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Open-Minded

Time-Resolved X-Ray Diffraction from Coherent Acoustic Phonons Analyzed by Combining Microscopic Modeling with Deep Learning Based Strain Retrieval

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Introduction

- Absorption of ultrashort optical pulses in solids
  - Quasi-instantaneous increase of stress/pressure
  - Acoustic strain waves (*coherent acoustic phonons*)
- Aim of this project: *Quantitative* investigation of the excitation and transient evolution of acoustic waves in metal films (Ti, Al, Pt, Pd, Au) deposited on 100-oriented GaAs
- Experimental tool: Time resolved X-ray diffraction using ultrashort X-ray pulses at 4.5 keV from a lab-based fs laser-plasma X-ray source [1]
- Measurement of the transient evolution of the GaAs (400) Bragg reflection in an optical pump – X-ray probe experiment
- New method for direct strain retrieval using deep neural networks (DNN) to solve phase problem

Experiment

- Pump light is absorbed in the first few nanometers of the metal film (thickness ~100 nm)
  - No direct pumping of the GaAs substrate
- Quasi-instantaneous increase of stress in the metal
- Electronic and thermal contributions

$$\sigma = \sigma_e(t,z) + \sigma_{th}(t,z)$$

$$\delta\sigma = -\gamma_e C_e \delta T_e - \gamma_{th} C_{th} \delta \mathrm{T_{th}} \text{ [1]}$$

- Spatial shape and temporal dependence of the generated stress strongly depend on the properties of the absorbing metal
- Subsequent release of strain waves from the surface and the interface following the equation of elasticity

$$\rho \frac{\partial^2 u}{\partial t^2} = \frac{\partial \sigma}{\partial z}$$

- Strain waves hitting the interface are partly transmitted into the GaAs
  - Detectable change of lattice constant
- Simulation of acoustic equations and dynamical X-ray diffraction theory (DXRD) yields inside into physical properties

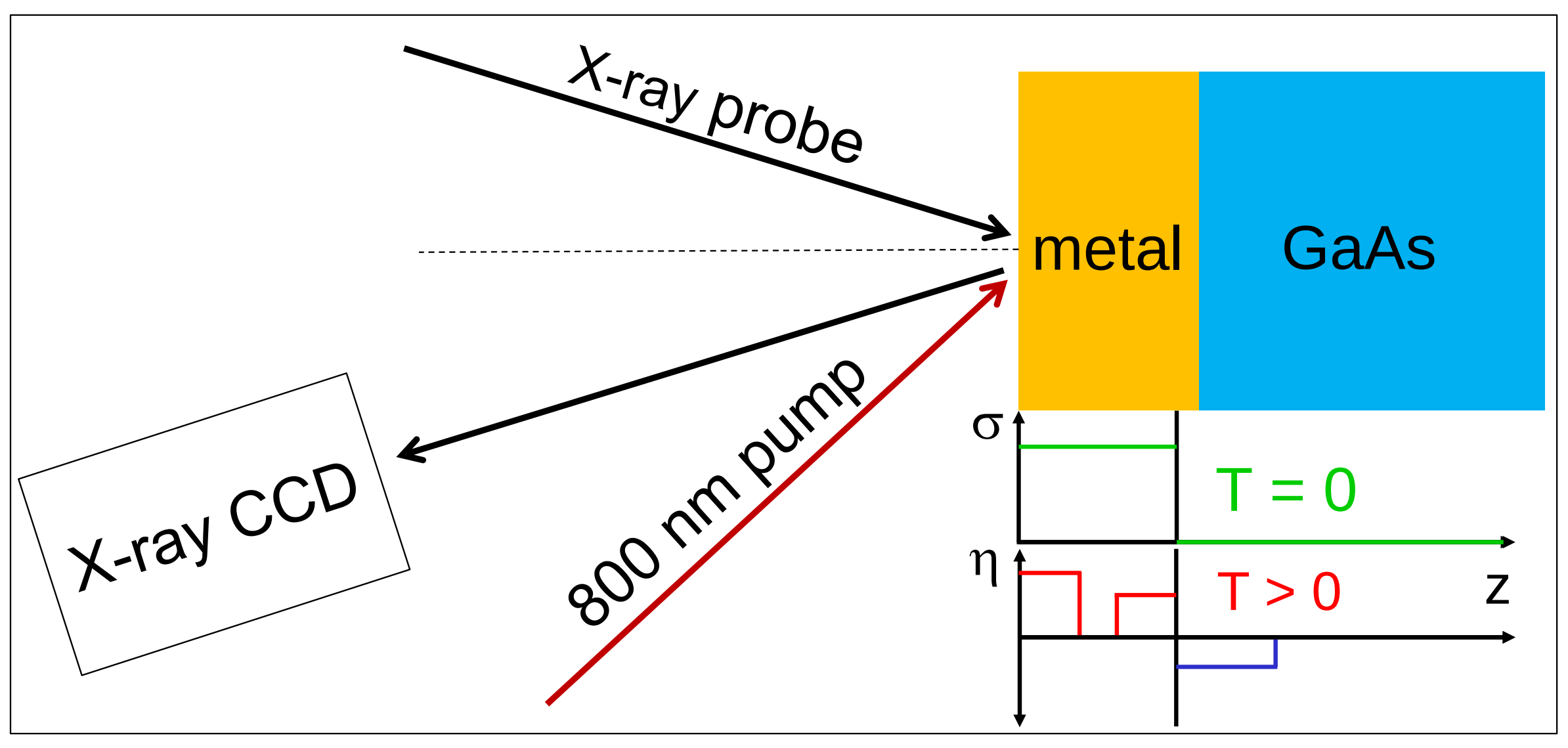


Fig 1: Measurement configuration, initial stress for T = 0 and strain for T > 0 for time- and space-independent starting conditions

