Laser induced structural dynamics in colloidal gold nanoparticles

C. Pan[1], T. J. Albert[1], A. R. Ziefuss[2], L. Kremeyer[1],
A. Plech[3], S. Barcikowski[2] and K. Sokolowski-Tinten[1]

[1]Faculty of Physics and Center for Nanointegration Duisburg-Essen, University of Duisburg-Essen, Lotharstrasse 1, 47057 Duisburg, Germany.
[2]Faculty of Chemistry and Center for Nanointegration Duisburg-Essen, University of Duisburg-Essen, Lotharstrasse 1, 47057 Duisburg, Germany.
[3]ANKA Institute for Synchrotron Radiation, Karlsruhe Institute for Technology, Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany.

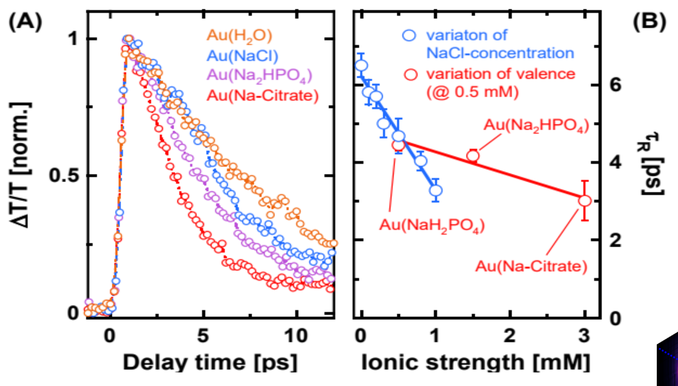
Introduction

Laser induced energy transfer and dissipation of nanoparticles in a liquid environment are of specific interest due to their relevance in a wide variety of applications. While numerous studies have addressed this issue, the results are largely based on optical pump-probe experiments, which only provide information related to electron dynamics.

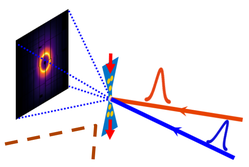
Here, we use time-resolved X-ray scattering at the ALVRA PRIME endstation of SwissFEL to characterize laser induced structural dynamics in colloidal gold nanoparticles. Based on the analysis of diffraction intensity and peak shift, we can derive information of the temperature and strain evolution of the nanoparticles after laser excitation. In addition, by adding different inorganic anions and varying their concentration, we address the influence of the chemical environment at the solid-liquid interface. Our measurements provide direct and quantitative information on the structural response of colloidal nanoparticles and are useful to understand the fundamental mechanisms of energy transfer and dissipation in this system.

Motivation and principle

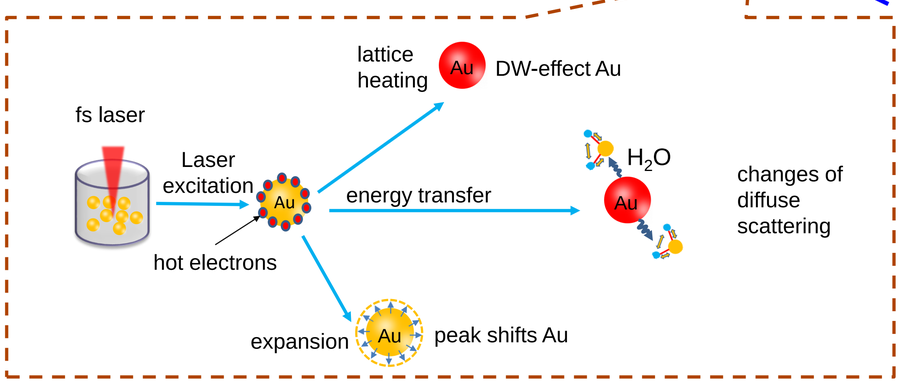
Motivation: all-optical pump-probe-results



