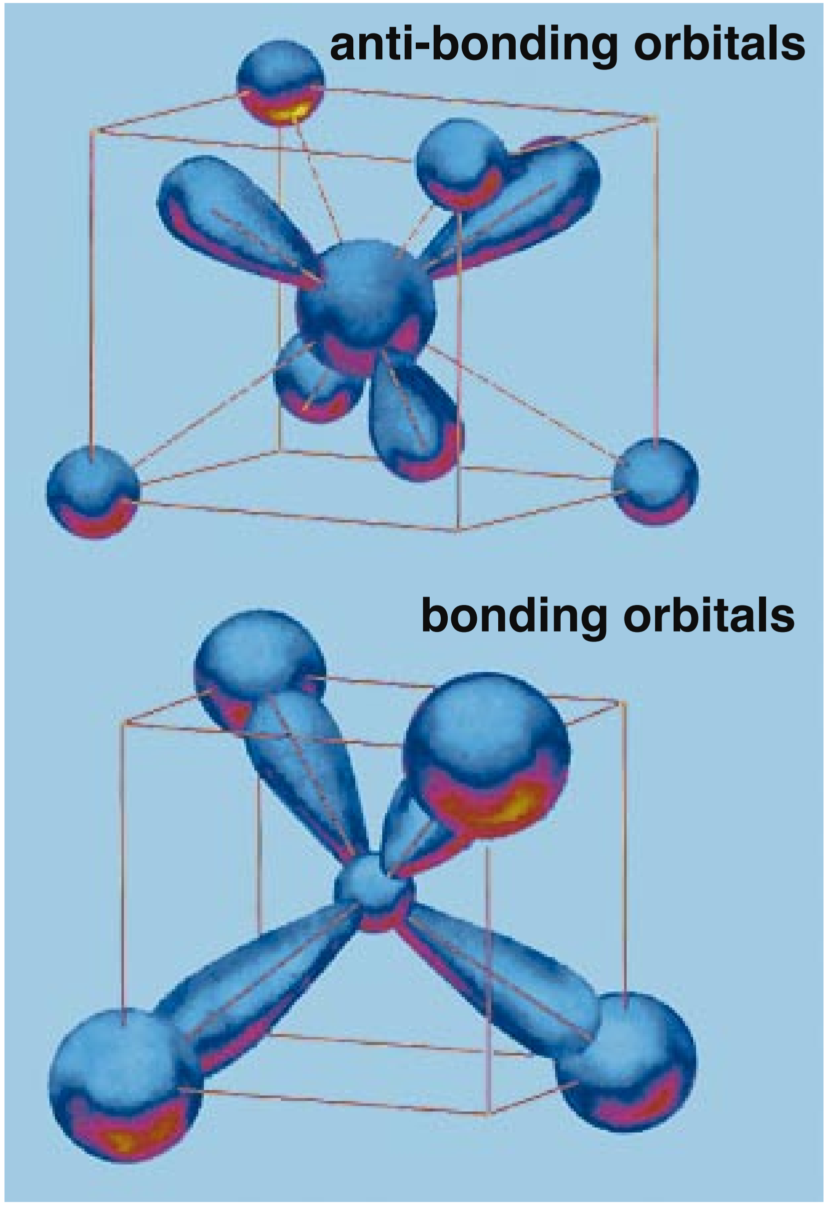
X-rays:

10^4 -10^5 Ti-K_α photons (4.51 keV)

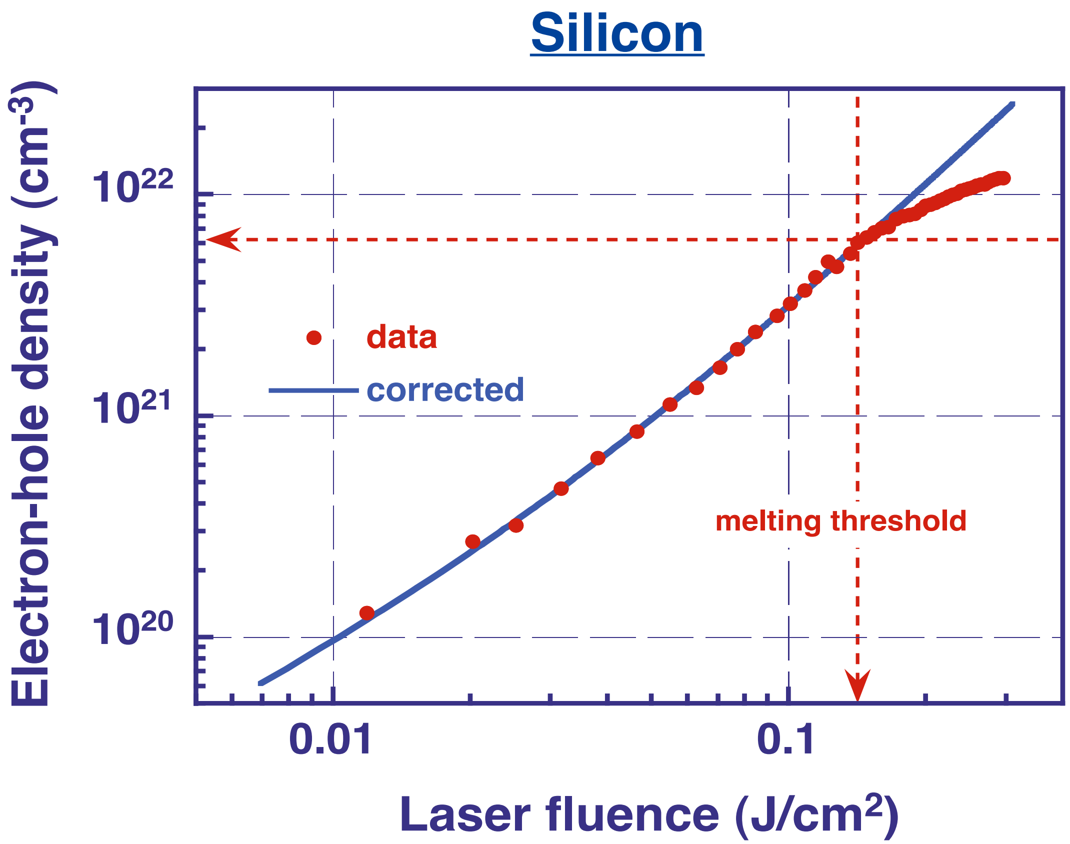
Ultrafast non-thermal melting

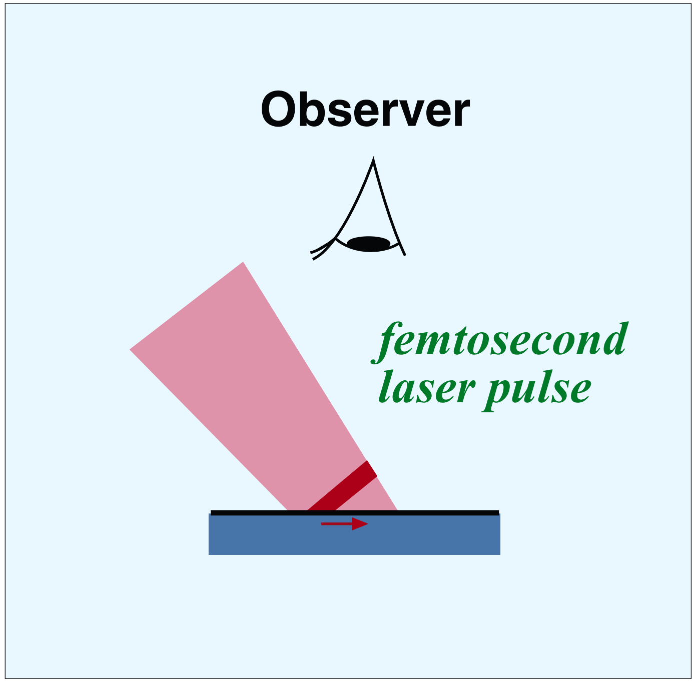
Band and bond picture of electronic excitation



