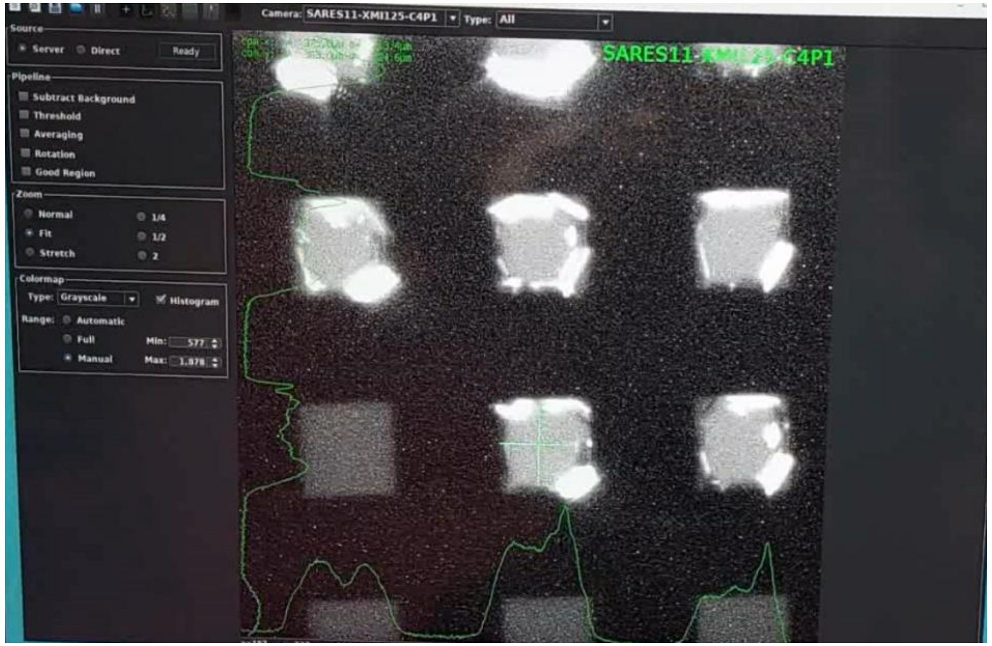
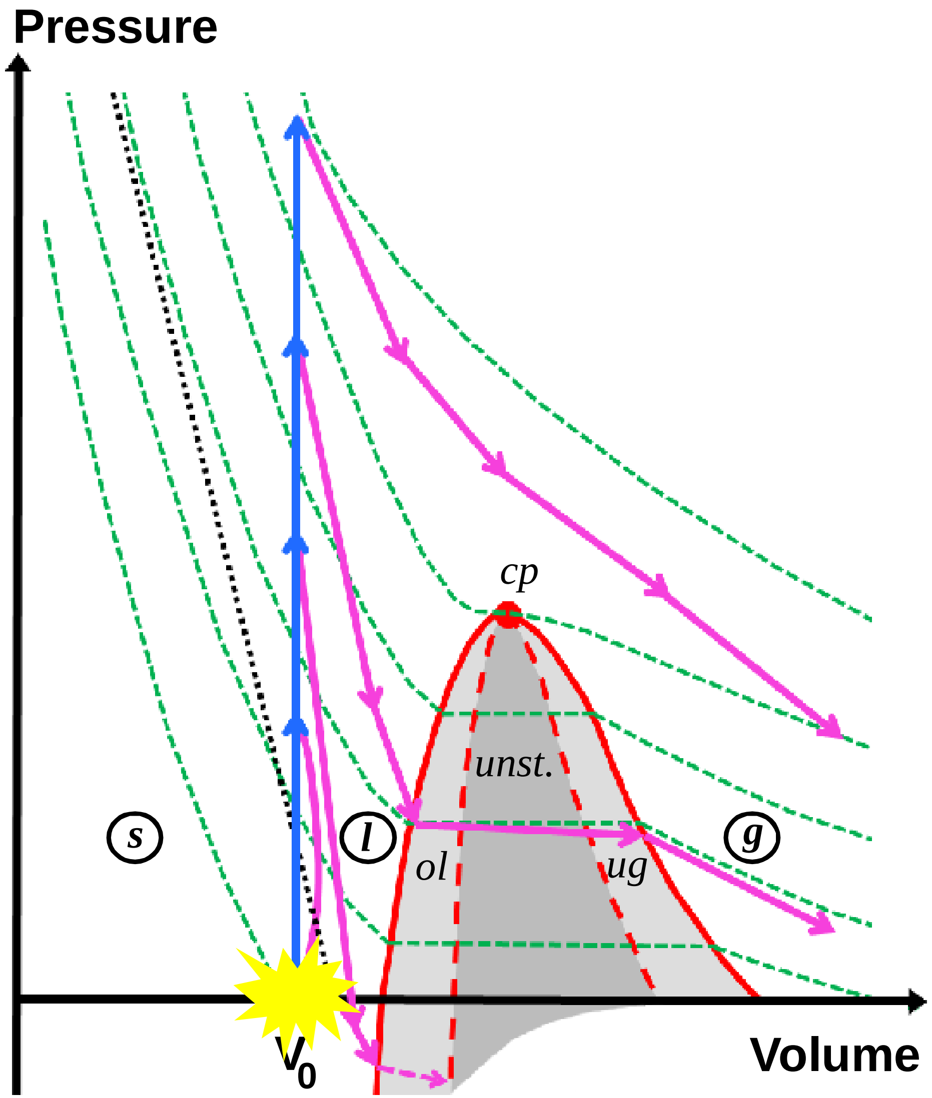
Irreversible* material dynamics studied with time-resolved X-ray scattering

K. Sokolowski-Tinten

*not in a thermodynamical sense



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***
  - ⇒ *supercooling & rapid solidification*
  - ⇒ ***mesoscale structure formation***

⇒ strong thermal & mechanical nonequilibrium

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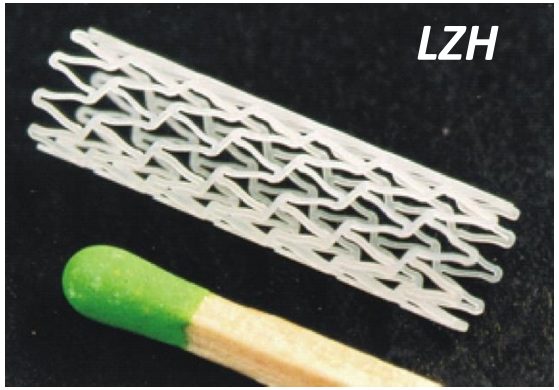
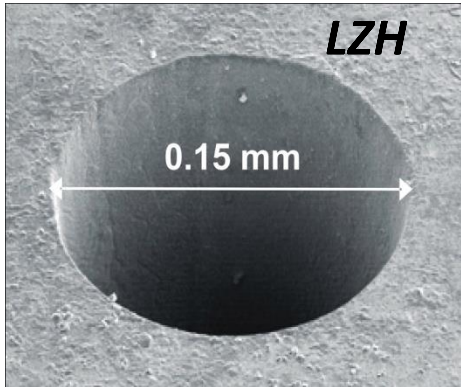
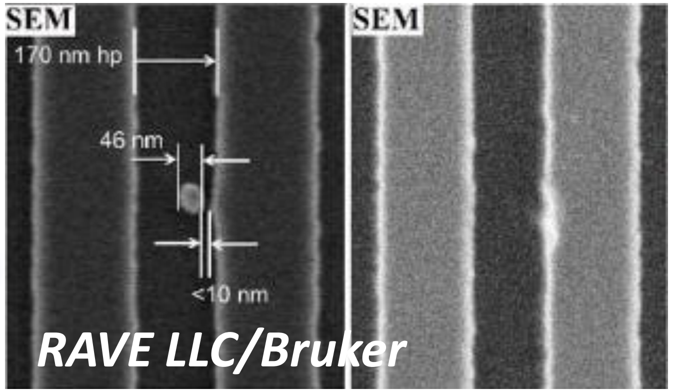
Applications of ultrafast lasers



