Irreversible material dynamics studied with time-resolved X-ray scattering

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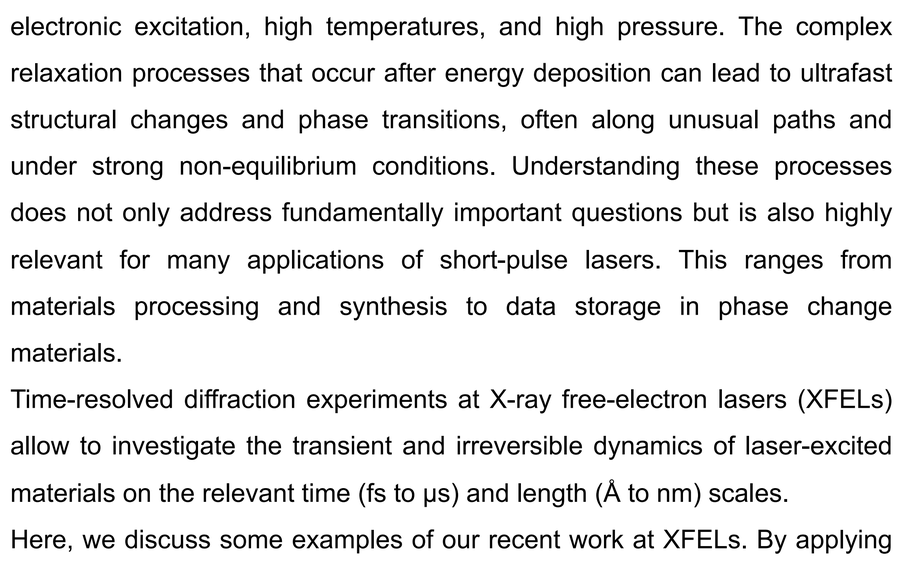
Introduction

Irradiation of a solid with an ultrashort laser pulse leads to states of strong electronic excitation, high temperatures, and high pressure. The complex relaxation processes that occur after energy deposition can lead to ultrafast structural changes and phase transitions, often along unusual paths and under strong non-equilibrium conditions. Understanding these processes does not only address fundamentally important questions but is also highly relevant for many applications of short-pulse lasers. This ranges from materials processing and synthesis to data storage in phase change materials.

Time-resolved diffraction experiments at X-ray free-electron lasers (XFELs) allow to investigate the transient and irreversible dynamics of laser-excited materials on the relevant time (fs to µs) and length (Å to nm) scales.

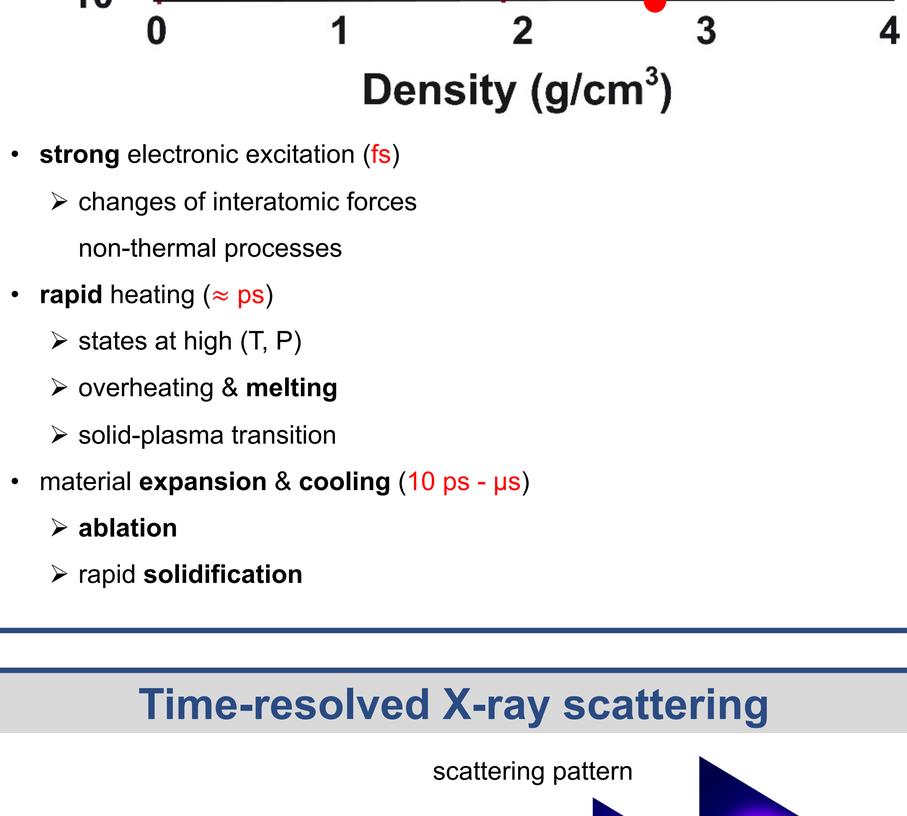
Here, we discuss some examples of our recent work at XFELs. By applying time-resolved wide-angle and small-angle X-ray scattering in combination with a unique thin-film sample design we have investigated the non-equilibrium structural dynamics of various materials after short pulse laser excitation.