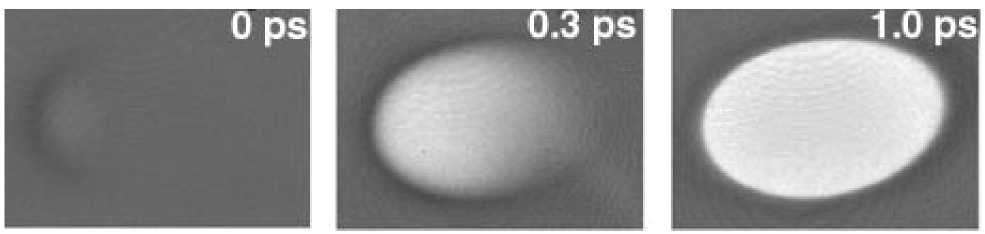


Optical evidence of ultrafast, non-thermal melting



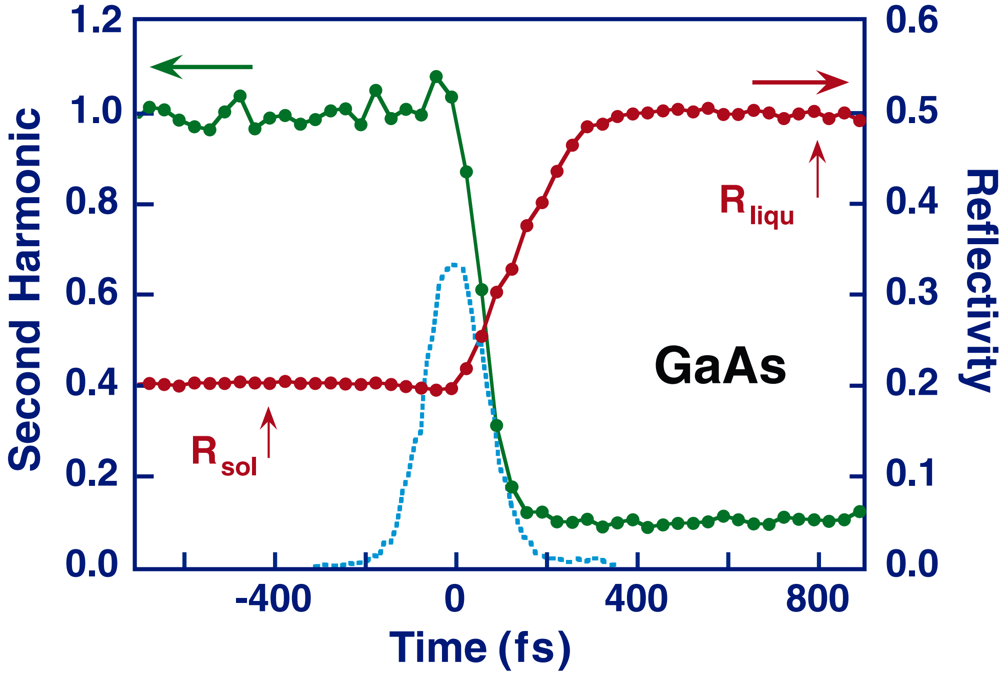


Optical micrographs: Snapshots of the laser-excited silicon surface

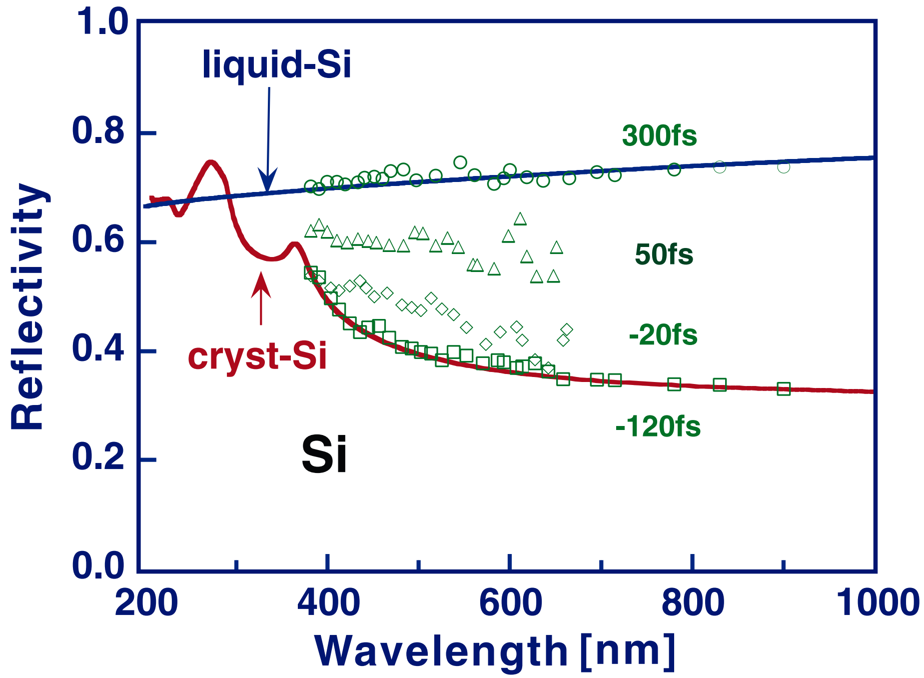


More optical evidence of non-thermal melting

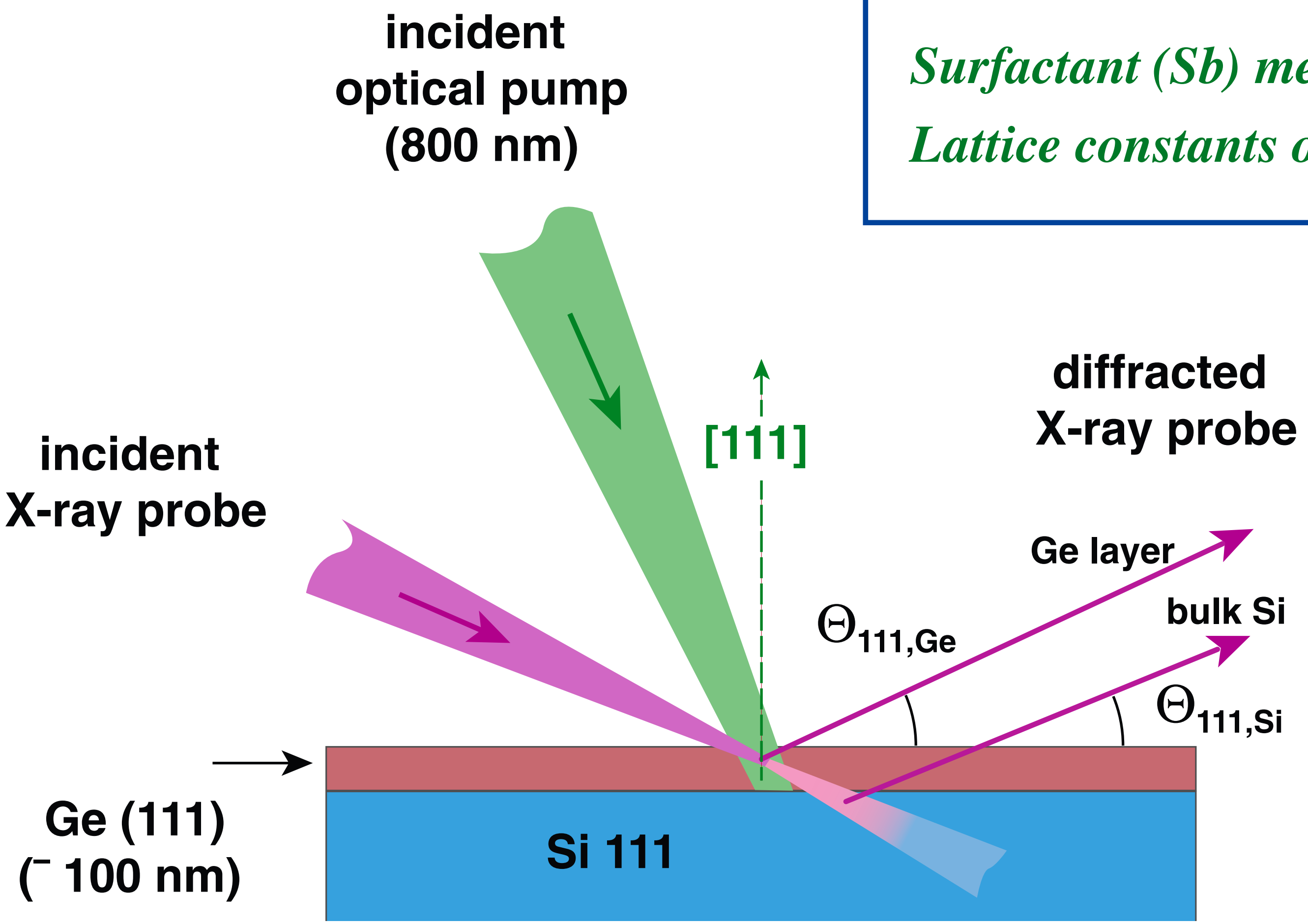
Time dependence of the reflected Second Harmonic and the optical reflectivity (GaAs)



Time-resolved spectra of the optical reflectivity (Si)



Experiments on Ge:Si hetero-epitaxial structures



Ge hetero-epitaxial crystal layers on Si (111)

Surfactant (Sb) mediated growth of Ge.

Lattice constants of Ge and Si not matched.