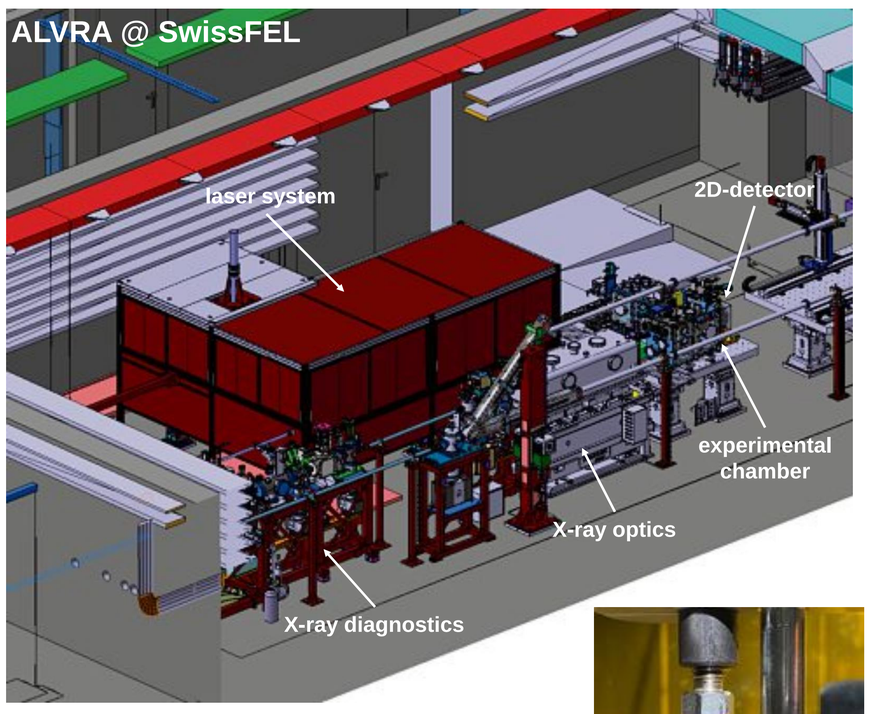
A. Ziefuss et al., (2021, under review)



Principle: ultrafast X-ray diffraction

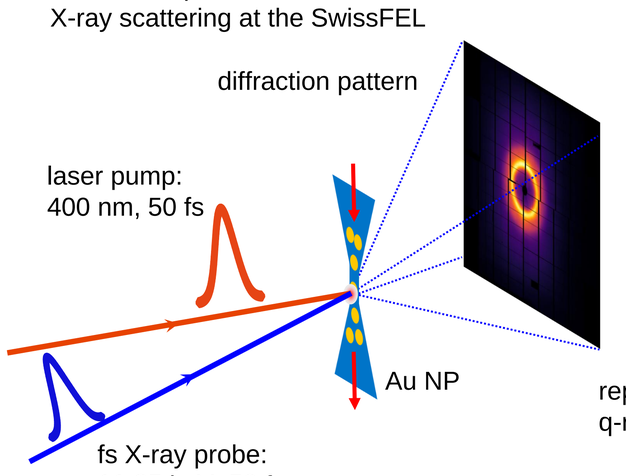


tr-X-ray diffraction at the SwissFEL



ALVRA: Experimental station for X-ray scattering at the SwissFEL

liquid jet, 3.5 m / s



repetition rate: 100 Hz
q-range: 0.4 Å^{-1} < q < 6.7 Å^{-1}

Samples: Colloidal Au NP
- laser ablation synthesis
- surfactant-free
- variation og the chemical environment

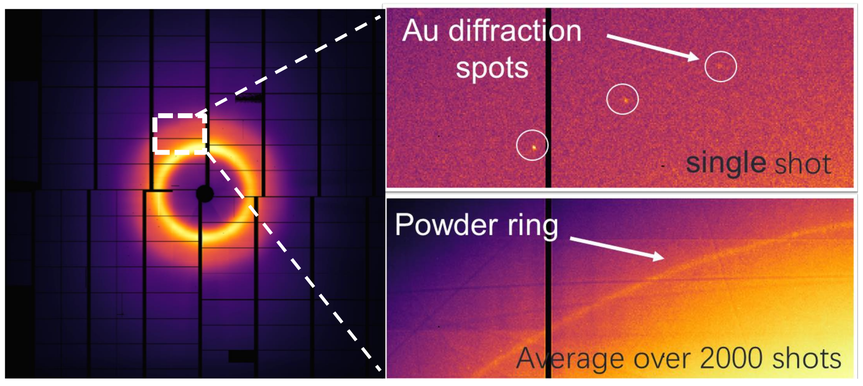
Chem. Rev., 2017, 117(5): 3990-4103.