Strong laser-excitation of solids



- **strong** electronic excitation (fs)
  - ➢ changes of interatomic forces
    non-thermal processes
- **rapid** heating (≈ ps)
  - ➢ states at high (T, P)
  - ➢ overheating & **melting**
  - ➢ solid-plasma transition
- material **expansion** & **cooling** (10 ps - µs)
  - ➢ **ablation**
  - ➢ rapid **solidification**

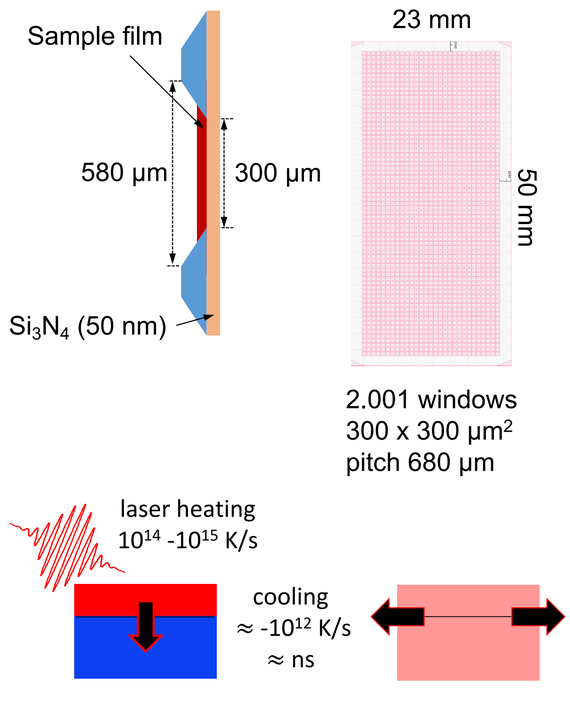
Time-resolved X-ray scattering

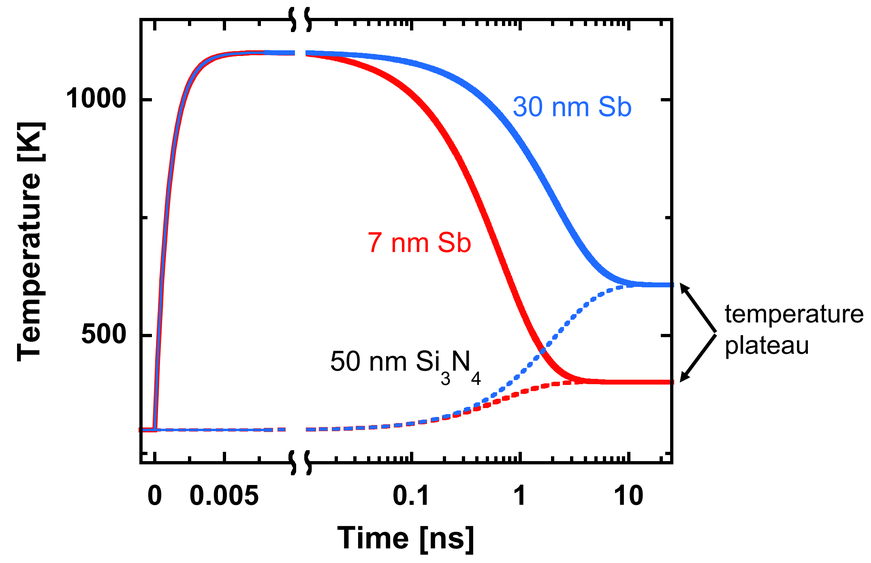