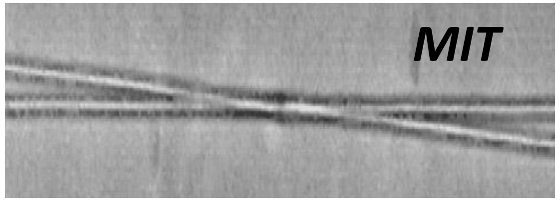
cardiovascular implants



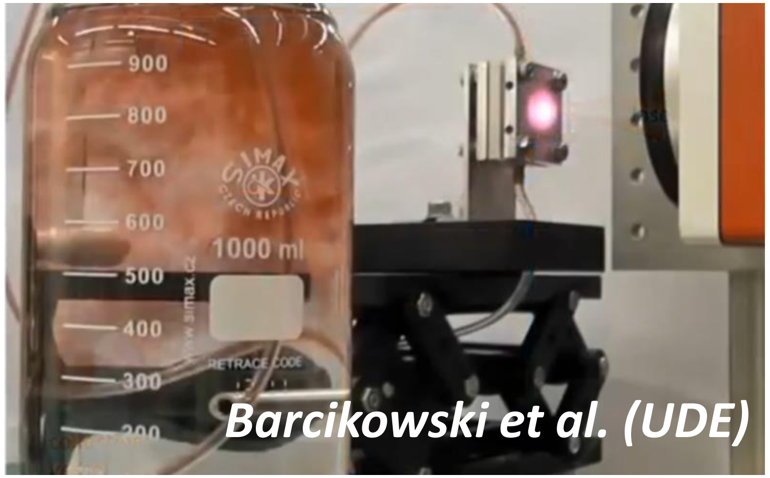
fuel injection nozzel



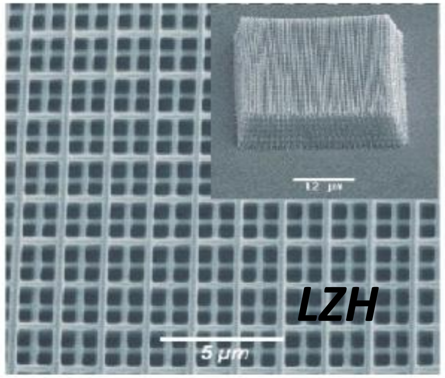
photomask repair



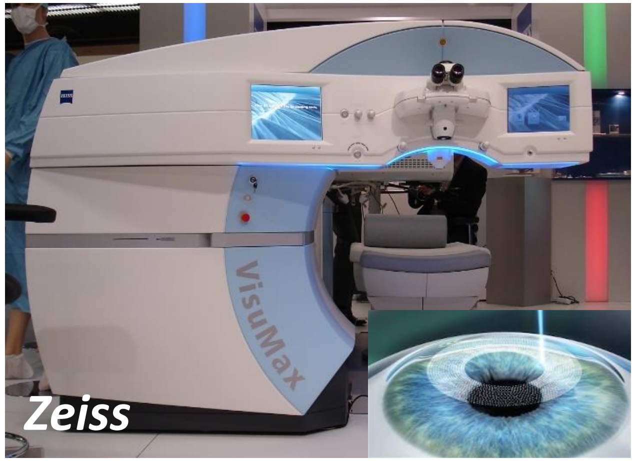
waveguide X-coupler



nano-particle synthesis



photonic crystal



ophtomalogy

⇒ **irreversible** material dynamics

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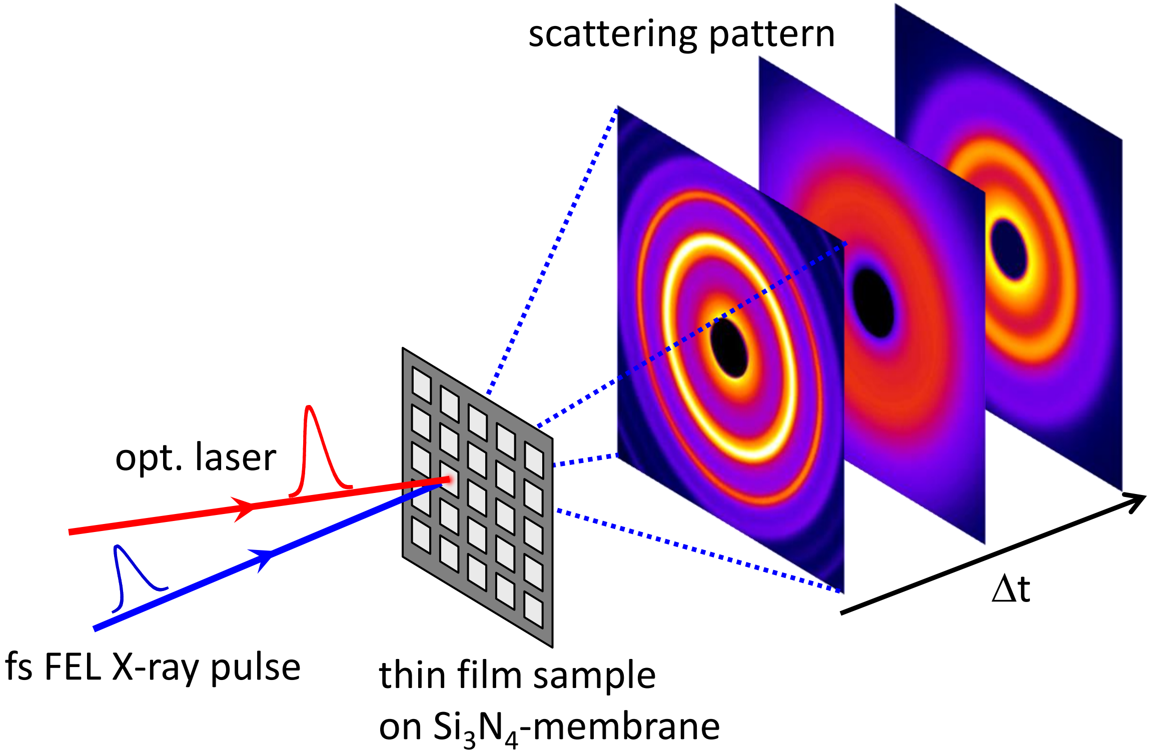
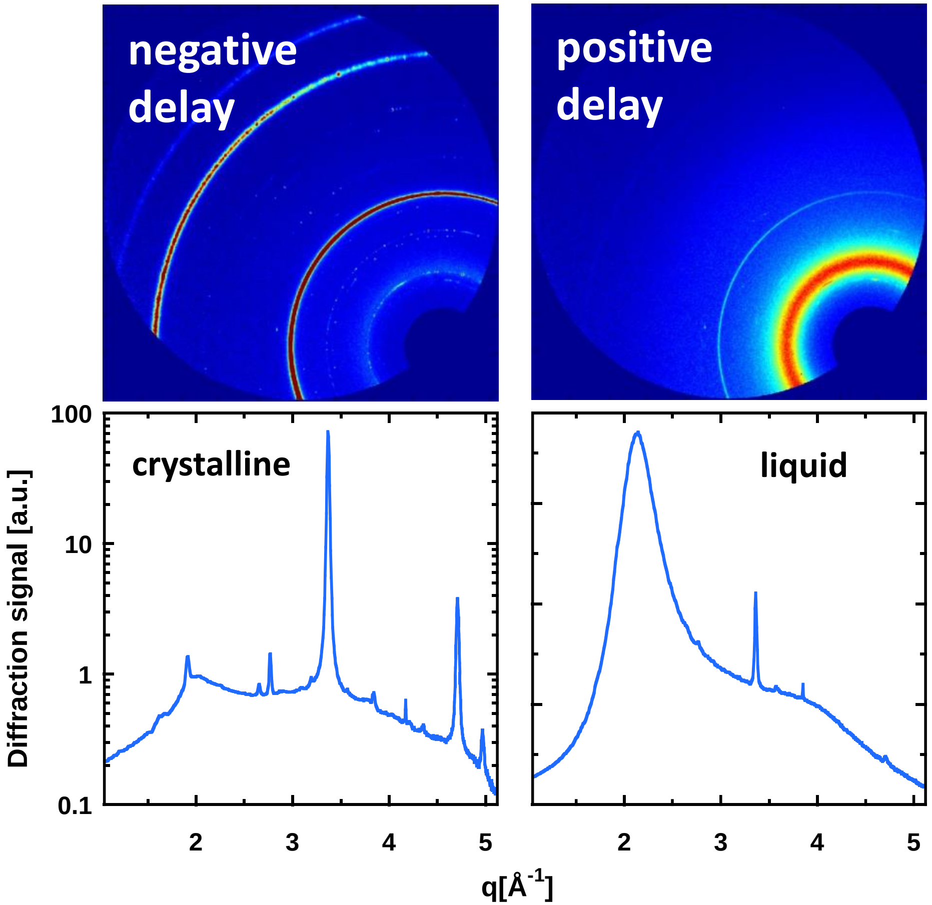
Time-resolved wide- and small angle X-ray scattering: Optical pump – X-ray probe experiment





<u>Outline:</u>

- ultrafast melting in fs-excited Fe
- fs laser ablation
- laser-induced period surface structures (LIPSS)

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Acknowledgements

melting of Fe:

SwissFEL
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PSI
C. Bacellar, C. Cirelli & Alvra-team

IP PAN **R. Sobierajski** et al.

