EPJ Web of Conferences **205**, 08005 (2019) https://doi.org/10.1051/epjconf/201920508005
*UP 2018*

# Structural phase transitions and phase ordering at surfaces probed by ultrafast LEED

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**Abstract.** We demonstrate the capability of ultrafast low-energy electron diffraction to resolve phase-ordering kinetics and structural phase transitions on their intrinsic time scales with ultimate surface sensitivity.

## 1 Introduction

Solid state surface systems are particularly attractive because of their strongly altered physical and chemical properties compared with the bulk [1]. The lower dimensionality and broken symmetry of a surface gives rise to a variety of phenomena associated with modified electronic, lattice and spin structures, which in many cases are correlated with each other. We have recently developed ultrafast low-energy electron diffraction (ULEED) to explore optically-induced structural dynamics in the time-domain, exploiting the superior surface sensitivity and momentum resolution of LEED to resolve structural phase transitions and phase-ordering processes at surfaces [2,3]. Here, we present the observation of phase-ordering mechanisms and phason dynamics in the incommensurate charge density wave (CDW) phase of 1T-$TaS_2$, enabled by time-resolved spot-profile analysis [3]. Furthermore, we report on the first time-resolved LEED studies of the structural phase transition associated with the insulator-metal transition in atomic Indium chains on a Si(111) surface.

## 2 Experiment

To realize ultrafast low-energy electron diffraction (ULEED), we employ low-energy electron pulses in a laser-pump/electron-probe scheme (Fig. 1a). Within this approach, a nanoscopic needle emitter triggered by 400 nm laser pulses (40 fs duration) via two-photon photoemission (2PPE) is utilized in a miniaturized electrostatic lens geometry as a high-brightness source. The resulting electron pulses exhibit a minimal duration down to 1.3 ps at the sample for electron energies of 20-200 eV [3,4]. The strongly confined emission area leads to a high transversal coherence length of the pulsed electron beam, resulting in a momentum resolution of $\Delta k_s = 0.03$ Å$^{-1}$, which corresponds to a transfer width of $2\pi/\Delta k_s$ = 21 nm, at a spot size on the sample below 100 µm (FWHM).

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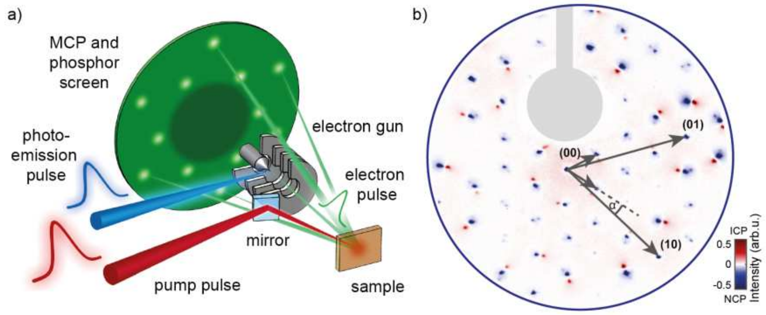


**Fig. 1.** a, Sketch of the optical-pump electron-probe scheme of the ULEED experiment. b, Difference image of the NC (blue) and IC (red) diffraction patterns (adapted from Ref. [3]).

## 3 Results and Discussion

We investigate the structural dynamics induced by the transformation of charge density waves at the surface of the quasi two-dimensional material 1T-$TaS_2$ [3]. Specifically, we map the laser-driven transition from the room-temperature nearly-commensurate (NC) to the high temperature incommensurate (IC) CDW phase (Fig. 1b). Utilizing the high momentum resolution of the ULEED setup, we perform a thorough spot profile analysis of the appearing IC CDW diffraction peaks and observe a coarsening behaviour in the newly created IC phase (Fig. 1c). This growth of the IC CDW coherence length is attributed to the annihilation of dislocation-type topological defects. To corroborate our experimental observations, we perform numerical simulations of the IC CDW phase-ordering process in a time-dependent Ginzburg-Landau approach.