- *Selective laser excitation of the Ge layer*
- *Selective X-ray probing of the Ge layer*

Parameters of the X-ray diffraction experiments

- **X-Ray probe pulse: Ti-K_α laser plasma source (λ = 0.274 nm)**

  (100 mJ, 120 fs laser pulses, 10 Hz, Titanium wire target)

- **Optical pump pulse: Ti-Sapphire laser (λ = 800 nm)**

  (120 fs, laser-excited area on the sample ≈ 0.2 × 0.4 mm^2)

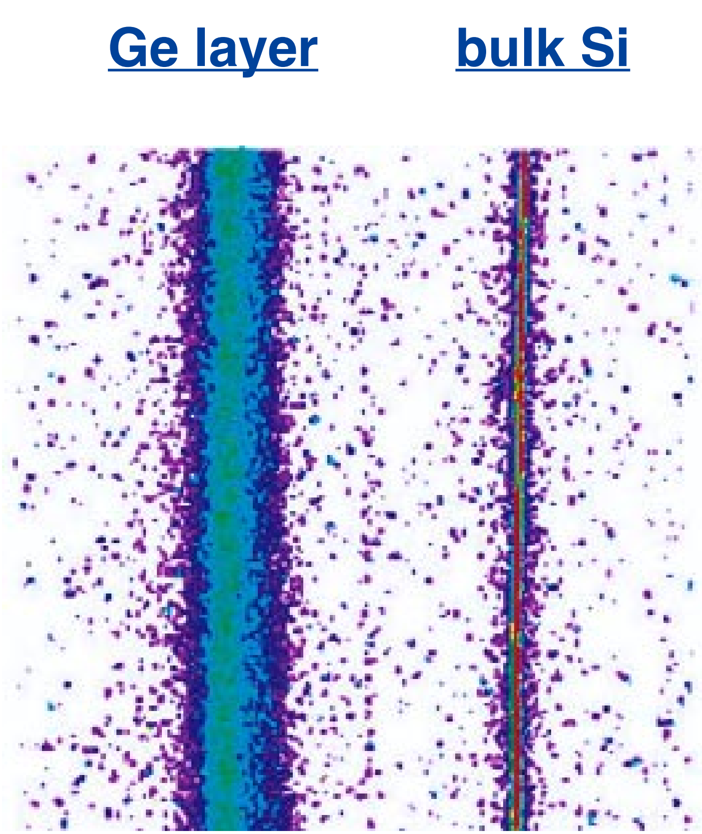
- **Raster scanning of (111)-Ge: Si-wafers**

- **Data accumulation over several 100 laser pulses**

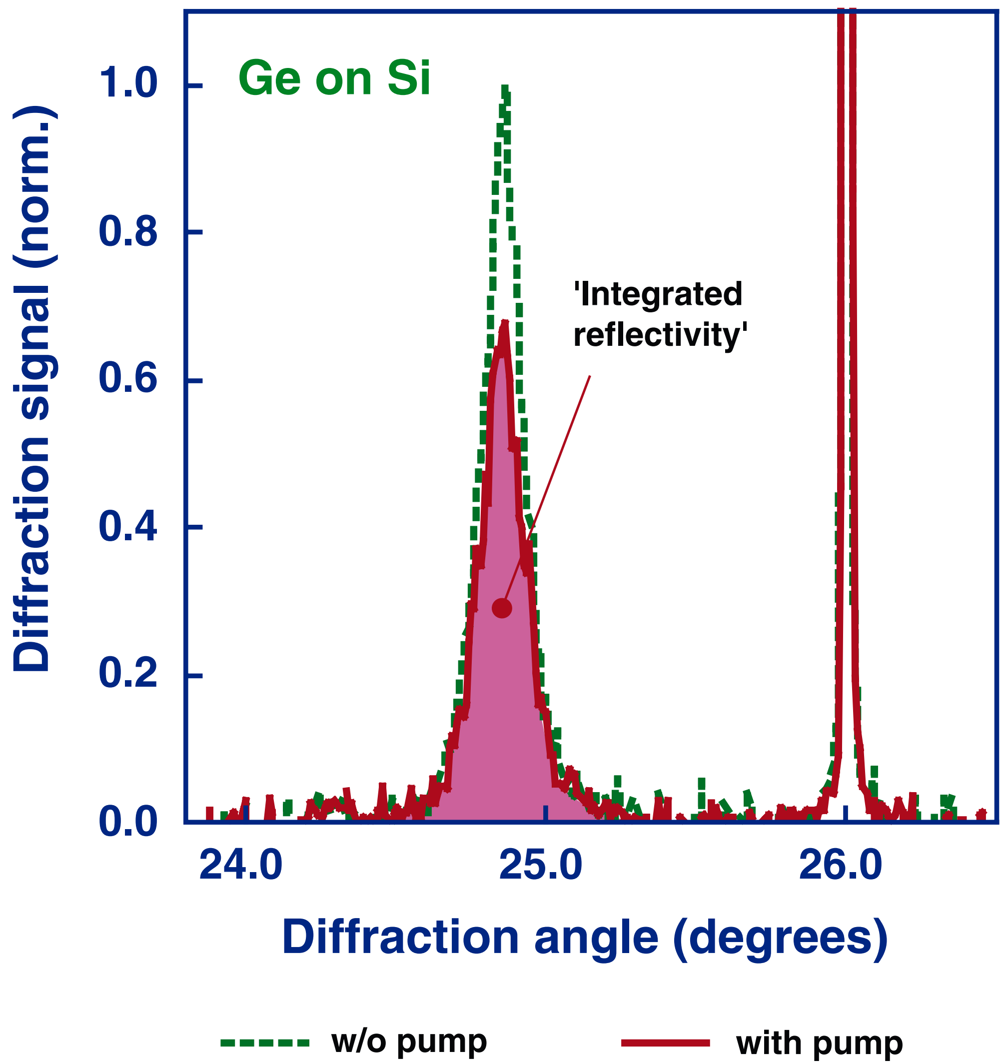
- **X-ray focusing with toroidal Si (311) mirror**

