Ultrafast nano-imaging of the order parameter in a structural phase transition

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Over the past decades, ultrafast optical techniques have considerably shaped our understanding of homogeneous materials, while transmission electron microscopy has greatly contributed to elucidating atomic structures and compositions on the sub-nanometer scale. Combining these concepts, ultrafast transmission electron microscopy allows for resolving femtosecond dynamics in heterogeneous materials using imaging, diffraction, and spectroscopy in a laser pump/electron probe scheme [1].

Here, we demonstrate the ultrafast real-space mapping of the order parameter for a charge- density wave (CDW) phase transition in the correlated material 1*T*-TaS_2 using the Göttingen Ultrafast Transmission Electron Microscope (UTEM) [2]. In the experiments, a free-standing, single-crystalline 1*T*-TaS_2 thin film [3] is pumped out of the nearly commensurate CDW phase at room temperature towards the high-temperature incommensurate CDW phase using a spatially structured laser field distribution (see Fig. 1A for a schematic of the experimental setup). The transient state of the specimen is recorded using a direct electron detection camera in counting mode.

Specifically, we employ ultrafast dark-field imaging to follow the formation, evolution, and relaxation of CDW domain patterns on their intrinsic femtosecond to nanosecond timescales, yielding nanoscale access to the order parameter of the structural phase transition (see Fig. 1B and C). Additionally, we show that prominent features in the spatiotemporal domain evolution can be modeled in a time-dependent Ginzburg-Landau approach, allowing us to distinguish different regimes of the observed dynamics [4].

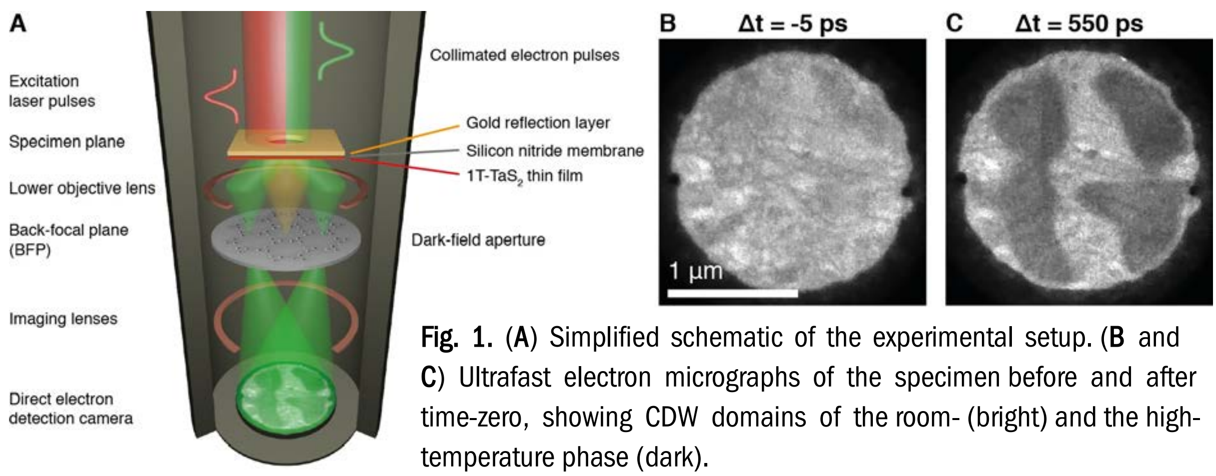

Fig. 1. (**A**) Simplified schematic of the experimental setup. (**B** and **C**) Ultrafast electron micrographs of the specimen before and after time-zero, showing CDW domains of the room- (bright) and the high-temperature phase (dark).

References

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[3] Th. Danz *et al.*, *J. Phys.: Condens. Matter* **28**, 356002 (2016). [4] Th. Danz *et al.*, in preparation.