X-Ray Source

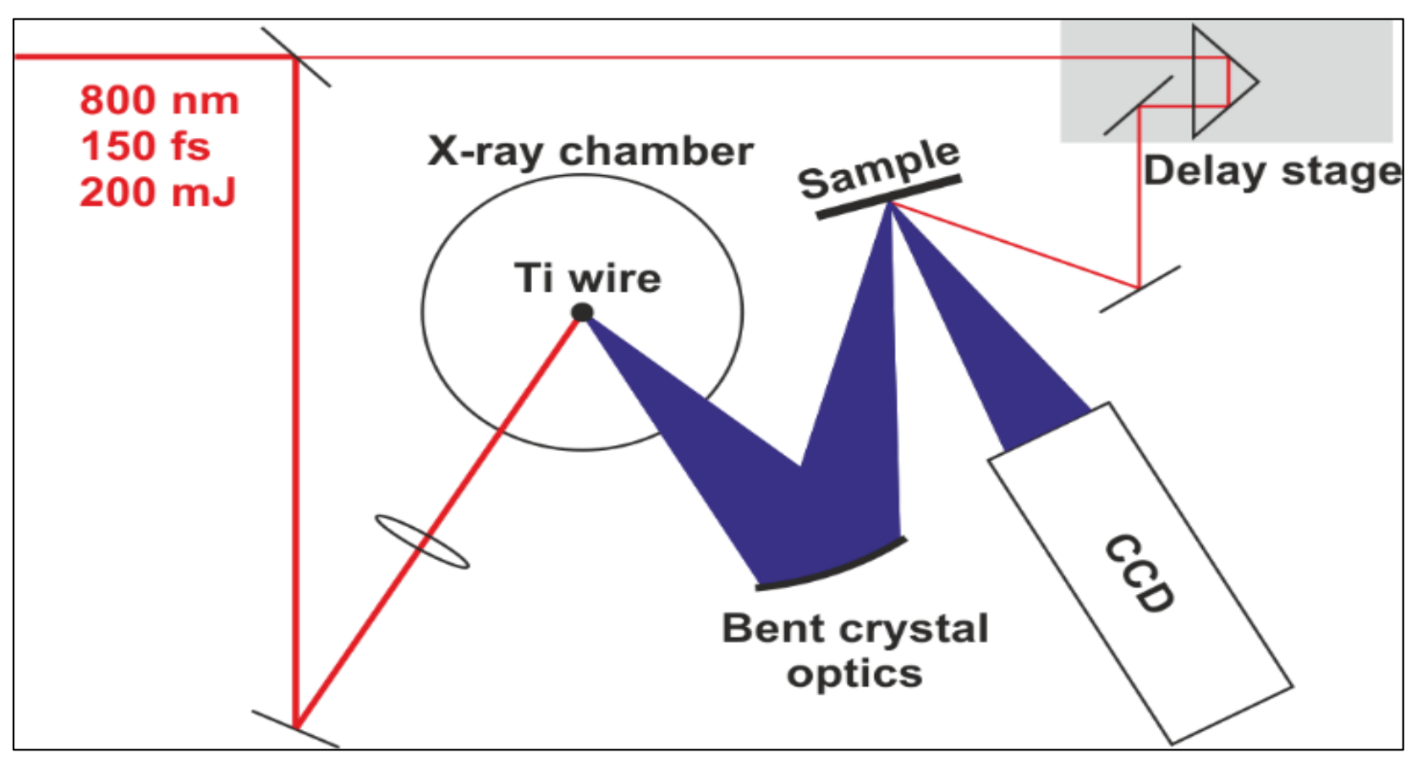


Fig 2: Setup

$$d_{FWHM}^{X-ray} = 80\ \mu m$$

$$d_{FWHM}^{laser} = 450\ \mu m$$

$$\theta_{pump-probe} = 50\,°$$

$$\theta_B^{400} = 76.629\,°$$

$$\frac{\Delta E}{E} = 10^{-4}$$

Afshari et *al.*, Struct. Dyn. **7**, 014301 (2020).

- Focusing of intense laser pulse on a Ti wire creates a plasma
- Plasma electrons are accelerated to relativistic energies and hit the Ti creating X-ray photons as in an X-ray tube
- Photons are collected and focused using a toroidally bent Ge crystal in Rowland-circle geometry
- Highly monochromatized