Faculty of Physics, WARSAW UNIVERSITY OF TECHNOLOGY **J. Antonowicz** et al.

European XFEL **P. Zalden**

R. van de Kruijs, UNIVERSITY OF TWENTE.

T. Albert, R. Müller, UNIVERSITÄT DUISBURG ESSEN

ablation:

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M. Jermann, UNIVERSITÄT DUISBURG ESSEN

LIPSS:

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IP PAN **R. Sobierajski** et al.

uOttawa **A. Rudenko**

J.-P. Colombier, LABORATOIRE HUBERT CURIEN, UMR • CNRS • 5516 • SAINT-ETIENNE

Y. Sun, SLAC NATIONAL ACCELERATOR LABORATORY

Faculty of Physics, WARSAW UNIVERSITY OF TECHNOLOGY **J. Antonowicz** et al.

D. Kaczmarek, **T. Albert**, UNIVERSITÄT DUISBURG ESSEN

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Non-Equilibrium Dynamics of Condensed Matter in the Time Domain

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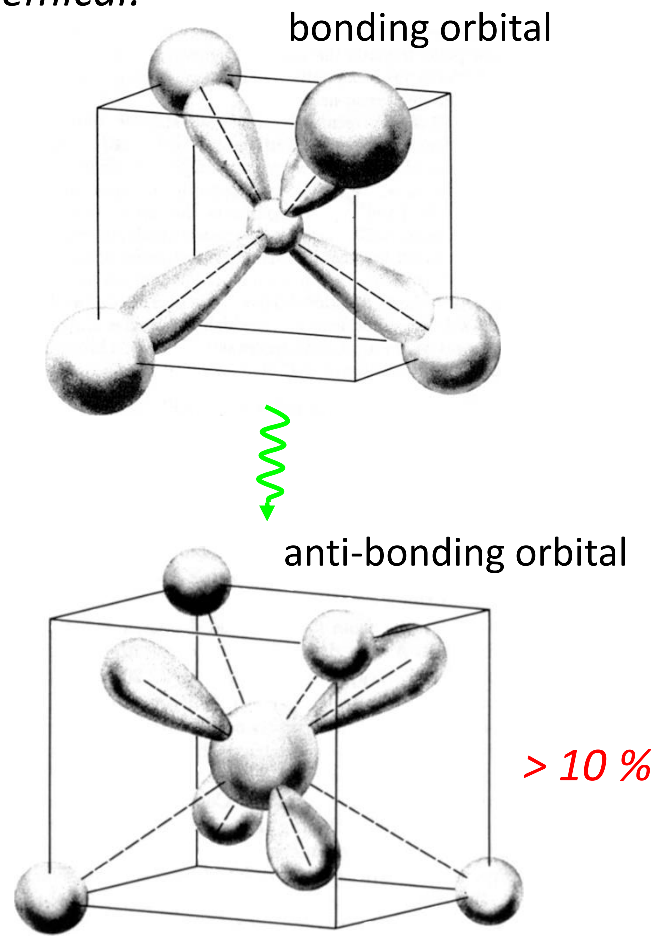
fs laser-induced melting of Fe

Strong laser excitation: Covalently bonded materials vs. metals

covalently bonded materials:

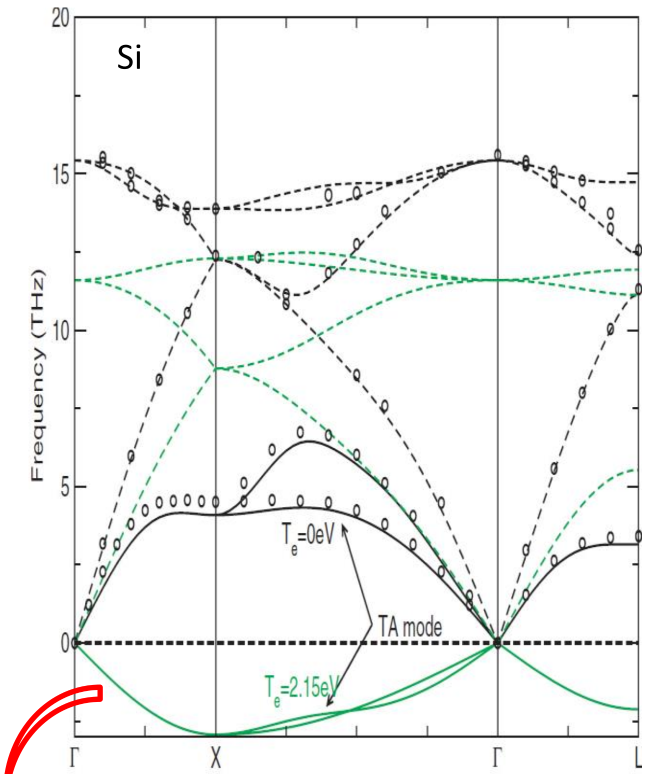
chemical:



> 10 % !!

J.C. Philipps,
"Bonds and bands in semiconductors"

solid state:

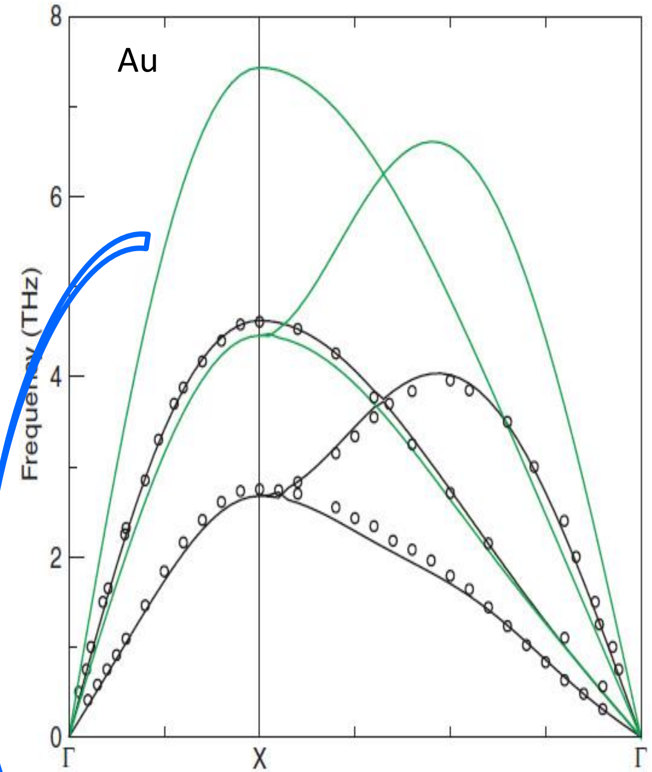


V. Recoules et al., PRL **96**, 055503 (2006)

imaginary frequencies

lattice destabilization
⇒ ***ultrafast disordering***

metals:



phonon **hardenening?!**

see: R. Ernstorfer et al., Science **323**, 1033 (2009).