  ‣ **Time-dependent diffraction patterns (rocking curves)**

Example of X-ray diffraction patterns

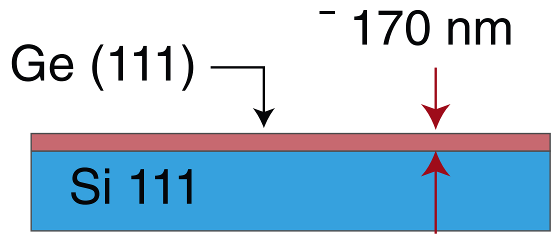


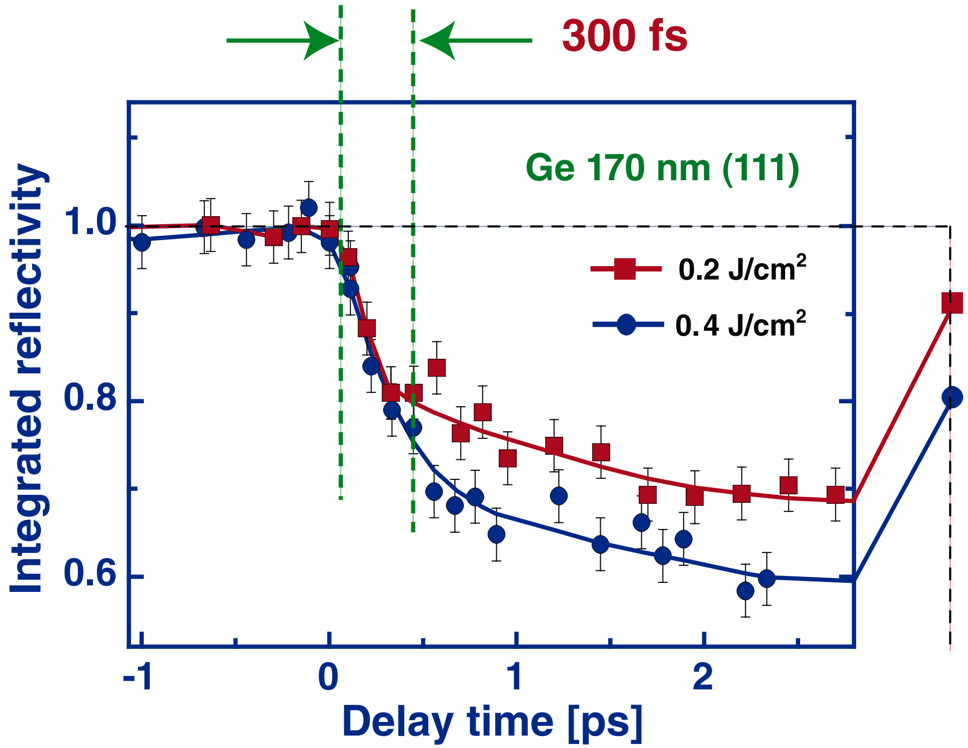
(111) Diffraction lines recorded on the CCD detector

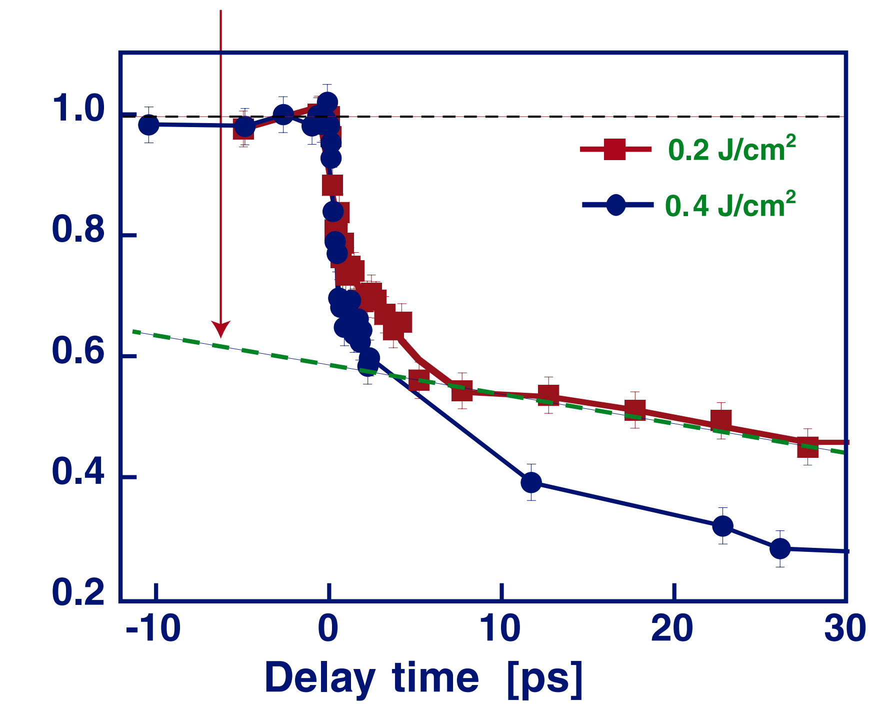


Ultrafast non-thermal melting of Ge

(111) Ge:Si heterostructures







X-ray diffraction experiments on Bismuth

Case study Bismuth

List of interesting properties

Structural distortion

Quasi-Peierls transition

Polymorphism

Lattice instabilities

A_{1g}-phonon modes (totally symmetric)