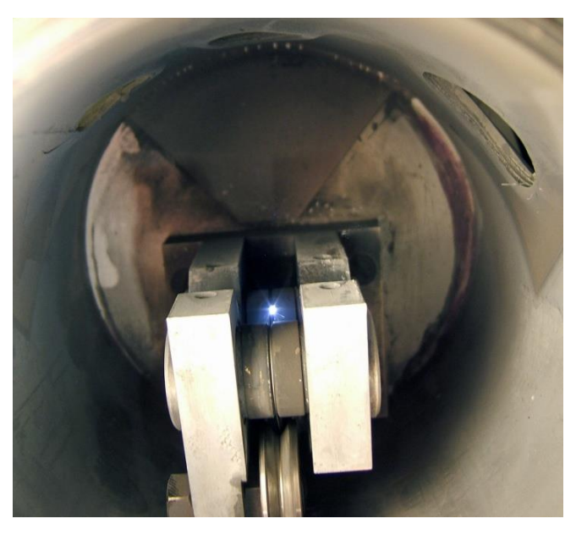


Fig 3: Wire and the laser induced plasma

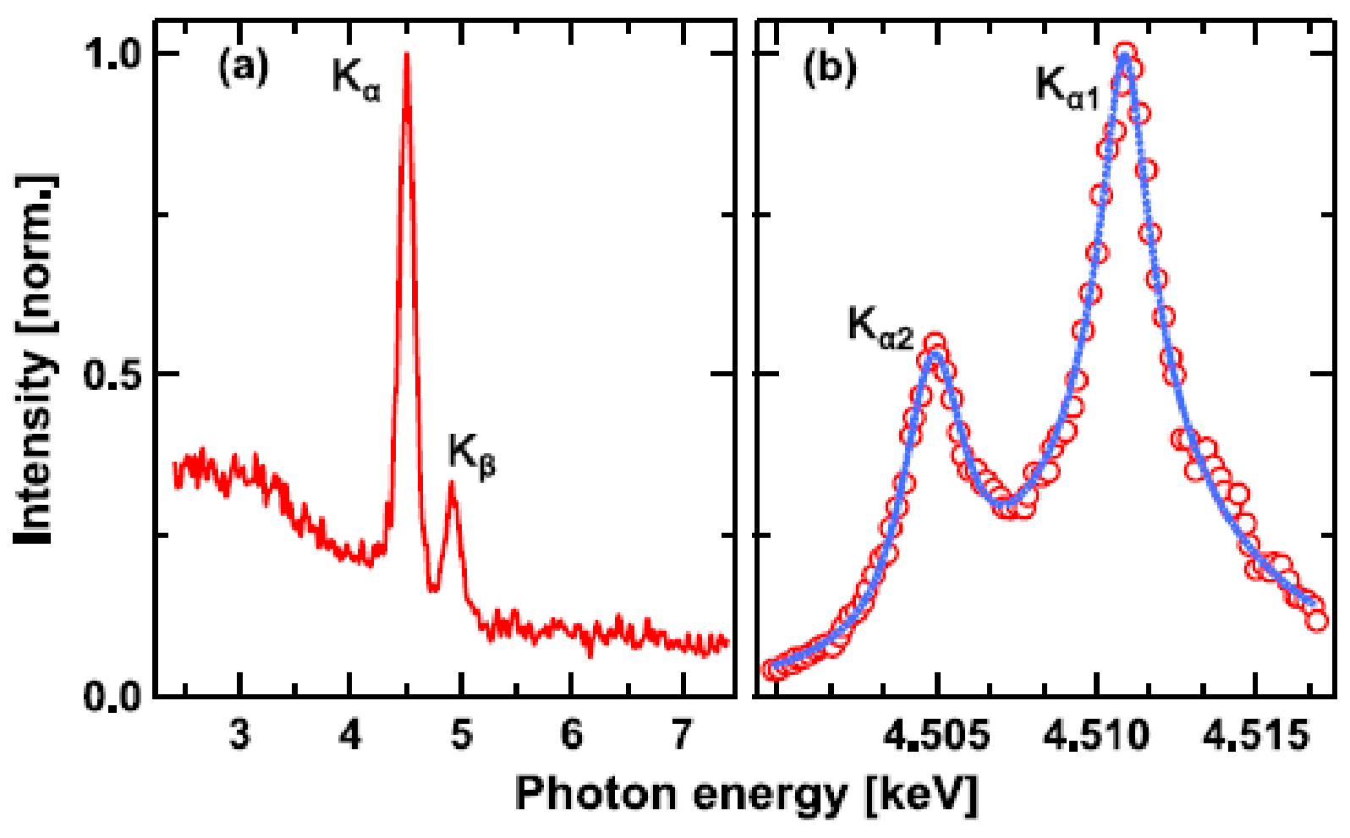


Fig 4: Spectrum of the X-ray plasma emission in low (a) and high (b) resolution

Transient Rocking Curves

- Upon excitation, strain waves travel back and forth in the metal film due to reflection at the interface and the surface
- Characteristic time scale: Travelling time of a pulse through the film

$$\mathrm{T_c} = \frac{s}{c} \approx 15.5\ ps \qquad \text{(e.g. aluminum)}$$

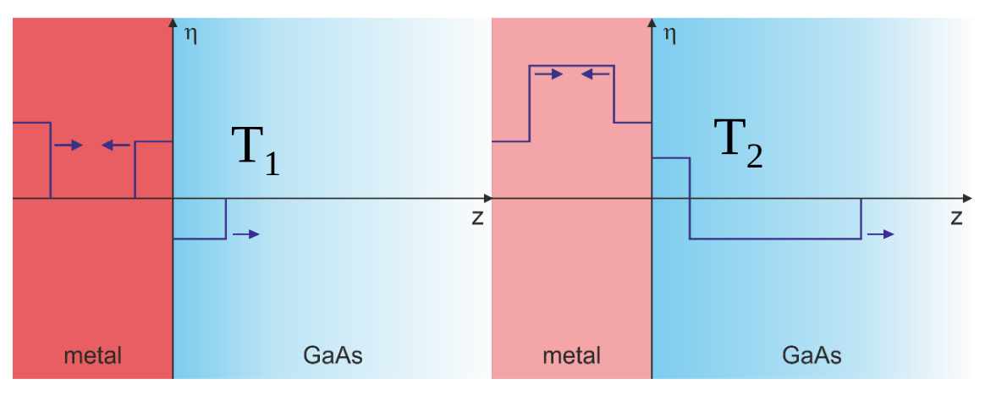


Fig 5: Strain profiles in the sample for two points in time $T_1 < T_2$

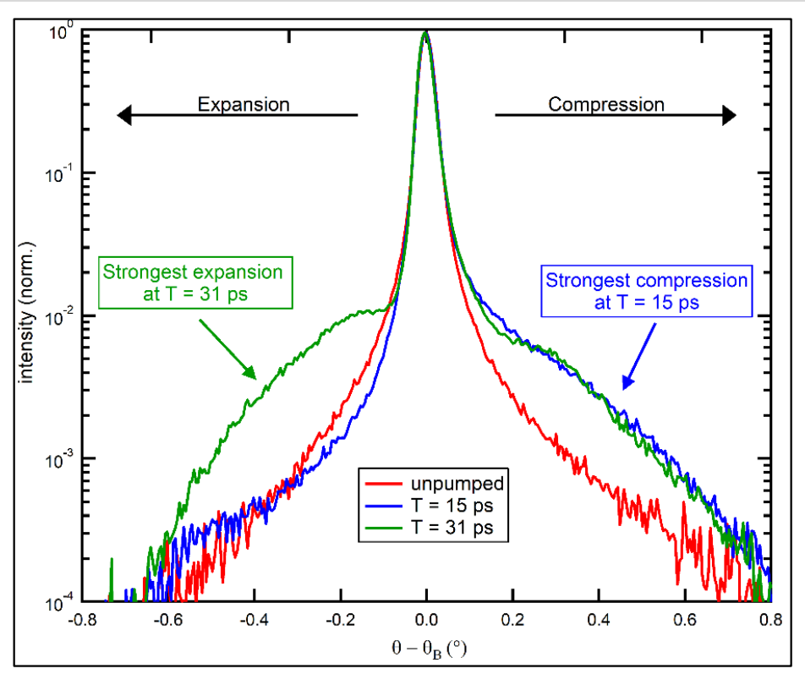


Fig 6: Comparison of the measured rocking curve

- Compression and expansion waves show up as small wings in the GaAs rocking curve (*high dynamic range required*)