melting of ("simple") metals:
thermal process!!
electron phonon coupling

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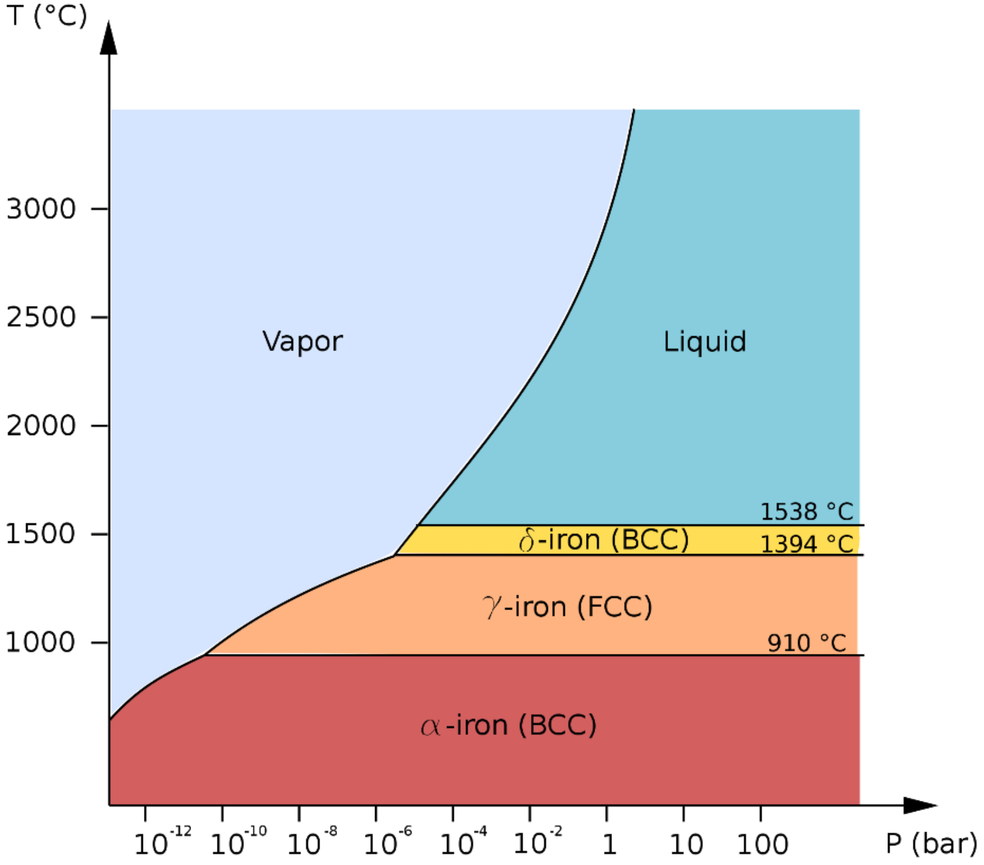
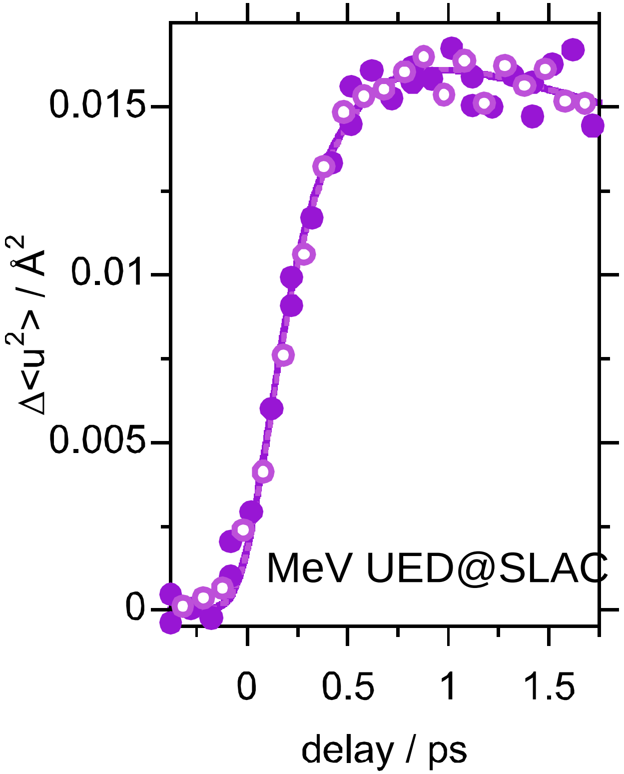
Why Fe?

reason 1: solid-solid transitions



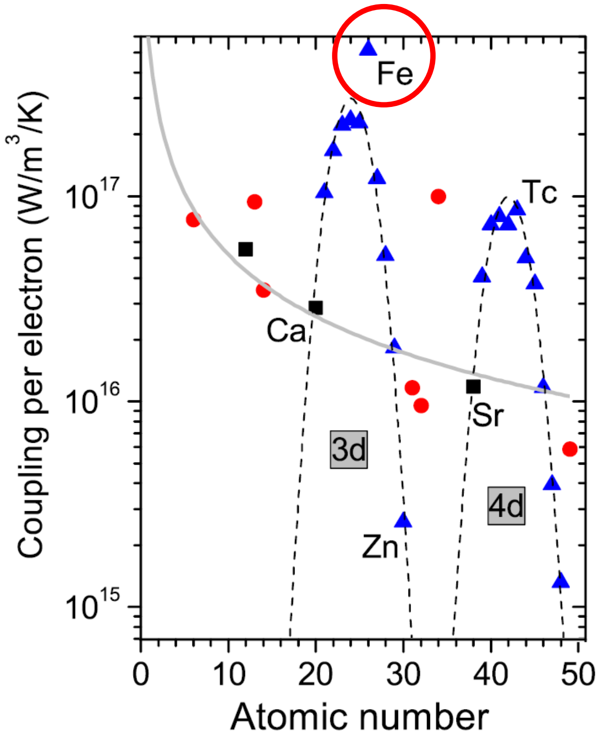
=> talk by R. Sobierajski

reason 2: very strong electron-phonon coupling



$$G \approx 1.5 \times 10^{18}\ \mathrm{W/(m^3K)}$$

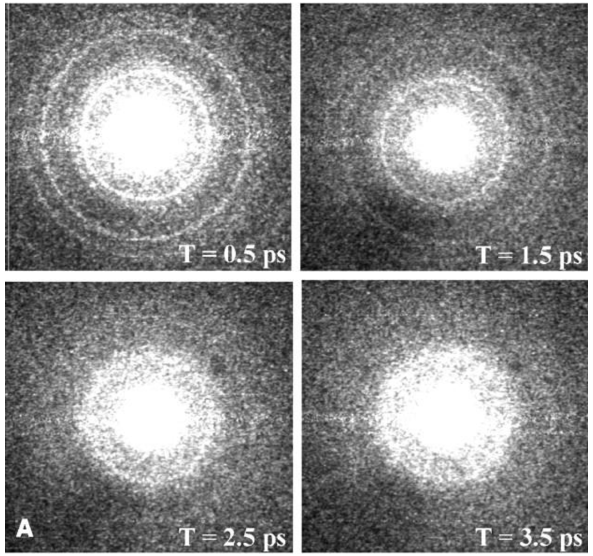
Rothenbach et al., PRB **100**, 174301 (2019)



Medvedev et al., PRB **102**, 064302 (2020)

Ultrafast melting of metals

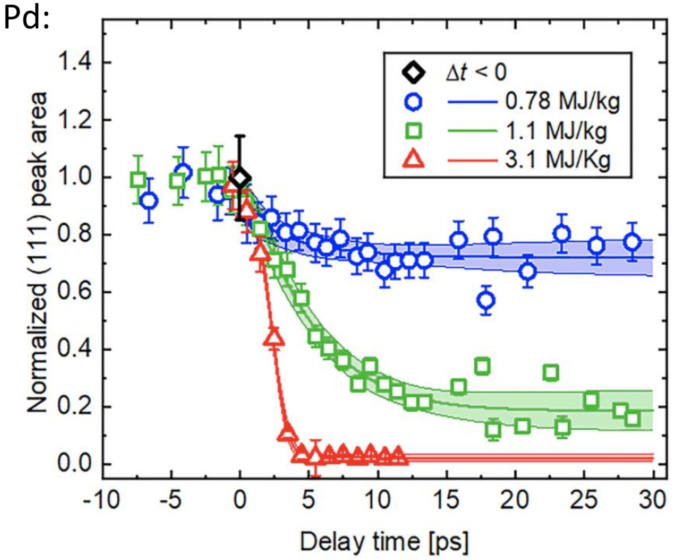
Al:



Siwick et al. Science **302**, 1382 (2003)

$\tau_{\min} \approx 3.5$ ps

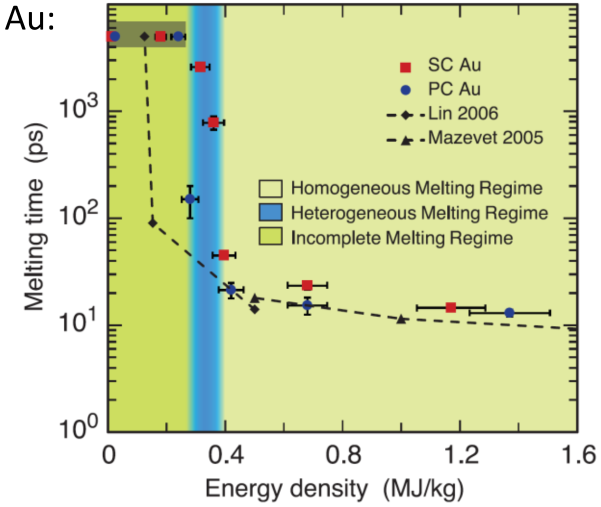
Pd:



Antonowicz et al. Acta Mat. **276**, 12043 (2024)

$\tau_{\min} \approx 2.5$ ps

Au:



Mo et al. Science **360**, 1451 (2018)
see also: Assefa et al., Sci. Adv. **6**, eaax2445 (2020)

$\tau_{\min} \approx 10 - 12$ ps

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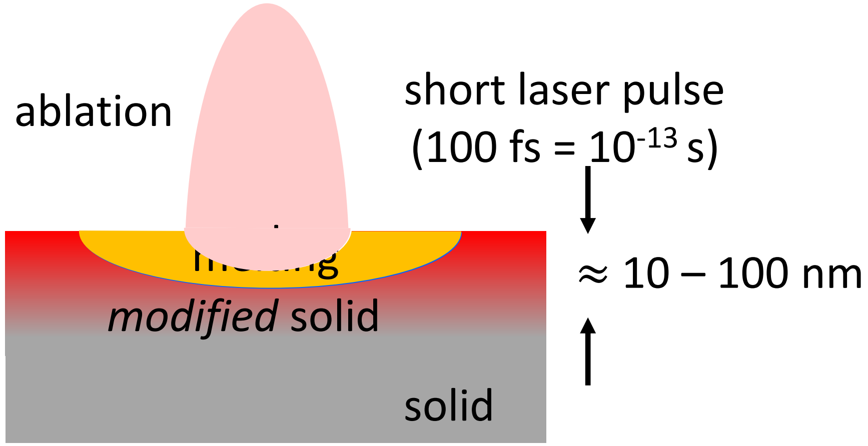
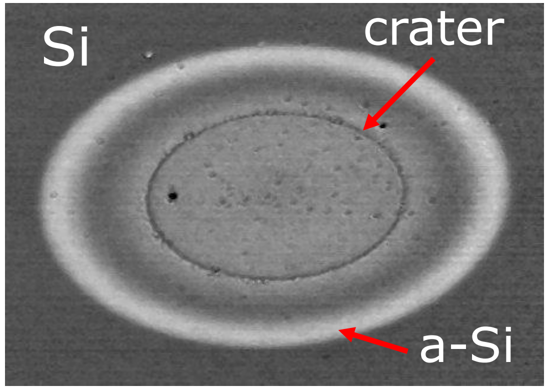
fs laser-induced ablation (of Au)

Fundamental aspects of laser ablation

laser ablation:

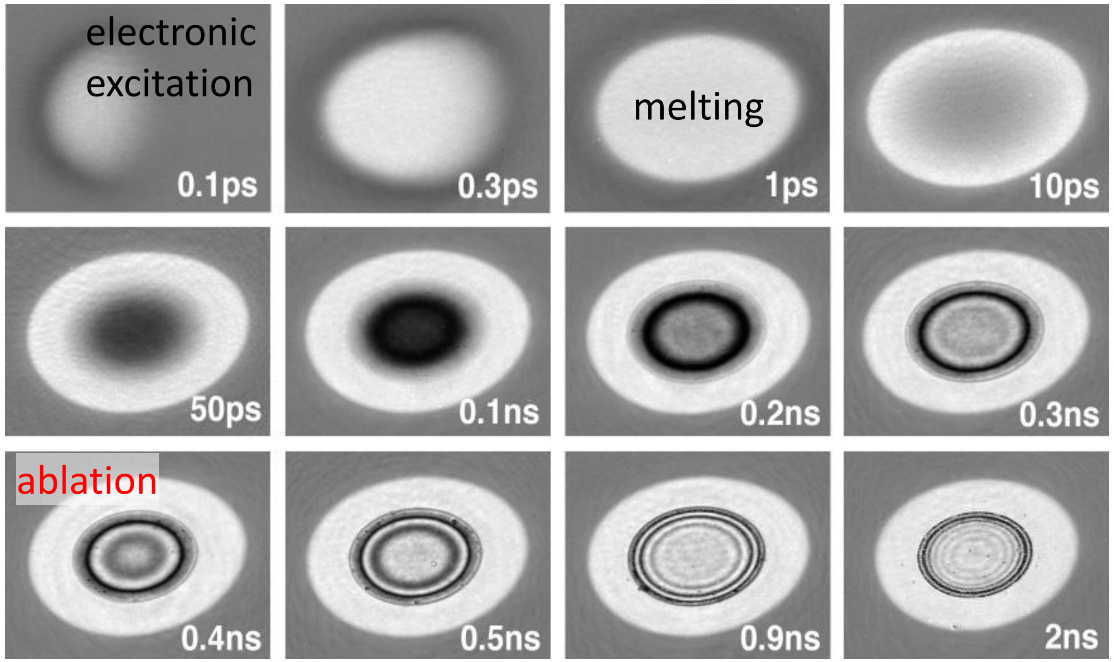
- removal of macroscopic amounts of material
- transition from condensed phase into volatile state.





t.r. imaging: fs-excited Si

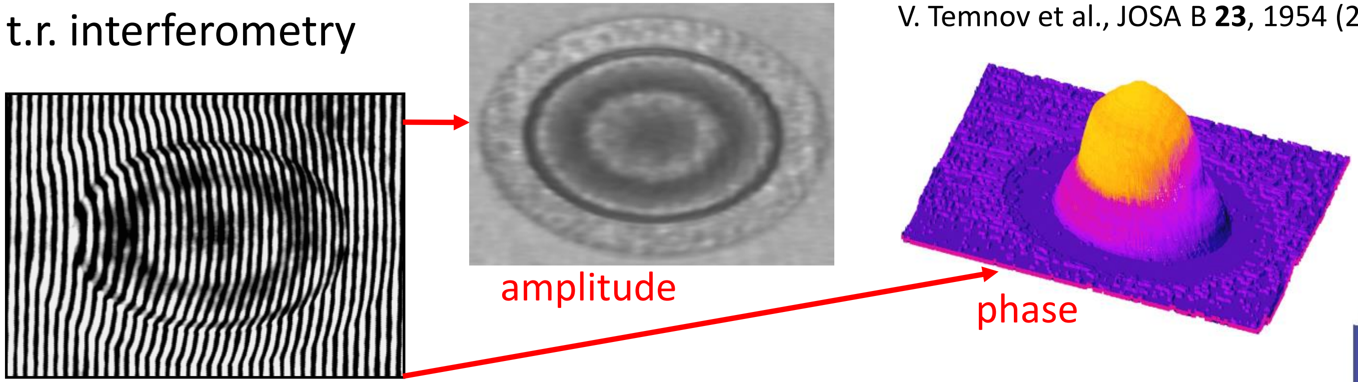
KST et al.,PRL **81**, 224 (1998)



