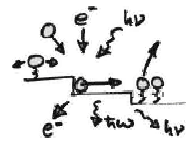
Laser-excited coherent and incoherent lattice vibrations studied by femtosecond X-ray diffraction



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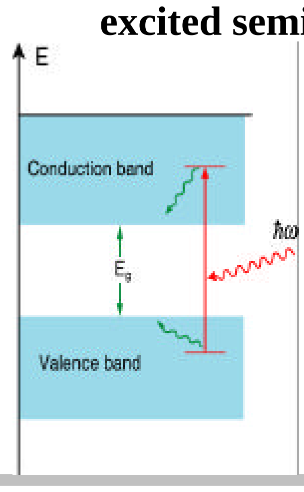
Summary

Ultrashort x-ray pulses offer a unique combination of atomic-scale spatial and temporal resolution, which permits /direct/ measurements of structural transients on an ultrafast time-scale. We have applied time-resolved X-ray diffraction using ultrashort, multi-keV X-ray pulses to study coherent and incoherent lattice vibrations in optically excited semiconductors and metals. Sub-300-femtosecond bursts of Ti-K_α - radiation (4.51 keV) are produced by focusing 120 fs laser pulses onto the surface of a moving Titanium-wire. In an optical pump / X-ray probe configuration transient changes in X-ray diffraction from (111)-oriented, single-crystalline thin films of Germanium and Bismuth have been measured. In Germanium the transient Debye-Waller effect allowed to follow directly the energy transfer from hot electrons to the lattice. In Bismuth the excitation of large amplitude coherent optical phonons is evidenced by a periodic modulation of the diffraction signals.

Energy relaxation

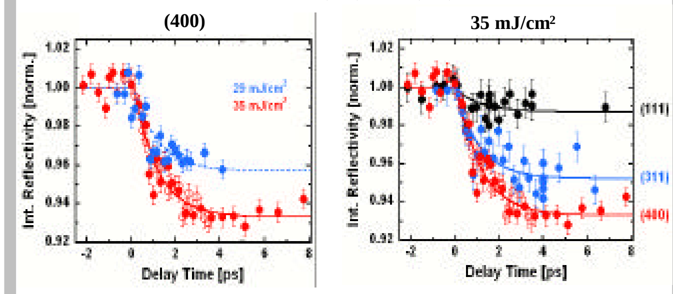
Dissipation of energy in optically excited semiconductors



1. Electron excitation
   <= fs-laser pulse
2. Relaxation and dissipation of electrons energy
   <=Electron-electron interaction
   <=Electron-phonon interaction

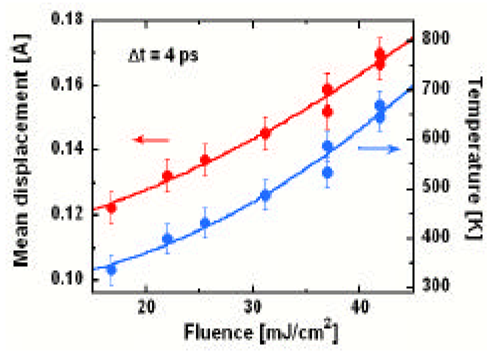
=> Heating of lattice

Transient DW-effect: *Lattice heating*



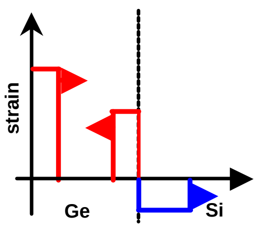
energy relaxation time: 1.1 ps

it is Debey-Waller !!