Experiments and Microscopic Modeling

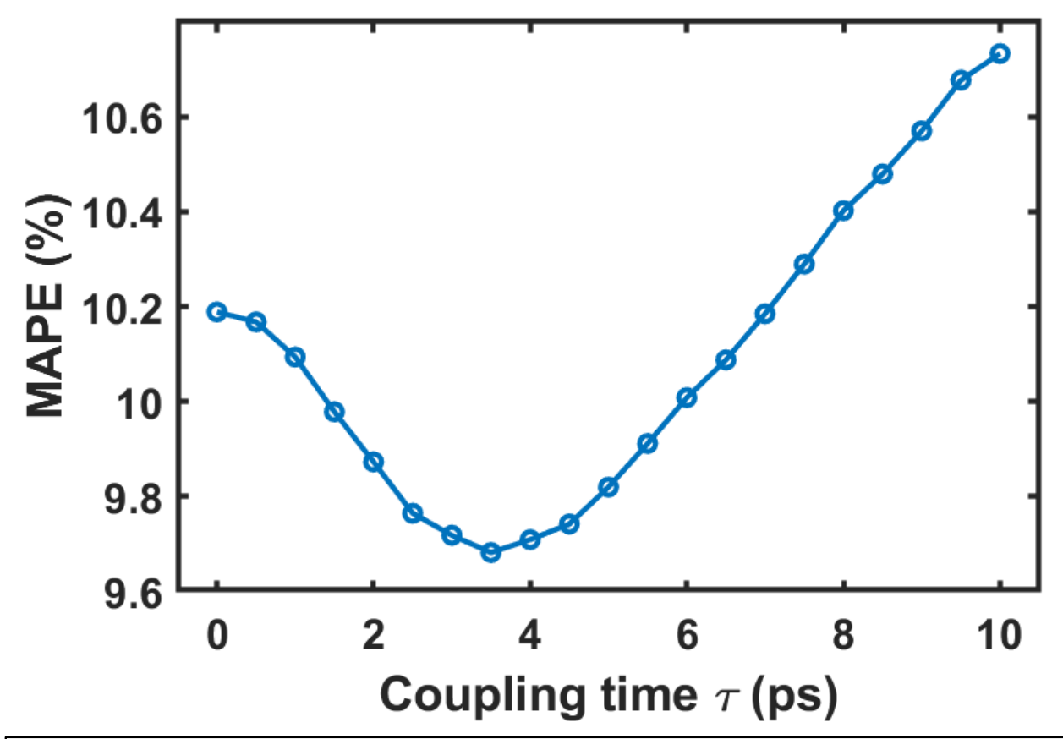
Au

Absorption of light within first few nm

Efficient electronic transport [3, 4]

Electron-phonon coupling on a „slow“ time scale

$$\sigma(t) = \sigma_\infty H(t) + \left[1 + \left(\frac{\gamma_e}{\gamma_L} - 1\right) e^{-\frac{t}{\tau}}\right] \quad [5]$$