Optical pump – X-ray probe experiment

irreversible dynamics → bright sources → X-ray free-electron lasers

Sample scheme to study irreversible dynamics

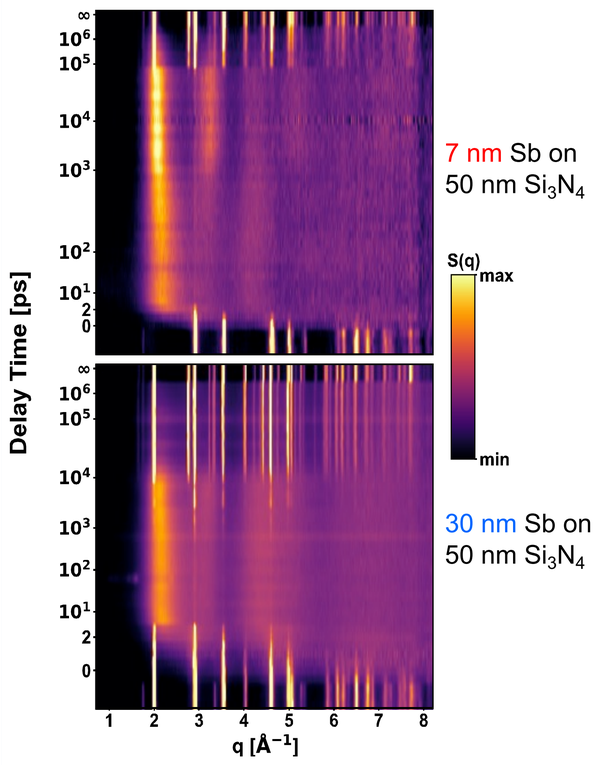


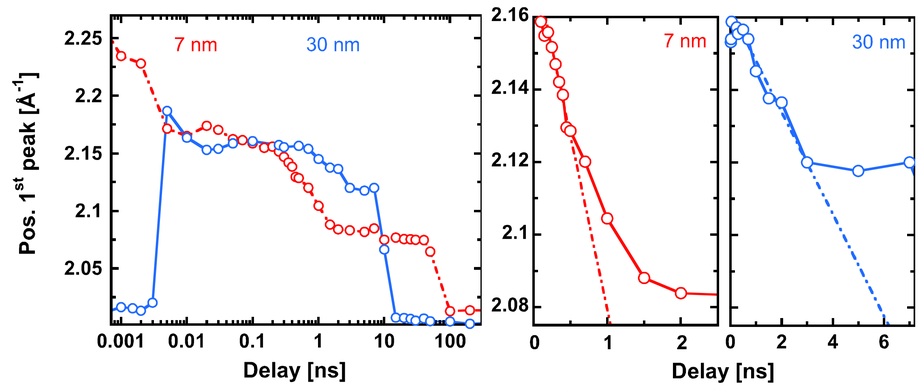


Thin films, deposited on free standing Si_3N_4-membranes

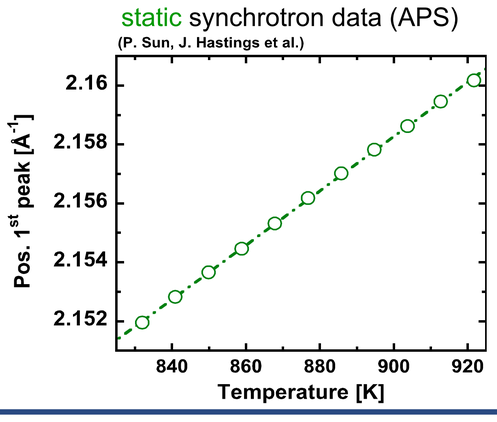
Rapid quenching & isothermal conditions

Rapidly quenched Sb: t.r. WAXS XPP @ LCLS





static synchrotron data (APS)
(P. Sun, J. Hastings et al.)

