Time-resolved Diffuse X-ray Scattering

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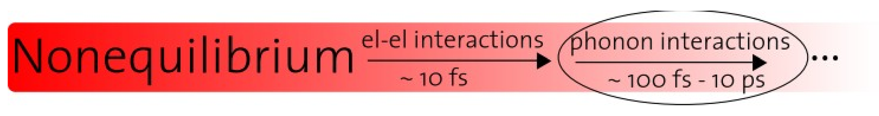
Motivation

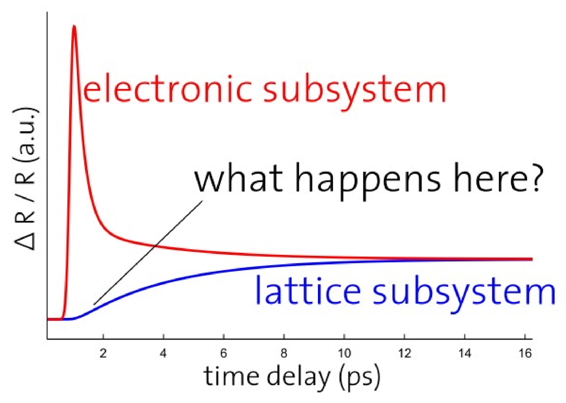
Relaxation processes of hot carriers play a crucial role in mainy highly interesting condensed matter systems (photovoltaic cells and semiconductors in general, superconductors, charge density wave systems, thermoelectrics...)

Relaxation dynamics governed by:
- Electron-electron scattering
- Electron-phonon scattering
- Phonon interaction (anharmonic coupling, decay)



Phonon timescale can be **< 100 fs** (typical vibration periods of lattice ions)

Optical pump-probe: Enough time resolution, but no information about momentum:



Generic optical pump-probe experiment

SwissFEL pulses will present an ideal opportunity to image nonequilibrium phonon processes with sufficient **temporal** and, because of its atomic scale wavelengths, **momentum** resolution.

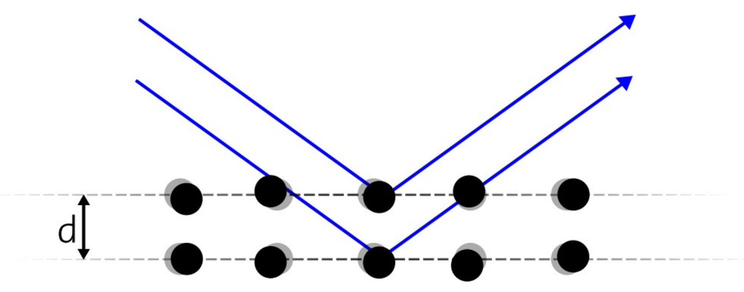
X-ray Diffuse Scattering..

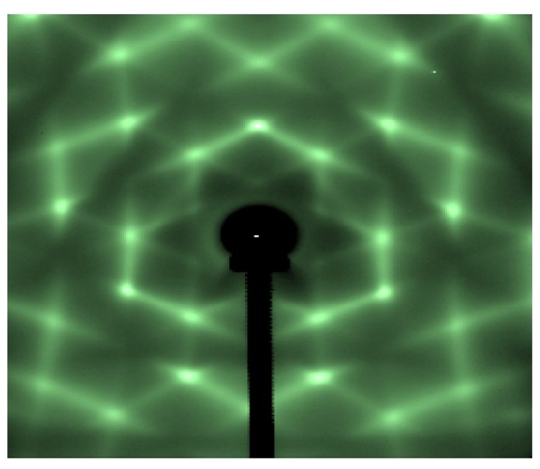
With low intensity X-ray sources, only Bragg scattering is observed: lattice quilibrium positions are imaged.

Temperature dependence of Bragg peaks: Debye-Waller factor.
The intensity that is lost in the Bragg peaks with rising temperature, or equivalently, with rising population of the phonon branches, is distributed into the diffuse scattering image.