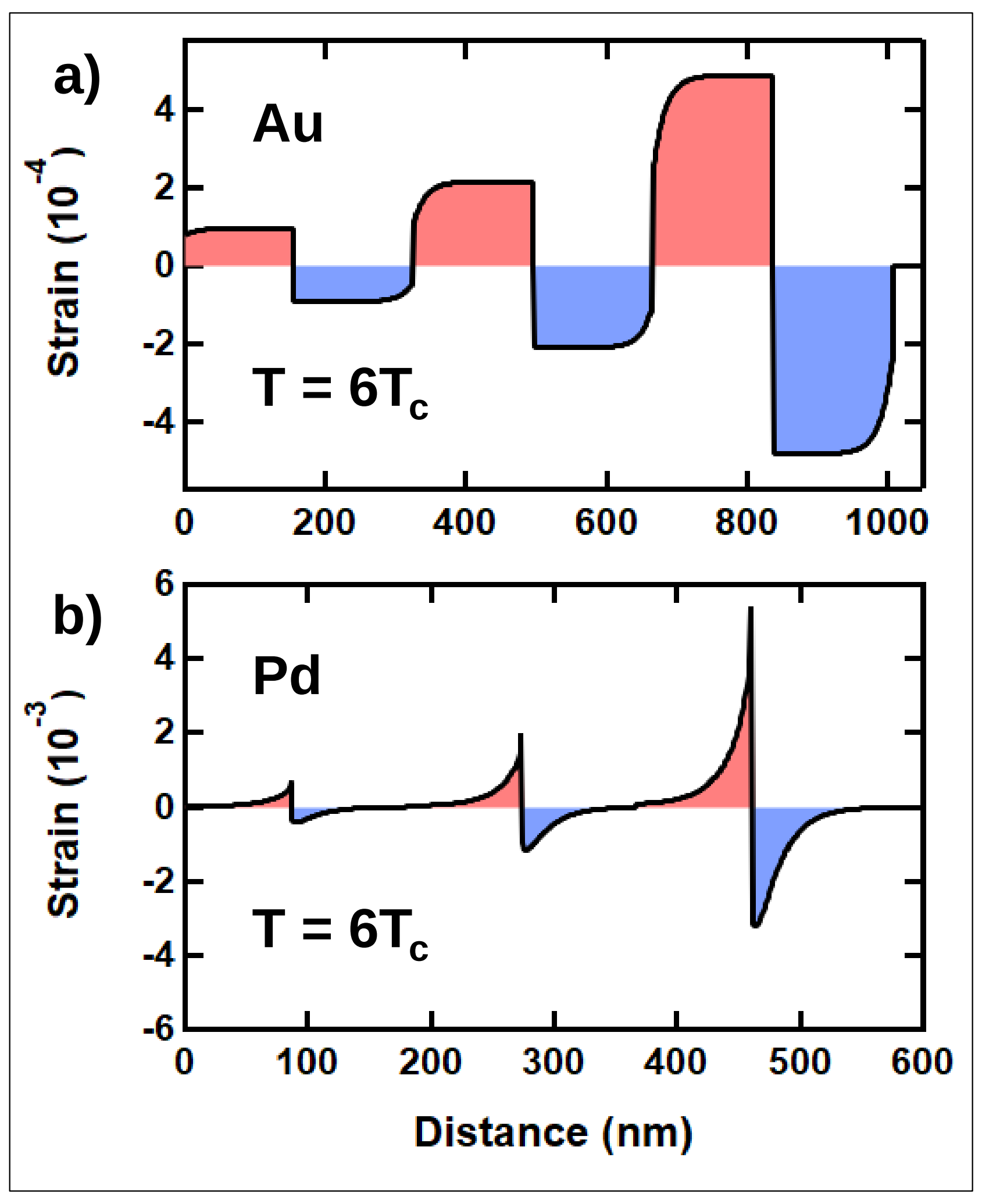


Fig 7: Strain pulses in GaAs for the Au coated (a) and Pd coated (b) sample after six characteristic times

Electron-phonon coupling, transport and their interplay determine acoustic response in these systems

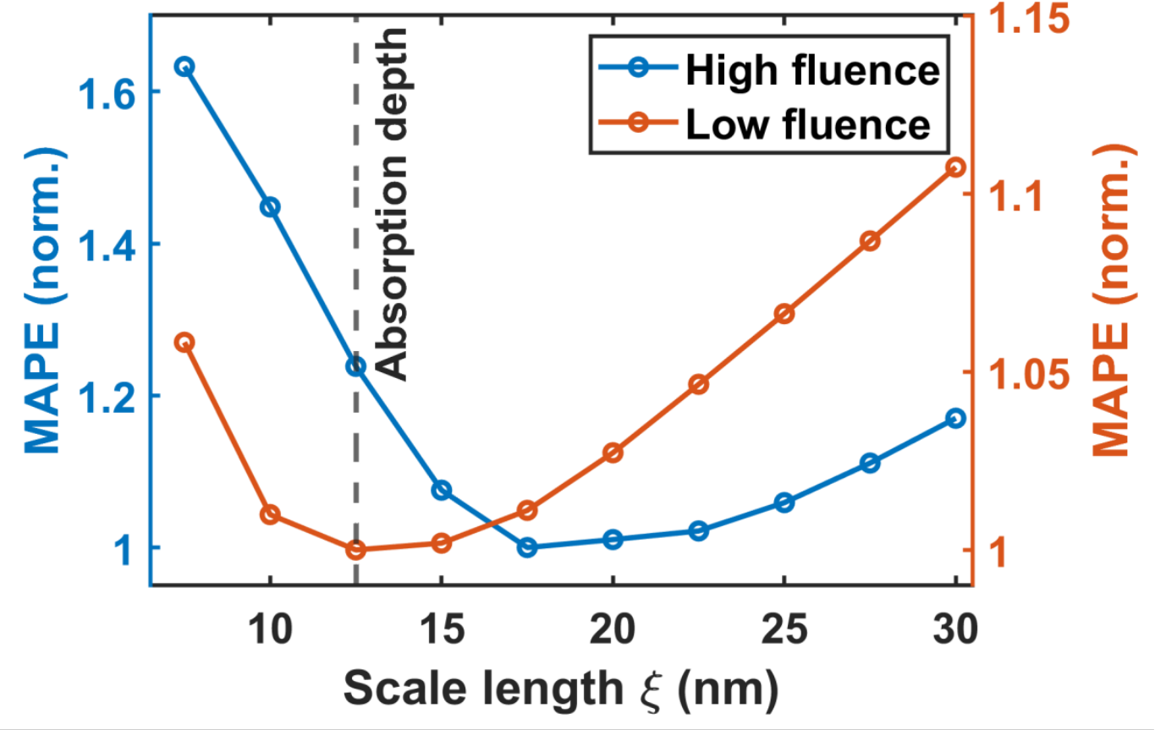
Pd

Absorption of light within first few nm

Ultrafast electron-phonon coupling + therm. diff. [5]

Simulation of the acoustic response [6]