transient Newton rings ⟹ *sharp* ablation front

t.r. interferometry

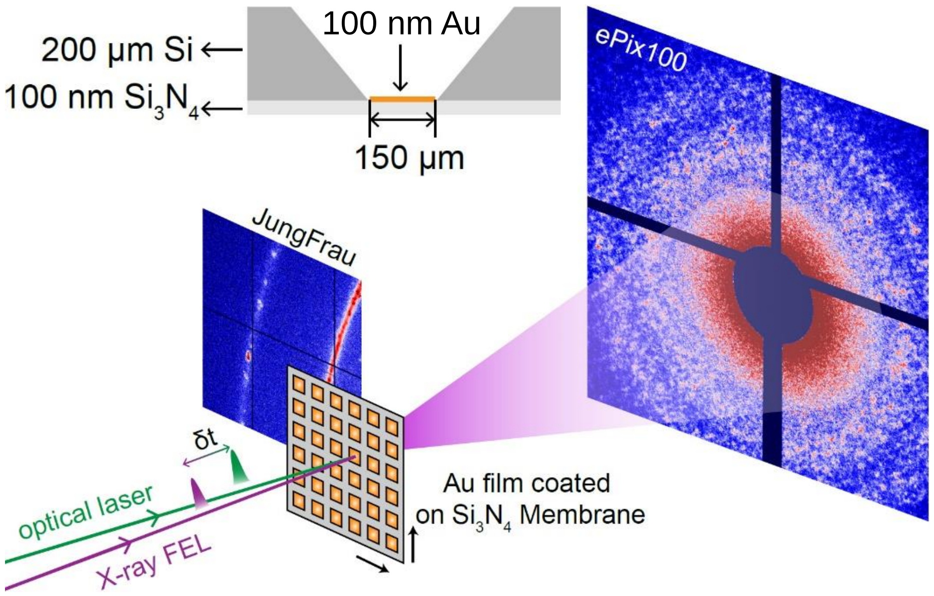
V. Temnov et al., JOSA B **23**, 1954 (2006)

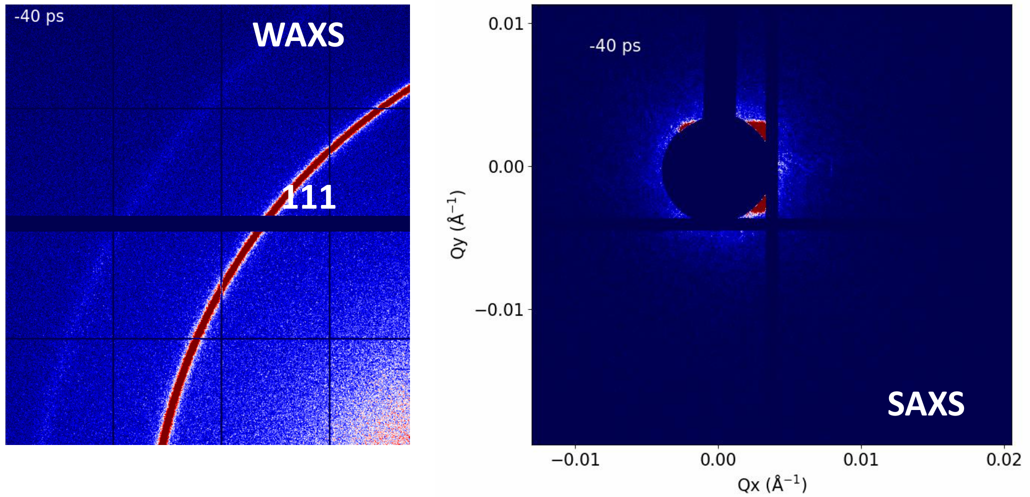


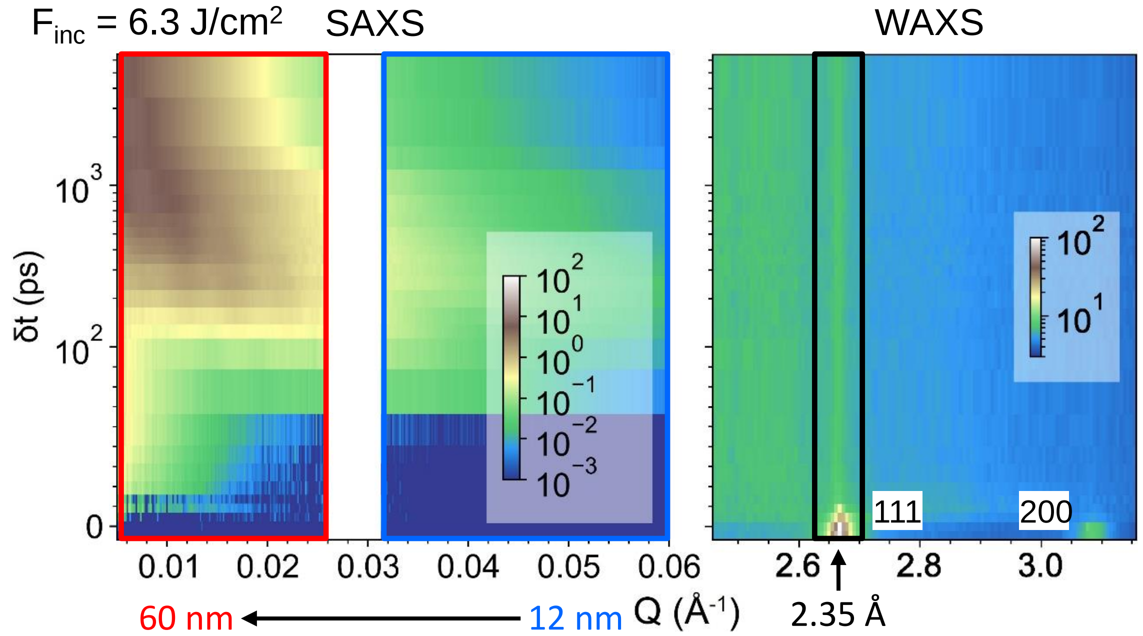
GaAs (1.8 ns)

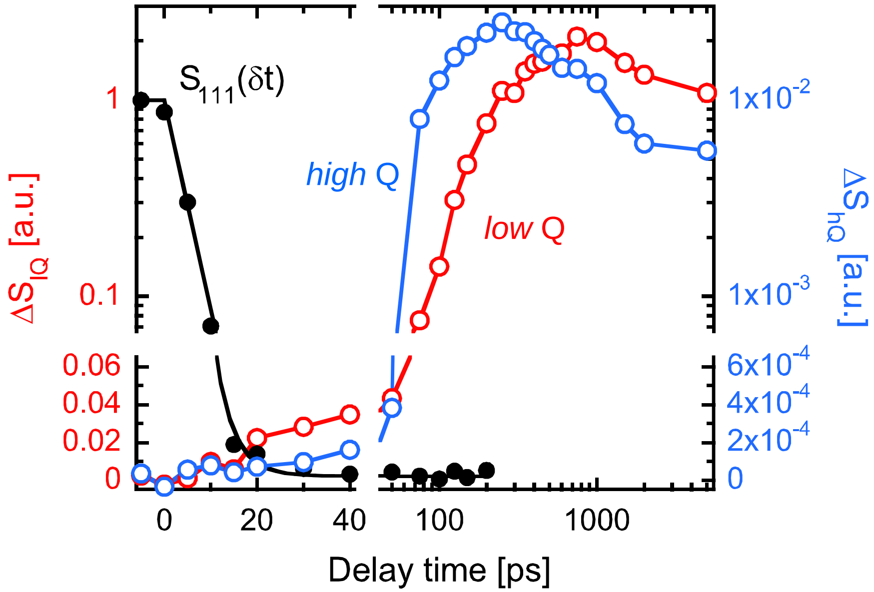
⟹ *transparent* ablation plume

Time-resolved SAXS & WAXS (XPP@LCLS)