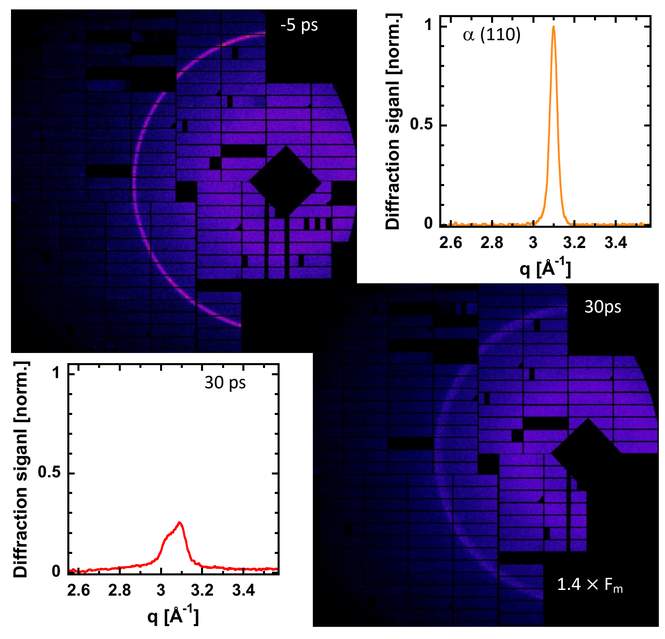


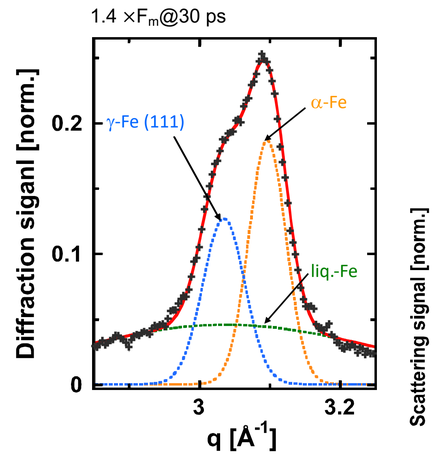
transient cooling rate:

$$\gamma_{7\,nm} = \frac{-0.096\ \text{Å}^{-1}/ns}{9.206\times10^{-5}\ \text{Å}^{-1}/K} = -1.04\times10^{12}\ K/s$$

$$\gamma_{30\,nm} = \frac{-0.015\ \text{Å}^{-1}/ns}{9.206\times10^{-5}\ \text{Å}^{-1}/K} = -1.63\times10^{11}\ K/s$$

Laser-induced phase transitions in thin Fe-films: FXE @ EuXFEL



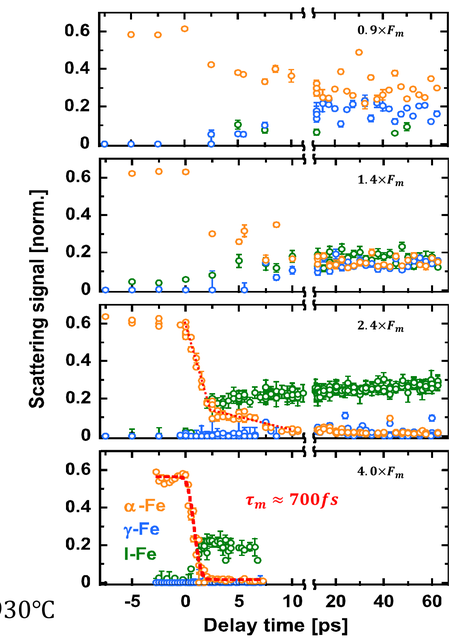


hypothesis: a → g transition

$F_m \mathrel{\widehat{=}} T_R = 20℃ \rightarrow T_m = 1538℃$

$F_{th} \approx 0.6\times F_m$

$\Rightarrow T_{th} \approx T_R + 0.6\times(T_m - T_R) = 930℃$



Acknowledgements

Collaborators

XPP @ LCLS: L. Kremeyer, P. Zalden, R. Sobierajski, J. Antonowicz, S. Wei, P. Sun, J. Hastings, M. Z. Mo, M. Zajac, A. Gleason, T. Sato, D. Zhu

FXE @ EuXFEL: A. Olczak, W. Gawełda, K. Georgarakis, P. Sun, P. Zalden, R. Sobierajski, J. Antonowicz

Use of the Linac Coherent Light Source (LCLS), SLAC National Accelerator Laboratory, is supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under Contract No. DE-AC02-76SF00515.
We acknowledge European XFEL in Schenefeld, Germany, for provision of X-ray free-electron laser beamtime at FXE and would like to thank the staff for their assistance.

Funding

Deutsche Forschungsgemeinschaft DFG in the frame of the Collaborative Research Center SFB1242 'Non-Equilibrium Dynamics of Condensed Matter in the Time Domain'

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