$$\sigma(z) = \sigma_\infty \exp\left(-\frac{z}{\xi}\right)$$



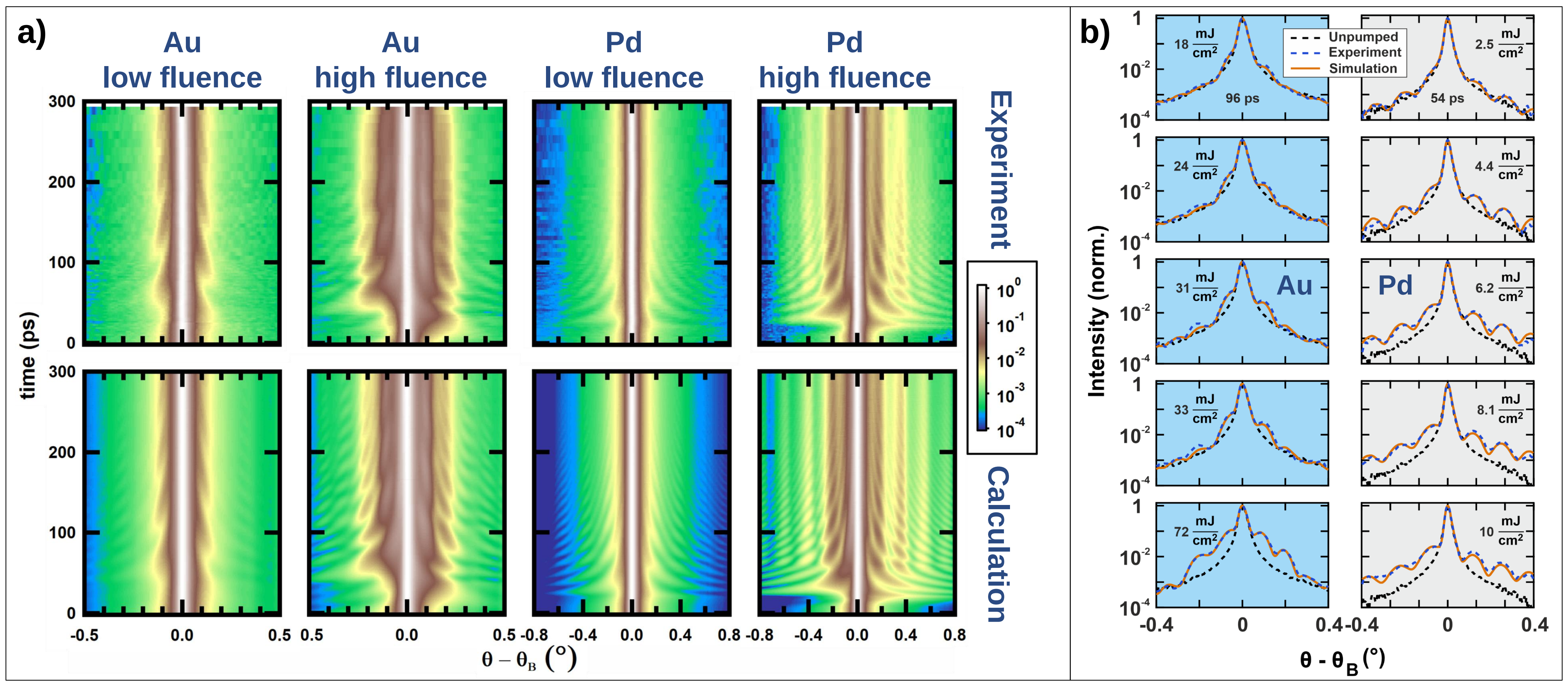


Fig 8: Comparison of experiment and modeling for the Au and Pd coated samples. a) depicts the transient rocking curves for two different fluences for both samples, where the upper panel .shows the measurement and the lower one the simulation. b) shows the comparison of experiment and simulation for different fluences

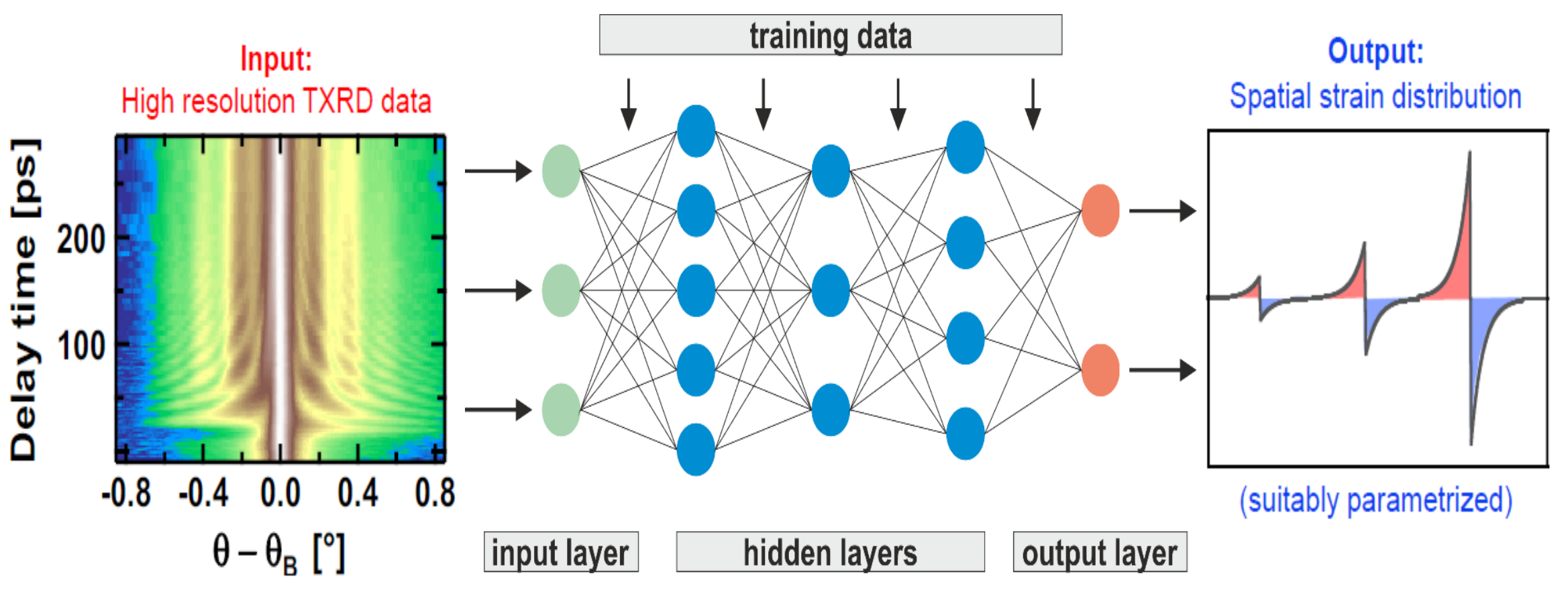
Direct Strain Retrieval Using Deep Neural Networks

- Phase problem in diffraction experiments

$$X(\vec{r}) = \hat{X}(\vec{r})\exp(i\phi(\vec{r})) \xrightarrow{\text{Measurement}} I(\vec{r}) = |X(\vec{r})|^2$$

- Idea: Train deep neural network (DNN) for retrieving strain from transient rocking curves (image recognition)
- Architecture of choice: Convolutional neural network (CNN) [8]
- Training data: Creation by simulating DXRD for random strain pulses