Increasing polarizability

monovalent salts	
NaOH	1 mM
NaCl	0.1 - 1mM
NaBr	0.1 – 1mM
multivalent salts	
Na_2HPO_4	1 mM
Na-Citrate	1 mM

Data processing and analysis

Data structure and features: strong water background v.s. weak gold signal



each dataset (2000 measurements)

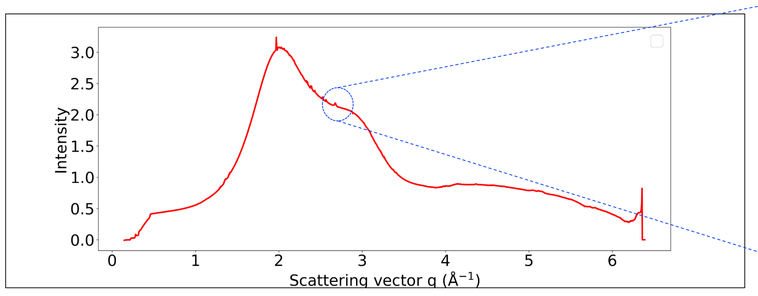
corresponding

- one concentration
- one laser energy
- one delay time
- nanoparticle size
- salt type

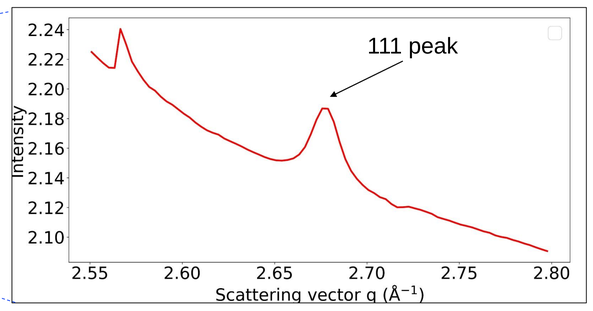
varying parameters

totally over 15000 datasets — **> 1 Petabyte**

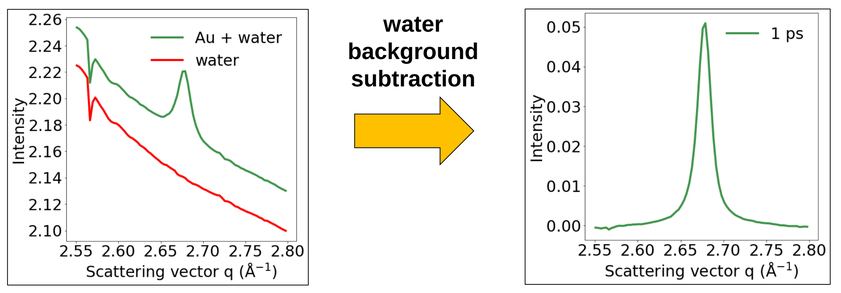
- azimuthal integration
- average
- normalization



- weak gold signal with a strong water background



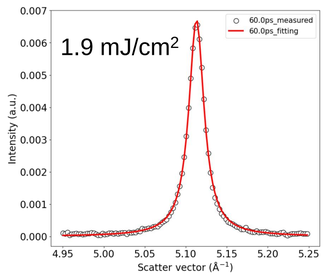
Water background subtraction and peak profile fitting



- intensity decrease
- peak shift

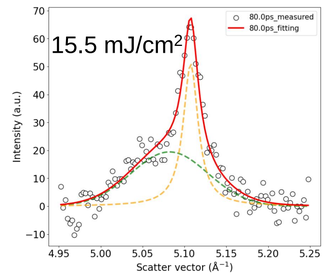
Low laser fluence (non-melting case)

- fitting function: Lorentzian
- symmetric



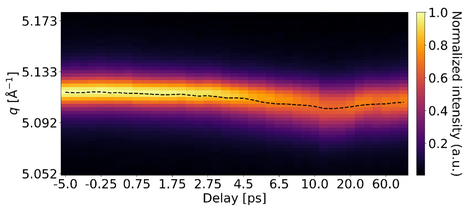
High laser fluence (melting case)

- fitting function: Lorentzian + Gaussian
- asymmetric



Results and discussion

Low laser fluence (non-melting case)