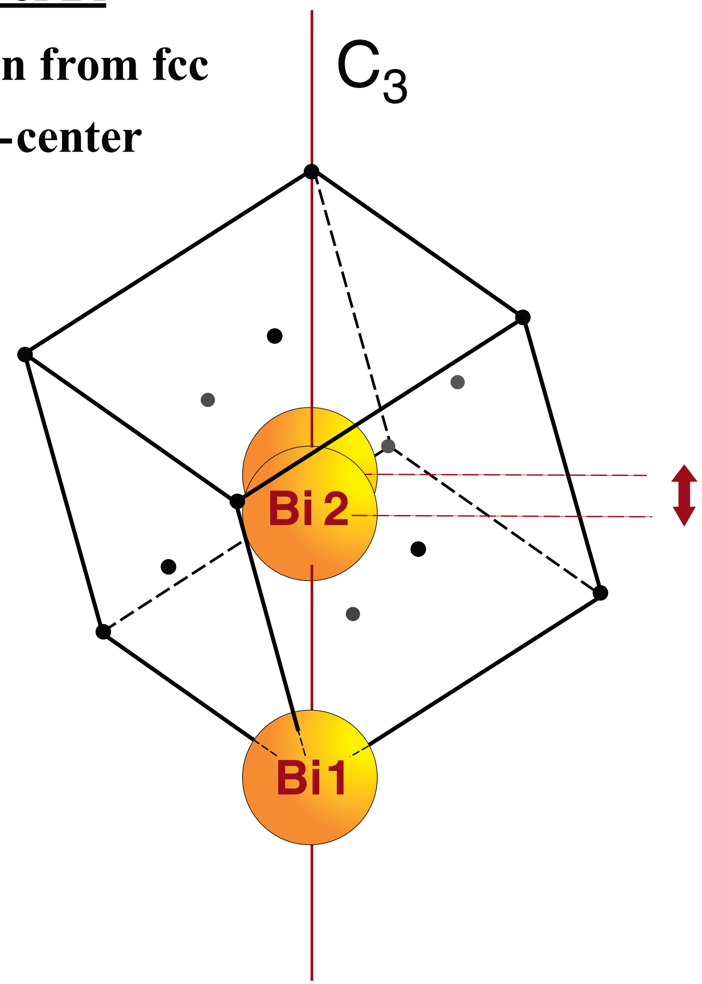
Strong diamagnetism

Semiconductor-semimetal-metal transitions

Crystal structure of Bi

a) Slight distortion from fcc

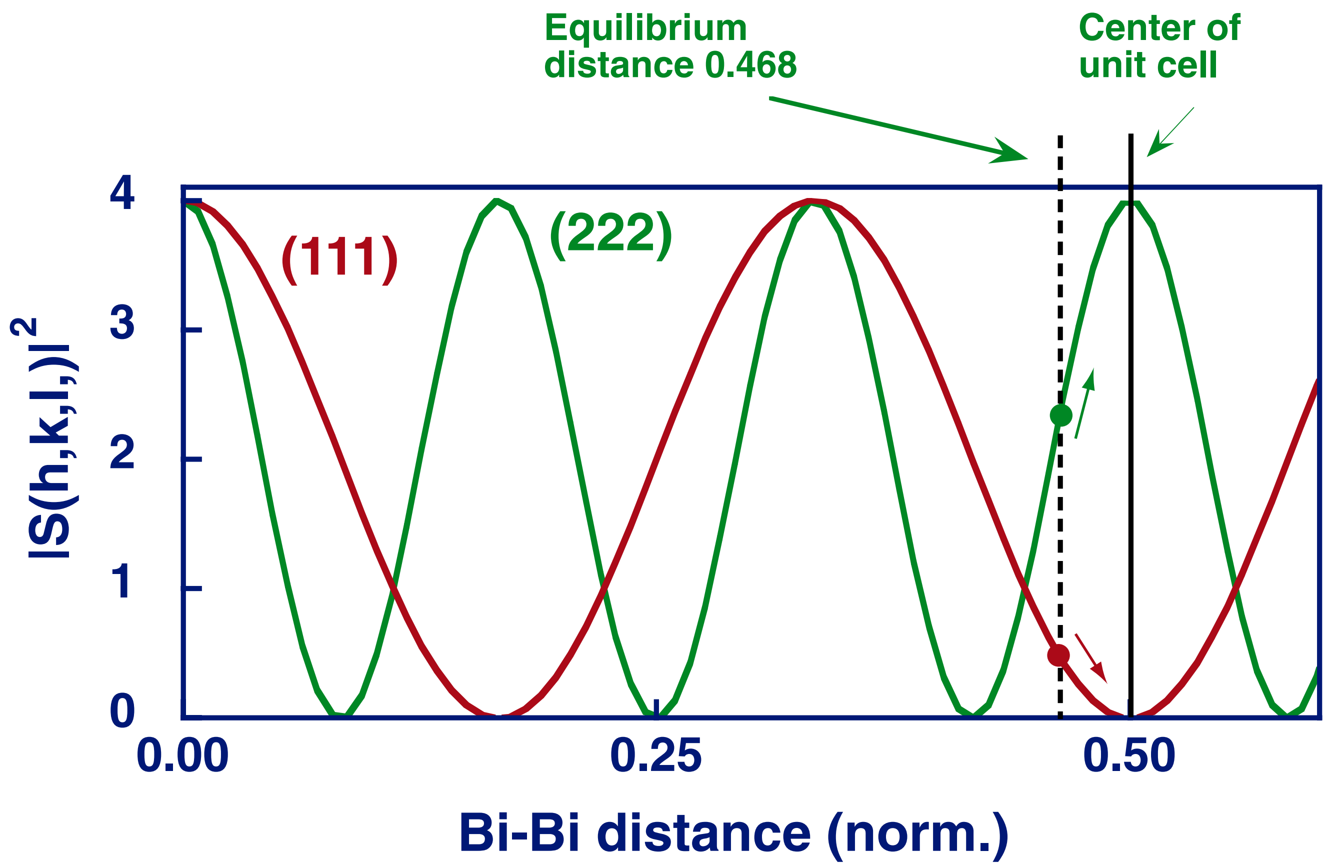
b) Bi2 slightly off-center



Key point:

Bi-Bi equilibrium distance easily affected by any kind of perturbation

X-ray geometrical structure factor of Bismuth



Decrease in the (111) X-ray diffraction

Increase in the (222) X-ray diffraction

... upon increase in the Bi-Bi distance

We regret that some material cannot be shown here in order to avoid conflict with a certain publisher.