Fig 9: Principle of a deep neural network



- Pulse parametrization:
  - Fourier expansion of the bipolar pulse **{S$_i$, C$_i$}**
  - Pulse thickness **d**
  - Interface reflectivity **R**

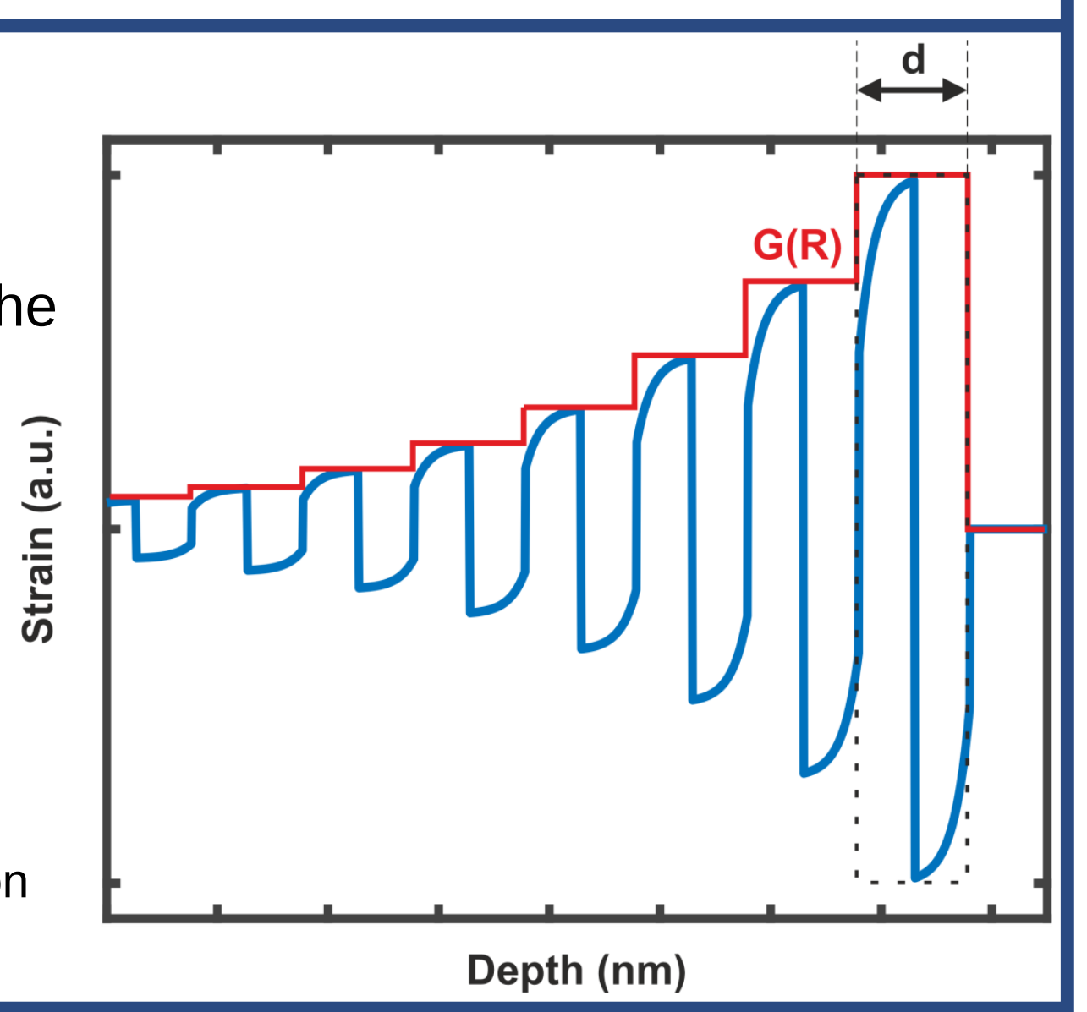


Fig 10: Pulse Parametrization

Au

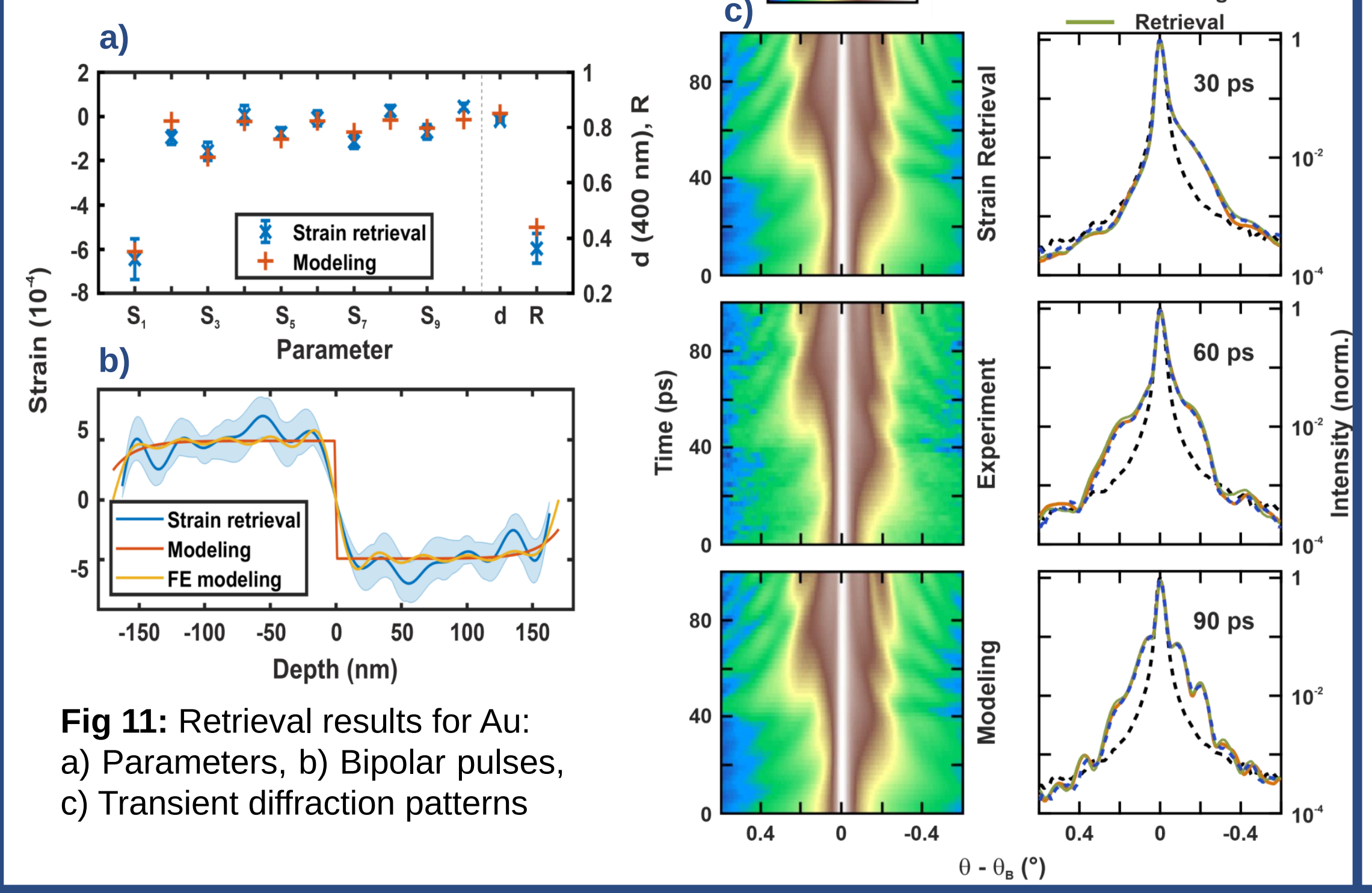


Fig 11: Retrieval results for Au:
a) Parameters, b) Bipolar pulses,
c) Transient diffraction patterns