In a second set of experiments, we study collective phase-excitations, so-called phasons, in the IC phase [5]. After exciting the sample with an intense light pulse, we monitor the subsequent energy redistribution from the electron- into the phason-/phonon-subsystems. To this end, we determine the time-dependent temperature changes in the respective subsystems by a comparison of the pump-induced intensity suppressions of Bragg- and CDW-satellite-reflections. Our results indicate a strong initial energy transfer to the phason system, followed by slower phason-phonon equilibration, which suggests an inhibited electron-phason relaxation channel. We attribute this observation to a gap-induced decoupling of electron and phason systems, resulting in the pronounced generation of hot phasons.

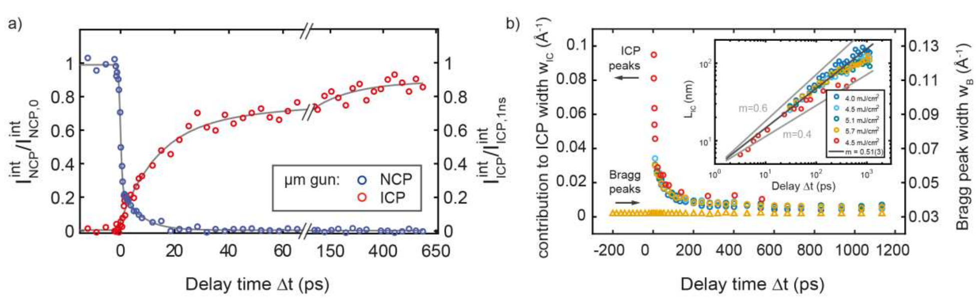


**Fig. 2.** a, Integrated CDW diffraction peak intensities recorded using the µm-sized gun. b, Time-dependent contribution to the width (FWHM) of the ICP diffraction peaks and width of lattice diffraction peaks for a range of optical-pump fluences. Red circles: data recorded with µm-sized gun. Inset: A double-logarithmic plot of the correlation length corrected for the instrument response function, indicating a power-law scaling for the phase-ordering kinetics (adapted from Ref. [3]).

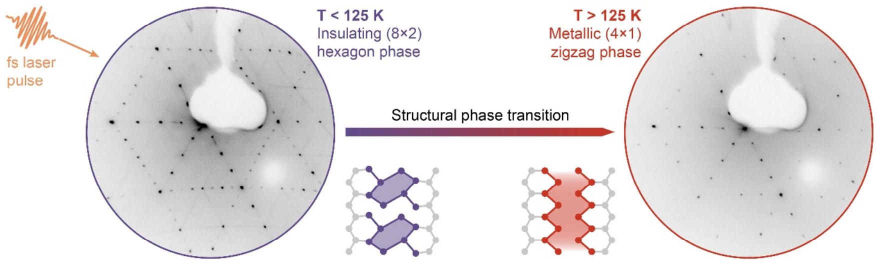


**Fig. 3.** Diffraction images and structure models of the low-temperature (8×2) and the high-temperature (4×1) phase of Indium atomic wires on a Si(111) surface recorded with 130eV electron pulses in our ULEED setup. The twofold streaks in the diffraction pattern of the (8×2) phase are the result of the missing correlation between adjacent atomic rows.

We further use the high surface sensitivity of our ULEED setup to study the surface-specific transition between two CDW states of self-assembled Indium wires on Silicon. The sample is prepared by *in situ* evaporation of about one monolayer of Indium on a clean Si(111) surface followed by subsequent annealing and cooling down to 60 K. Following optical excitation, the system undergoes a Peierls-type transition from a low-symmetry insulating to a high-symmetry metallic phase, accompanied by a structural phase transition between an (8×2) and a (4×1) superstructure [6]. We report on the first findings regarding the underlying structural dynamics probed by Ultrafast LEED.

## 4 Summary and Conclusion

In conclusion, using the example of 1T-$TaS_2$, we reported the first systematic observation of ultrafast phase-ordering kinetics and phason dynamics enabled by the development of ultrafast low-energy electron diffraction (ULEED). We further explore the potential of this new technique by studying surface-specific structural phase transitions, such as Indium atomic chains on Si(111). Serving as an ideal complement to ultrafast angle-resolved photoelectron spectroscopy (ARPES), we believe that ULEED will pave the way for a greater understanding of a multitude of surface phenomena and consequently the optical control of complex surface dynamics, including phase transitions and chemical reactions.

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