We detect

coherent optical phonons

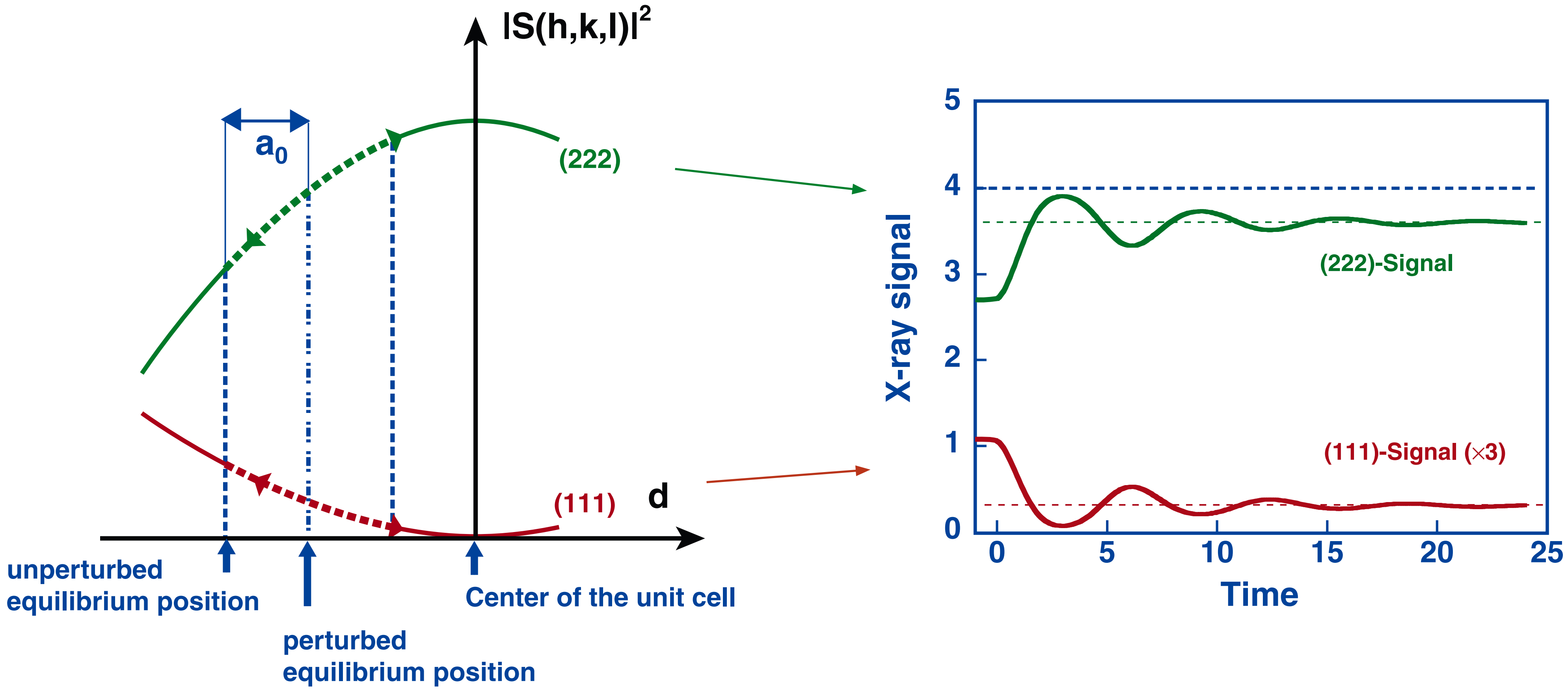
A_{1g}-optical phonon mode:

ν_{A1g} = 2.92 THz; T = 342 fs (unperturbed Bi)

ν_{obs} = 2.14 THz; T = 467 fs (highly excited Bi)

Displacive excitation of optical phonons

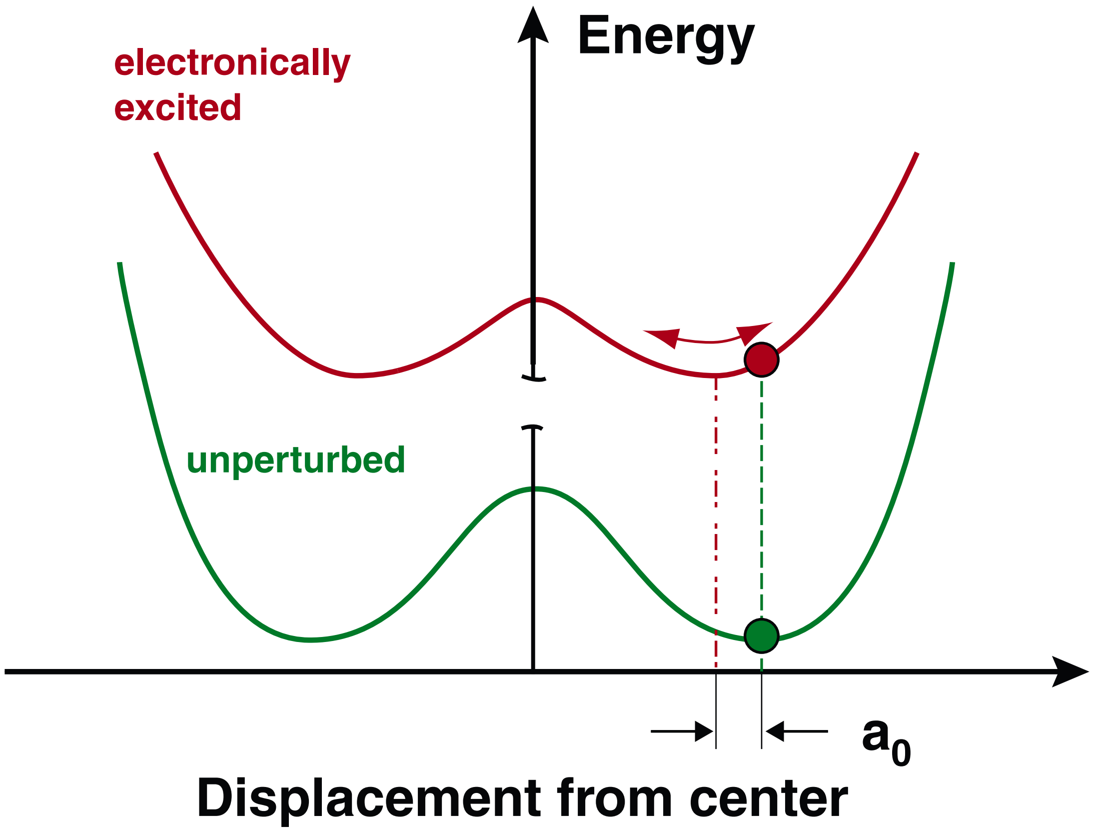
Interpretation of the oscillations

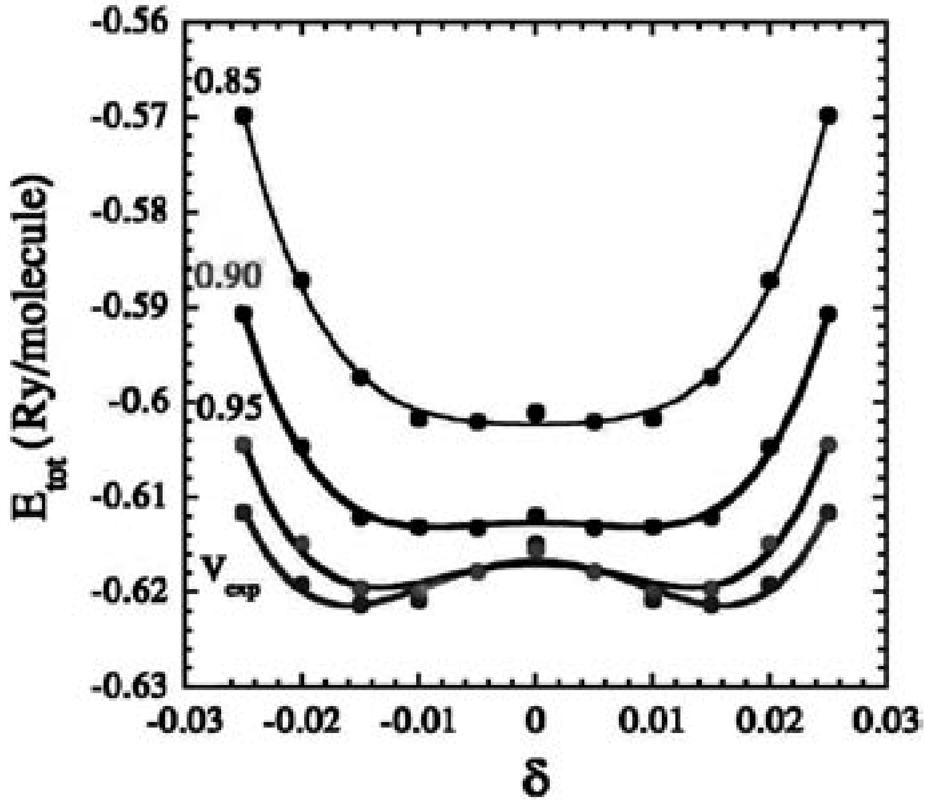


Estimated oscillation amplitude: $a_0 \approx 0.15 .. 0.2$ Å

(nearest neighbor distance: 3.47 Å)