Pd

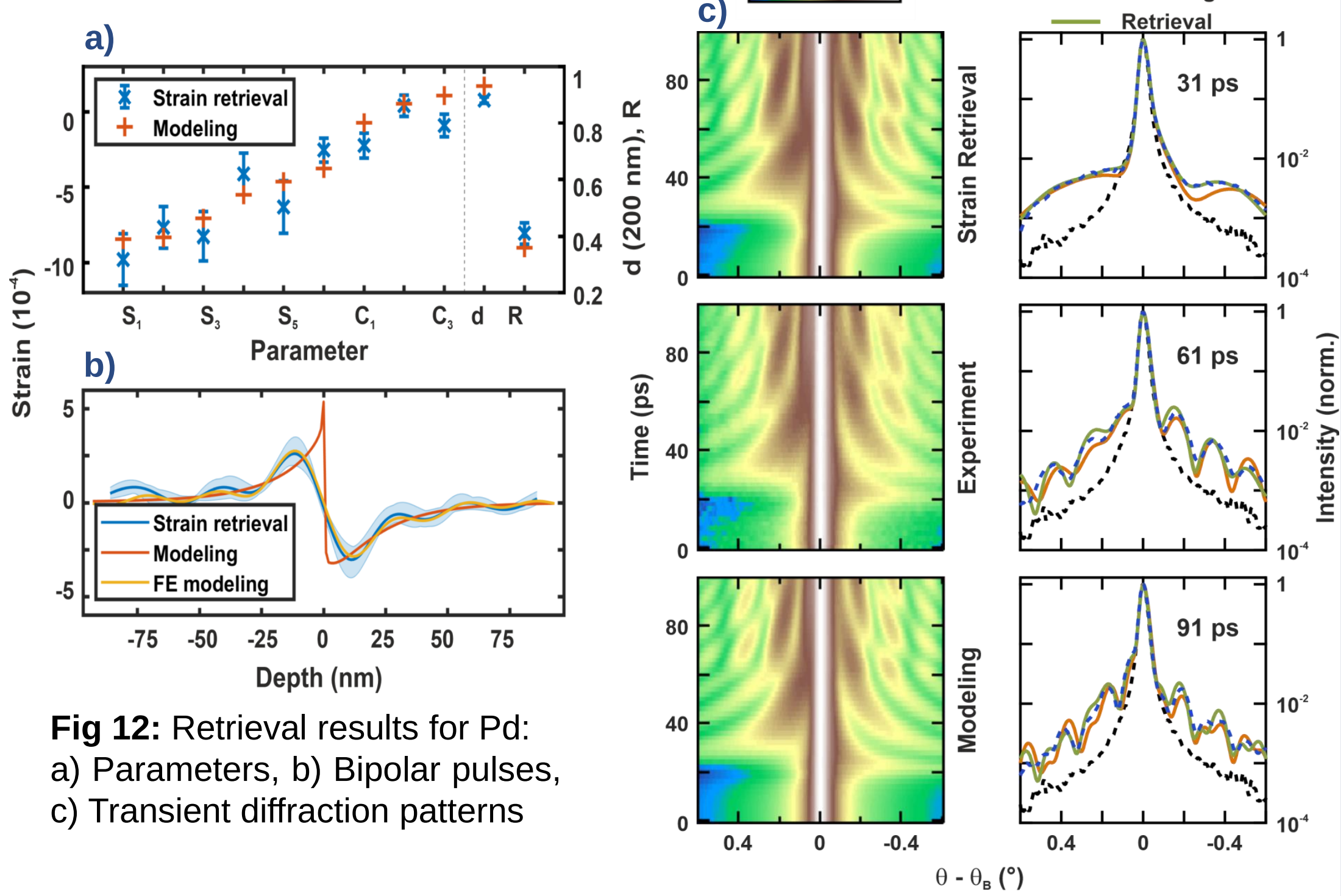


Fig 12: Retrieval results for Pd:
a) Parameters, b) Bipolar pulses,
c) Transient diffraction patterns

Conclusion and Outlook:

DNNs are capable of retrieving strain from time-resolved X-ray diffraction patterns

Switch to more favorable material systems (e.g. GaAs (111)) as next step

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