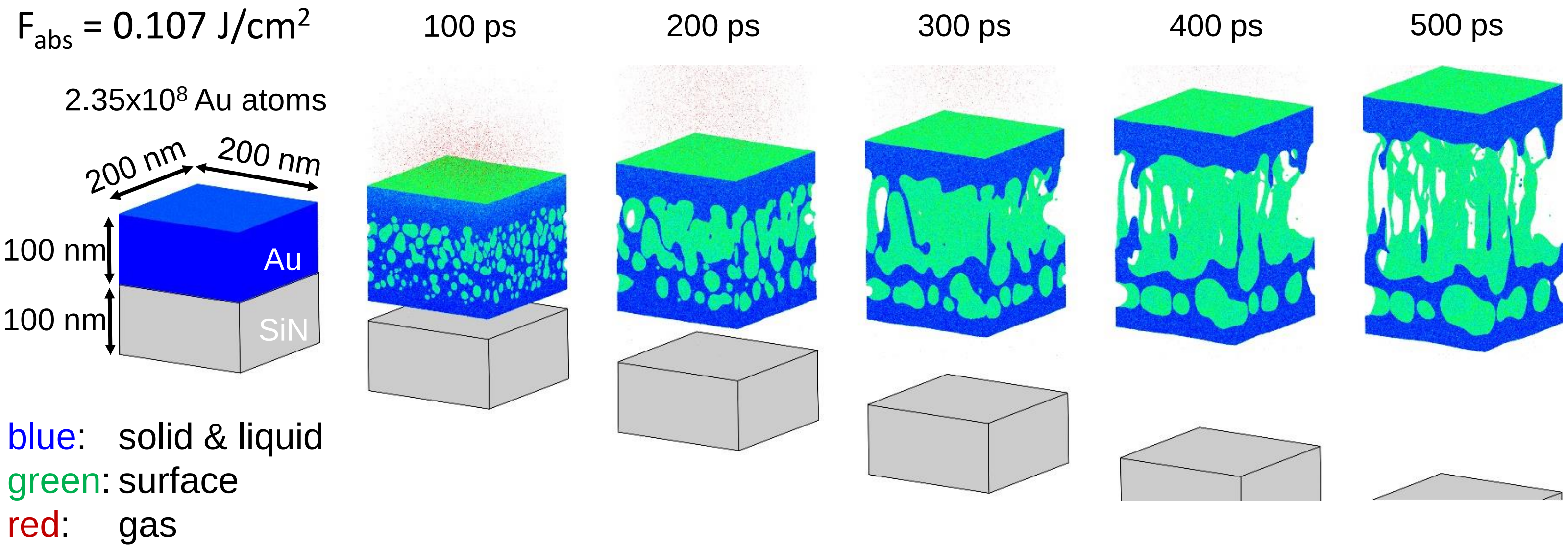


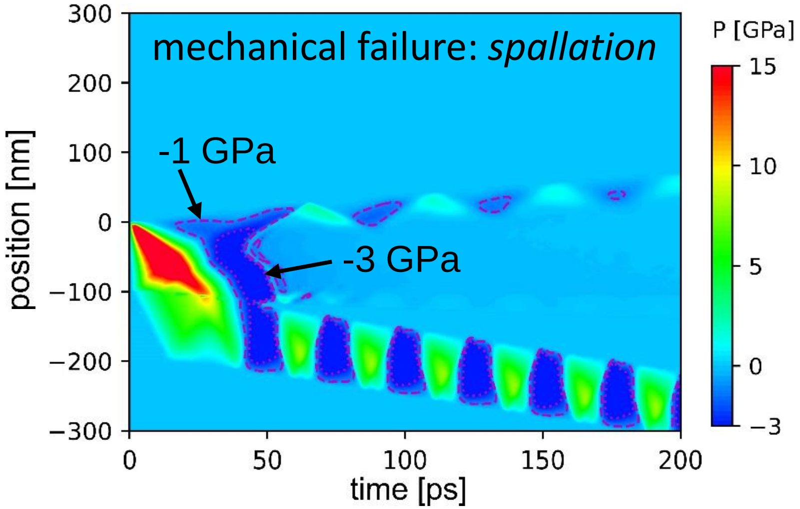


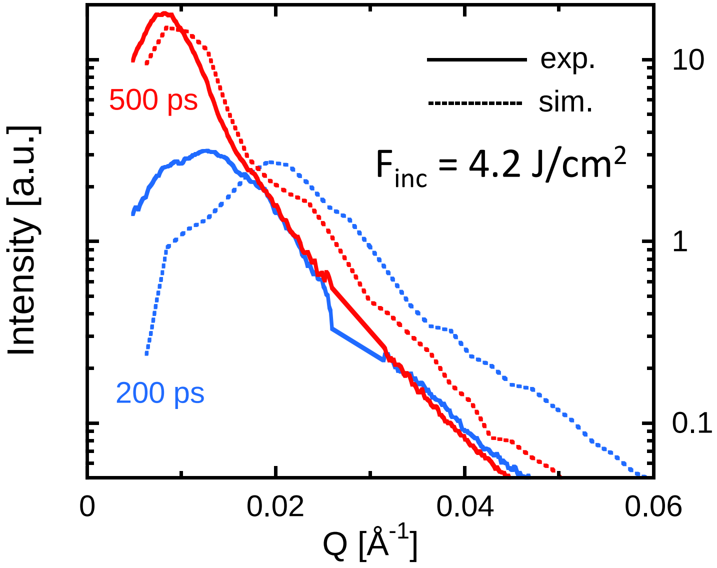


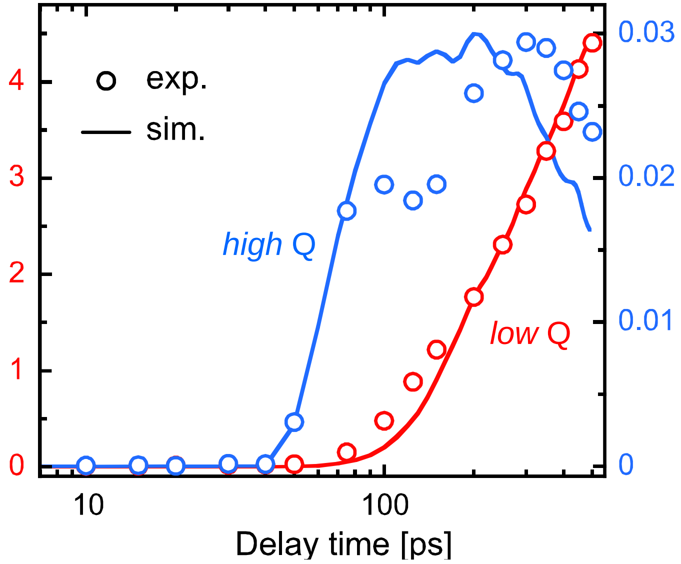
Large scale molecular dynamics

(L. Zhigilei et al.)





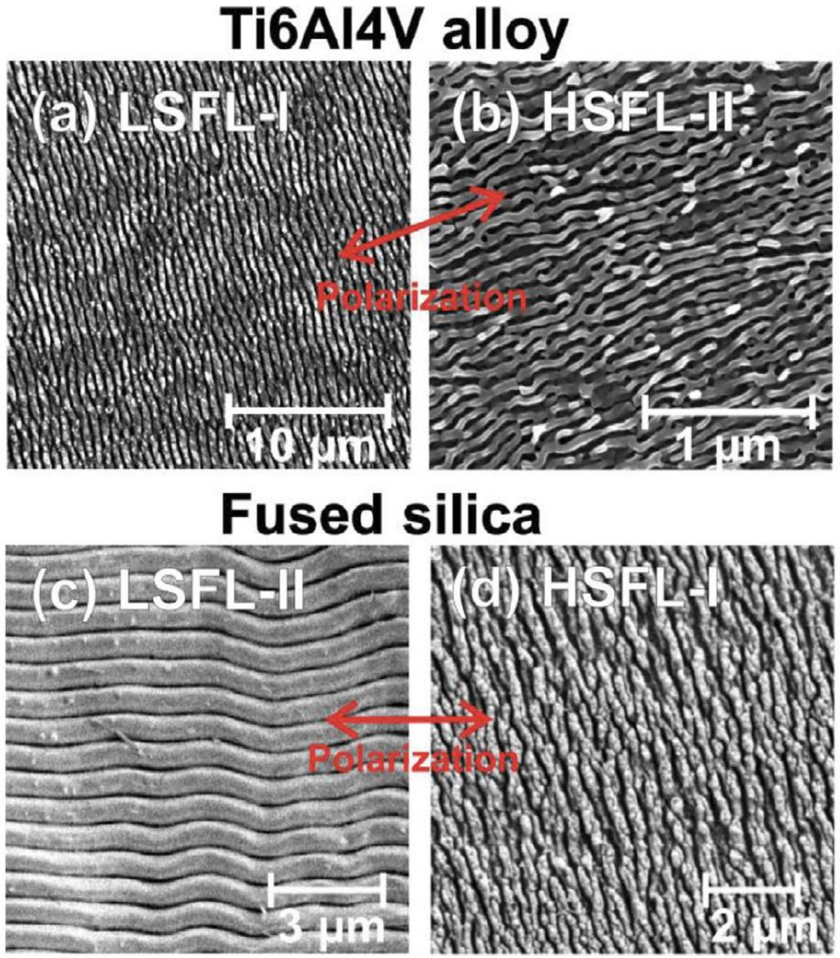




Y. Sun et al., (submitted, 2024)
arXiv:2407.10505v1

Laser-**I**nduced **P**eriodic **S**urface **S**tructures (LIPSS)