Displacive excitation of the A_{1g}-mode





Total energy (a) at E_F as a function of the internal displacement for different volumes

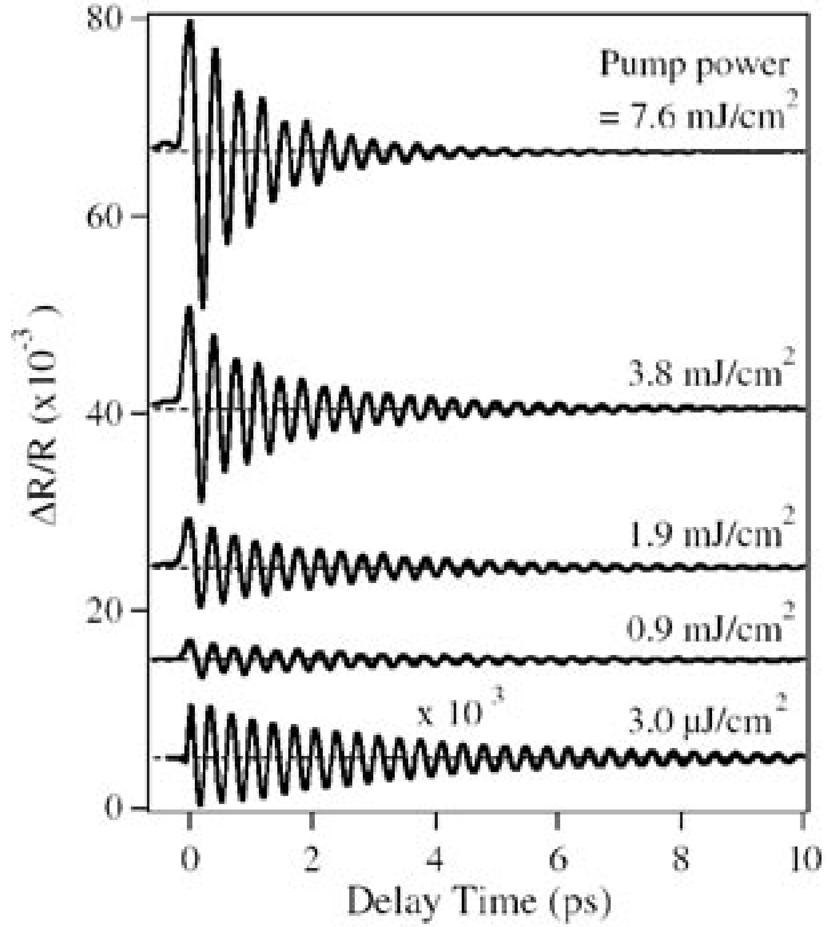
A. B. Shick et al.

Phys. Rev. B 60, 15485 (1999)

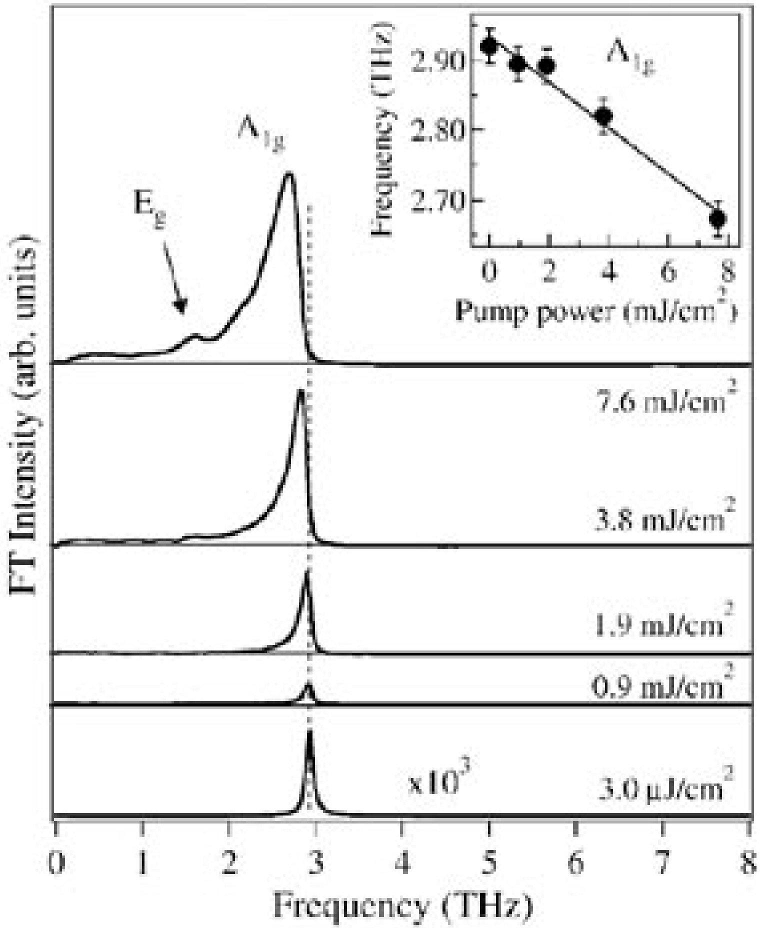
Optical phonon spectra from transient reflectivity

Time-resolved reflectivity of the intensely photoexcited Bi

(Hase et al. PRL <u>88</u>, 067401(2002))



The transient reflectivity change for Bi at various pump power densities. The dashed lines correspond to the zero level on the reflectivity change of each time-domain signal.