$$\vec{k}_{in} - \vec{k}_{out} = \vec{G}_{hkl} + \vec{q}_{phonon}$$

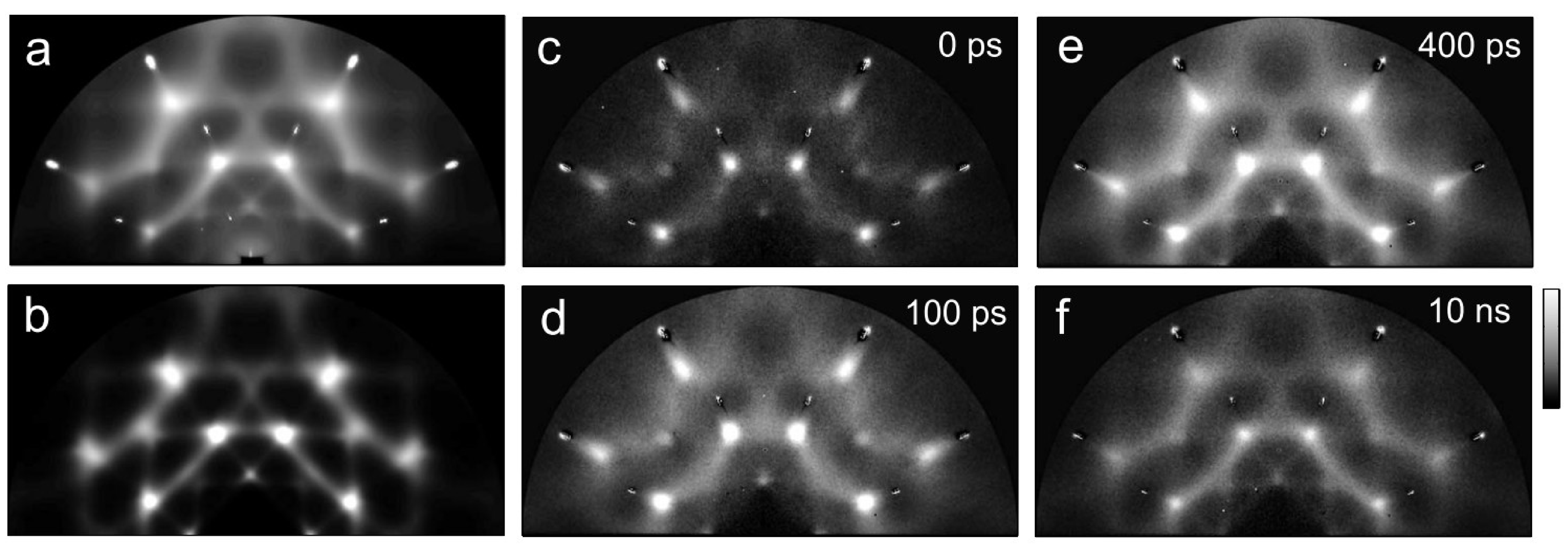
Lattice with phonons





Example of a thermal diffuse X-ray scattering image taken in transmission geometry at the Advanced Photon Source (APS) by Holt et al. [1]. Sample: Si(111).
- Intensity pattern: Inelastic scattering of thermal phonons.
- Information of thermal phonon population and phonon dispersion.