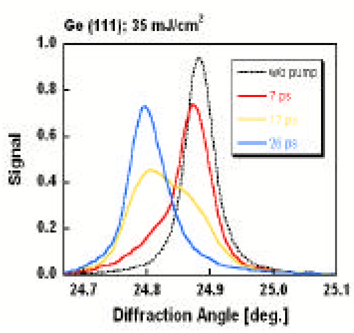


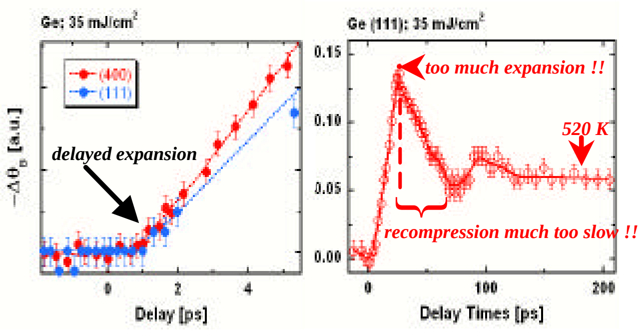
Coherent acoustic phonons

Thermoacoustic response: *Coherent acoustic phonons*



A. Cavalleri et al., PRL **85**, 586 (2000)

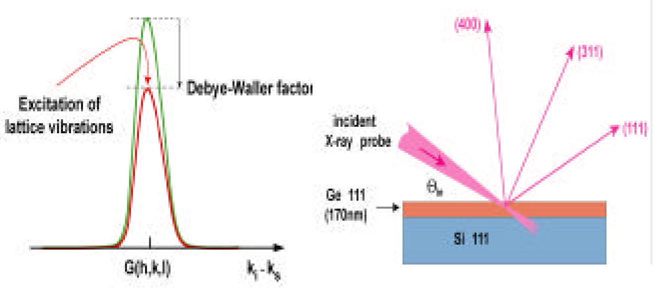




no *instantaneous* stress contribution

- **not only *thermal* stress**
- **not just *simple acoustics***

Debye-Waller factor



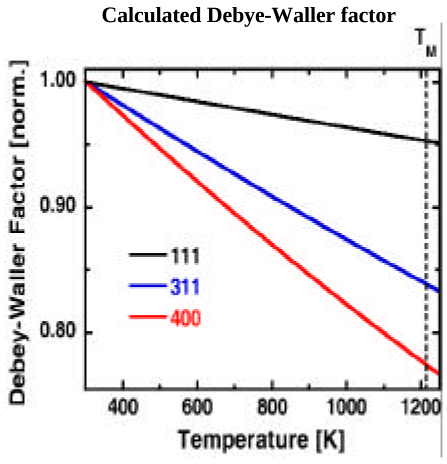
$$DW = \exp\left[-\frac{4}{3}\pi^2 \langle u^2 \rangle \Big/ d_{hkl}^2\right]$$

111-surface:
111-refl.: Þ Q_{in} = 24.9°; Q_{out} = 24.9°
311-refl.: Þ Q_{in} = 24.2°; Q_{out} = 48.8°
400-refl.: Þ Q_{in} = 21.6°; Q_{out} = 96.8°

Mean atomic displacement

No change of FWHM!

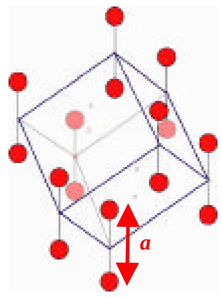
Calculated Debye-Waller factor



Coherent optical phonons

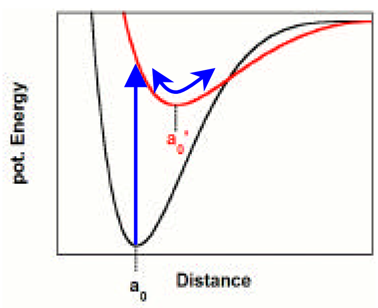
Bismuth

- semi-metal
- rhombohedral structure:
  - slightly distorted fcc
  - di-atomic basis
- excitation of *coherent optical phonons*



Key point:
Bi-Bi distance ***a*** (**0.468** × body diagonal ***c***)
readily affected by external pertubation: pressure, **optical excitation**

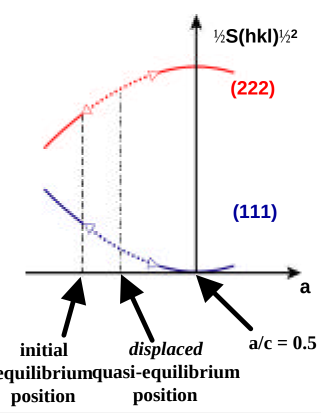
Displacive excitation of coherent phonons



New quasi-equilibrium position !

New potential energy curve !

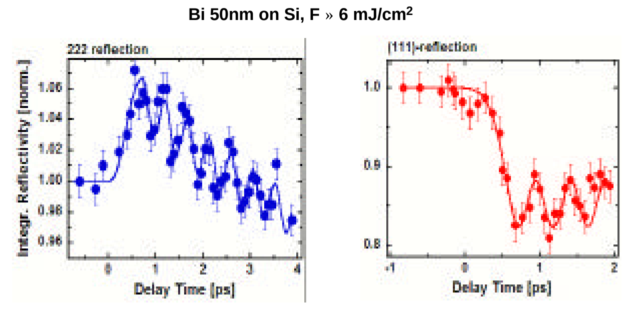
Geometrical structure factor of Bi



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

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

Coherent optical phonons



A_{1g} optical mode:
n_{abs} = **2.14 THz** (470 fs)
ν_0 = 2.92 THz (342 fs)

softening & anharmonicity

phonon amplitude: **0,15 – 0,25 Å**

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

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