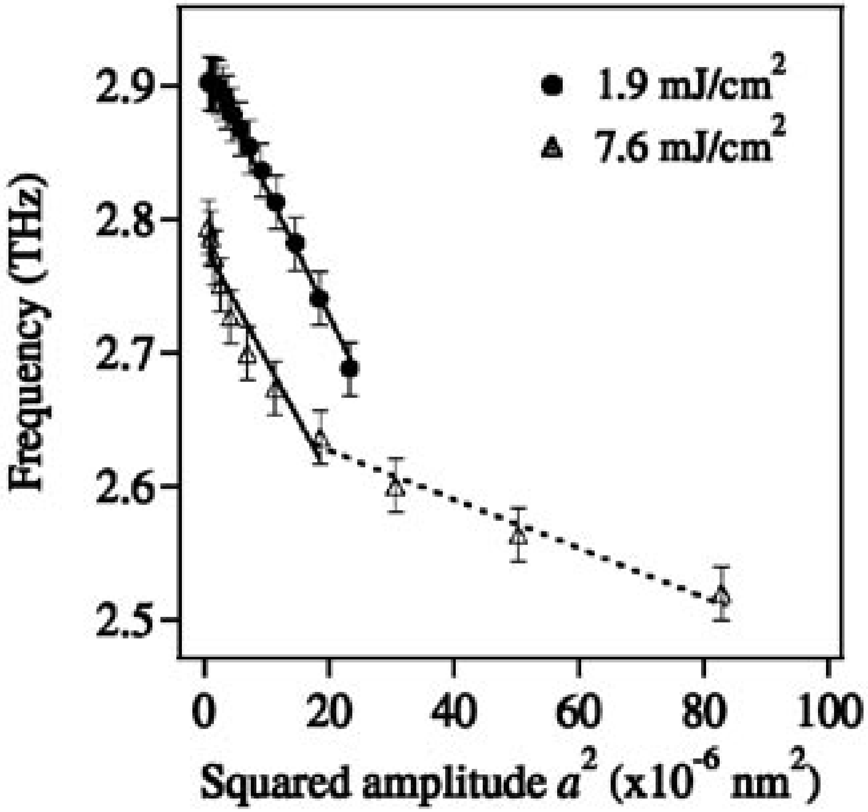


Fourier transformed spectra obtained from time-domain data in Fig. 1. The dashed line represents the A_{1g} frequency of 2.92 THz. Inset: the peak frequency of the A_{1g} mode as a function of the pump power density. The solid line represents a numerical fitting of the data by a linear function.

Discussion of the phonon frequency

Time-resolved reflectivity of the intensely photoexcited Bi

(Hase et al. PRL 88, 067401(2002))



Observed down-shift of the A_{1g}-phonon frequency is attributed to anhamonicity and mode softening.

Observed down-shift (Hase et al.):

2.92 THz --> 2.45 THz

Estimated phonon amplitude:

a (= a_0) ≤ 0.09 Å

Observed down-shifted frequency:

(X-ray experiments)

ν_{obs} = 2.14 THz

Estimated phonon amplitude:

a_0 ≈ 0.15 .. 0.2 Å

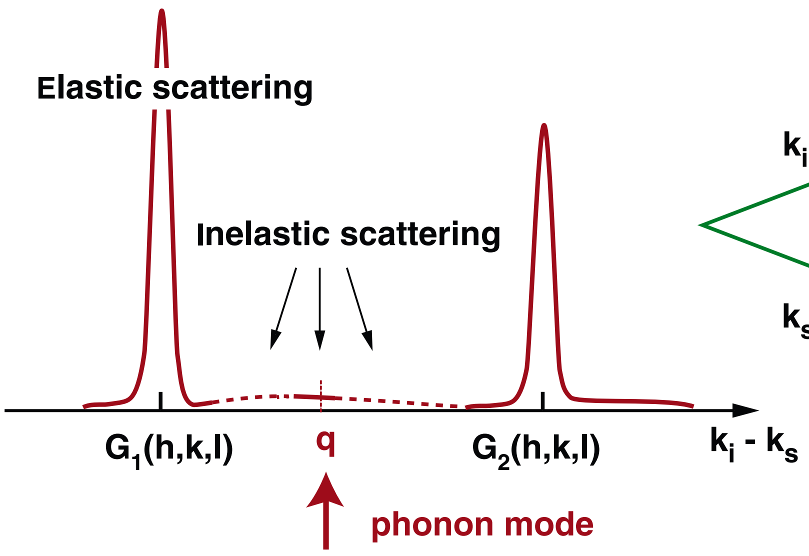
Conclusion

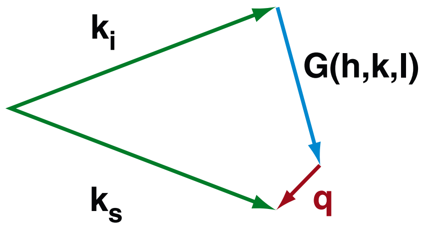
We have demonstrated:

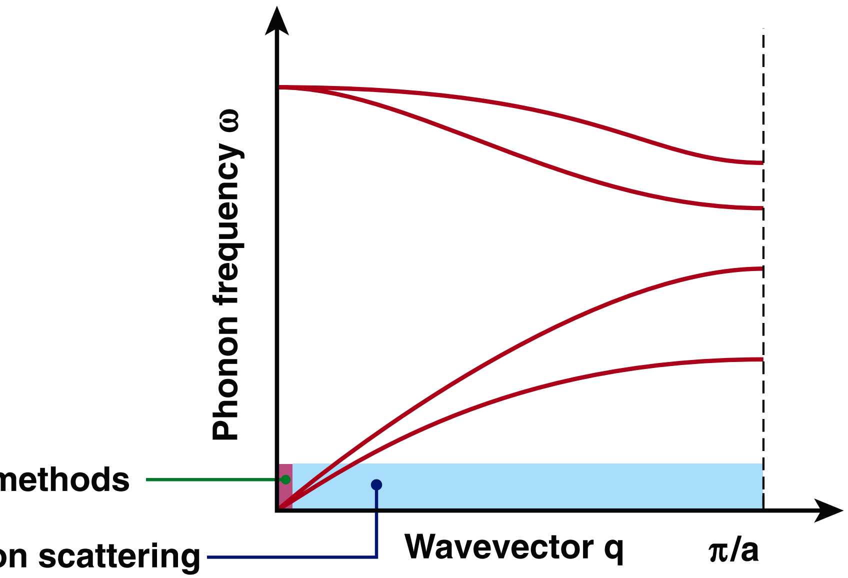
- Multi-keV X-ray diffraction with subpicosecond time resolution
- X-ray measurement accuracy of a few percent
- Non-thermal melting and rapid thermal melting processes
- Capability of measuring optical phonon oscillations via X-ray diffraction
- Borderline between very strongly driven lattice vibrations and the onset of structural transitions

Outlook: Lattice dynamics

Measurement of short wavelength phonons

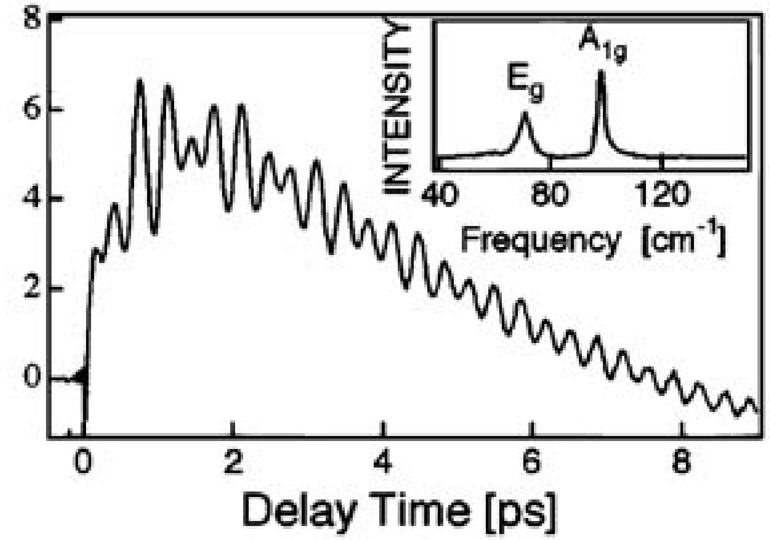






Conventional inelast. X-ray scattering:

Requires energy resolution $\Delta E/E \approx 10^{-6}$



Femtosecond X-ray scattering:

Direct measurement
of the phonon frequency