.. to study nonequilibrium phonons

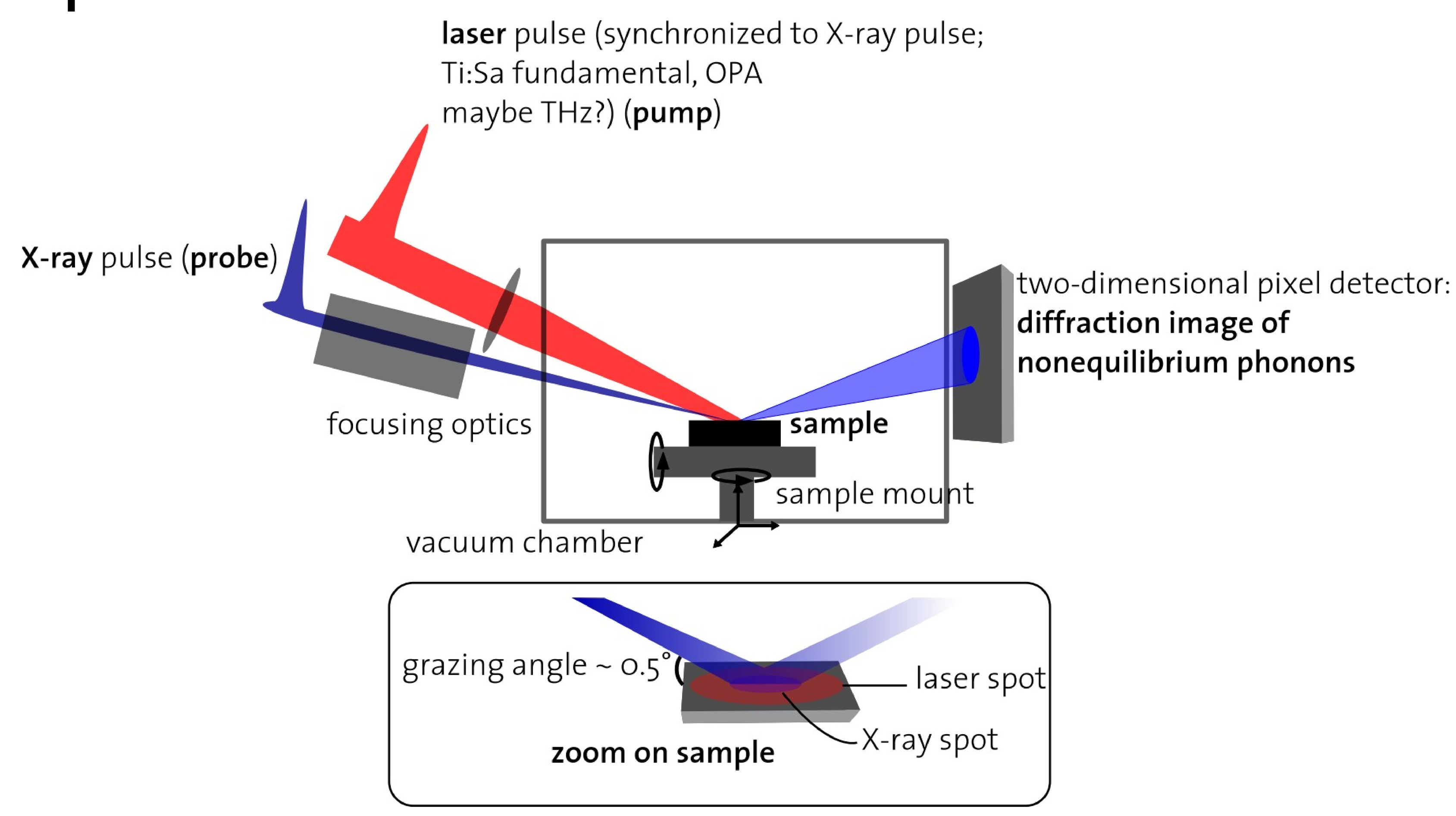


InP Data taken by Trigo et al. [2] at APS with a time resolution of 1 ps and 10^{10} photons/pulse at 15 keV.

a) Thermal diffuse scattering pattern
b) Calculated thermal diffuse scattering pattern from a Born model
c) – f) Images of the nonequilibrium phonon population with time delay t between laser excitation and X-ray probe. The differences between these images show that equilibrium could not yet have been reached.

For sub-picosecond time delays, the phonon distribution does not correspond to the Bose population of the phonon branches, but reveals **nonequilibrium dynamics**.

Experiment



X-ray beam is focused onto the sample in a grazing incidence geometry in order to ensure the entire probed volume is photoexcited by the laser pulse.

- Laser beam blocked: Thermal diffuse scattering image of the equilibrium phonon distribution on the two-dimensional pixel detector. The incoherent atomic motion throughout the Brillouin zone can be visualized because of the high X-ray photon flux of the SwissFEL pulses.

- Laser beam unblocked: Diffraction image of the nonequilibrium phonon distribution can be recorded with an adjustable time delay between laser pulse (pump) and X-ray pulse (probe).

SwissFEL Parameters

Parameter		Unit	Require -ment	Motivation /Remarks
Energy		keV	12	Sufficient q-resolution for small wavelength phonons needed (q-resolution also limited by X-ray penetration depth)
	stability	%	10	Of bandwidth
Bandwidth		%	< 1	
	stability	% bw	< 1	
Beam position	stability	μm	< 1	High stability needed because of grazing incidence
Beam size		μm	< 10	Grazing incidence requires small spot size (in one dimension, other dimension: several 100μm)
Photons per pulse			1012	Attenuation needed in case of sample damage
	stability		-	Single-Shot measurement
Pulse length		fs (FWHM)	20	Determines time resolution together with pump-probe jitter and pump pulse length
	stability	%	10	
Pulse arrival time	stability	fs	< 20	With respect to pump laser
Beam parameter changes during experiment				
Energy	range	eV	-	No change needed
	step	eV	-	No change needed
	scan	eV / sec	-	No change needed
Beam size			-	No change needed
Pulse length			-	No change needed
Beam geometry				
Beam slope	maximum	μrad	300	
Working distance	minimum	mm	500	vacuum chamber, sufficient space to guide laser beam onto sample (distance detector – last permanent optic: around 1000 mm). Roughly the same sample environment as needed by FEMTO group for several experiments.

References

[1] M. Holt et al., Phys. Rev. Lett. 83, 3317 (1999)

[2] M. Trigo et al., Phys. Rev. B 82, 235205 (2010)