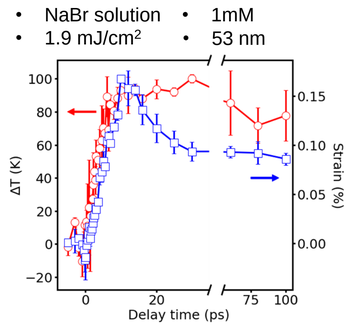


- separation for DW effect and expansion

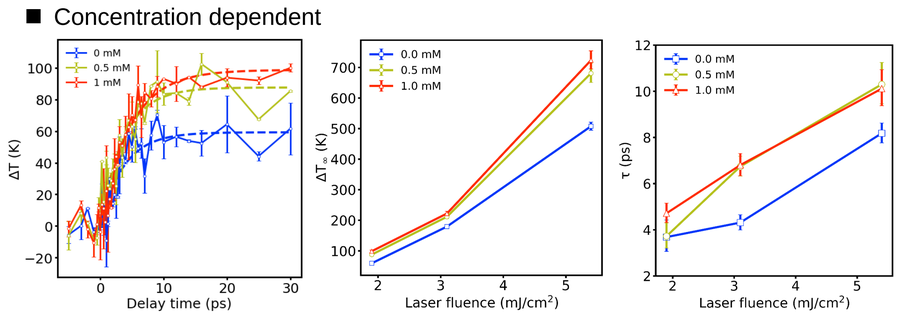
Debye-Waller(DW) factor:

$$-ln\left(\frac{I(\Delta t)}{I_0}\right) = \frac{1}{3}\Delta\langle u^2\rangle(\Delta t)\cdot G^2$$

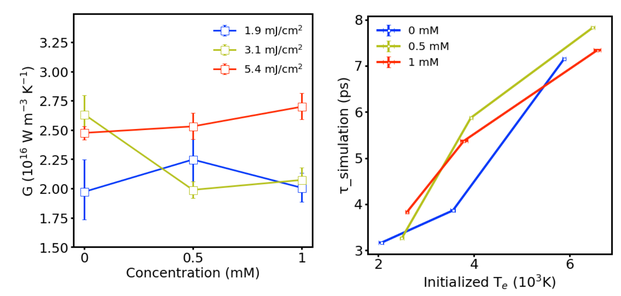
- NaBr solution
- 1.9 mJ/cm²
- 1mM
- 53 nm



- Concentration dependent

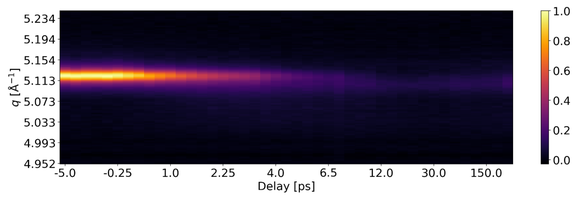


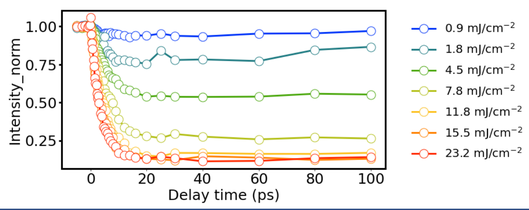
- TTM simulation



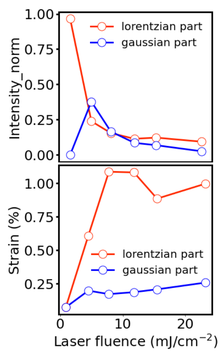
- No obvious concentration effect on electron-phonon coupling strength
- Stronger energy in-coupling in salt solution than pure water

High laser fluence (melting case)

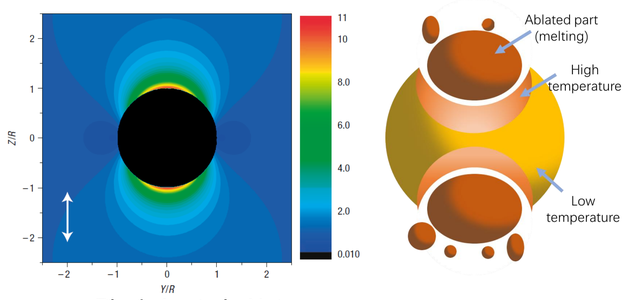




- delay: 300ps



- inhomogenous melting/ablation



Plech A, et al. Nature Physics, 2006, 2(1): 44-47.

Acknowledgements

Diffraction experiments were carried out at the SwissFEL, Paul Scherrer Institute (Villigen, Switzerland).
This project received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 801459 and the Deutsche Forschungsgemeinschaft (DFG) under Germany´s Excellence Strategy – EXC 2033 – 390677874 –RESOLV *and the Deutsche Forschungsgemeinschaft DFG in the frame of the collaborative research center SFB1242*

Contact: changji.pan@uni-due.de