LIPSS: periodic modifications of laser-irradiated surfaces



J. Bonse & S. Gräf, Laser & Phot. Rev. 14 2000215, (2020)

LSFL- *Low spatial frequency LIPSS*

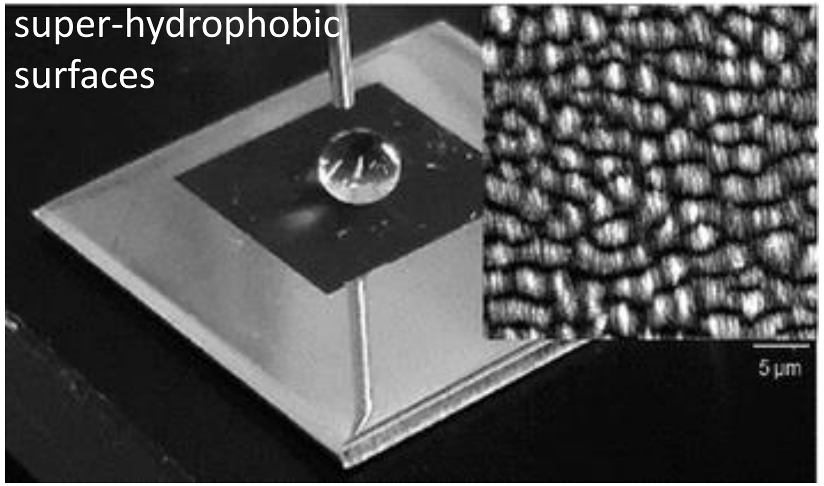
$\Lambda \approx \lambda$

HSFL- *High spatial frequency LIPSS*

$\Lambda \ll \lambda$

depend on …

- material
- polarization
- pulse duration
- fluence
- number of pulses
- environment
- …



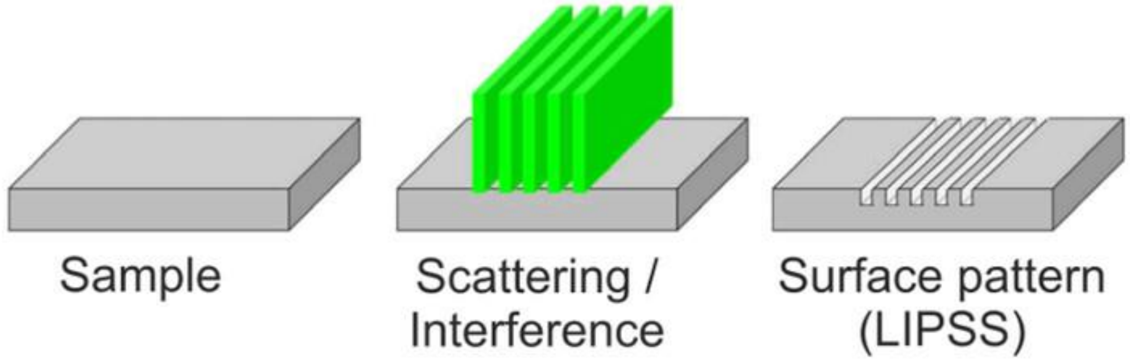
J. Reif, *Surface Functionalization by Laser-Induced Structuring,* in: Springer Series in Materials Science 274, 63 (2018)

night vision

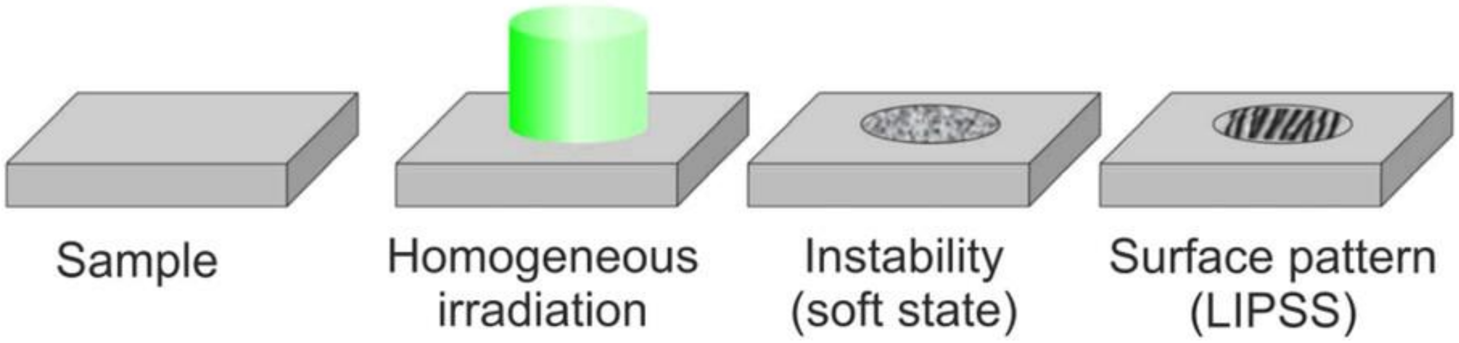
10MP sensor (sionyx.com)

LIPSS: mechanisms

electromagnetic interactions



matter reorganization



both processes contribute

see: A. Rudenko & J.-P. Colombier in "Ultrafast Laser Nanostructuring - The Pursuit of Extreme Scales", Springer Series in Opt. Sci. **209**, 257 (2023)

adapted from: O. Varlamova & J. Reif, J. of Laser Micro/Nanoengeering 8, 300 (2013)

essentially no time-resolved information on the relevant (nm/sub-µm) length scales

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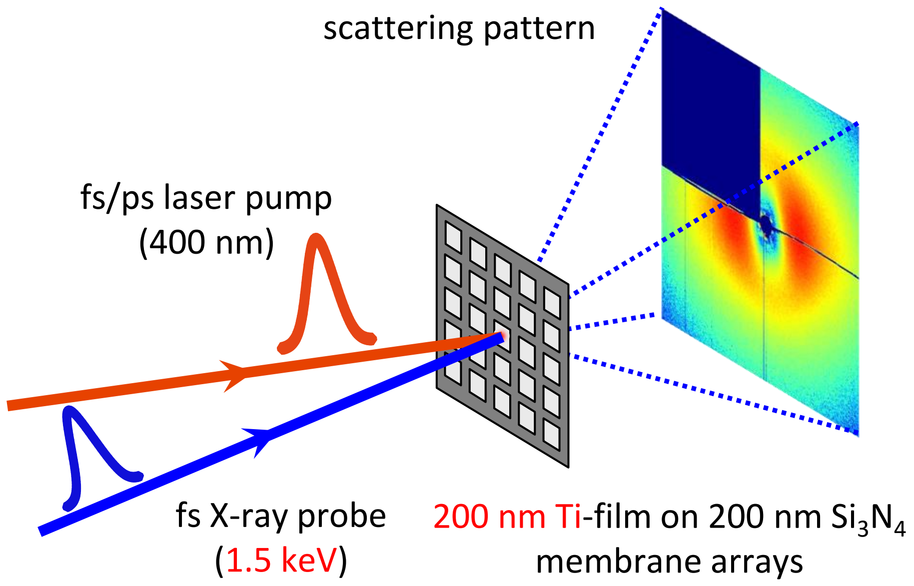
LIPSS on Ti: t.r. small-angle X-ray scattering (SCS@EuXFEL)



Unpublished

Thank you!

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