doi:10.1017/S1431927621010308
Microsc. Microanal. 27 (Suppl 1), 2021

Ultrafast nanoimaging of the order parameter in a structural phase transition

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Strategies to optically control and modify physical material properties have a strong impact in current and future technologies. The underlying dynamical processes generally take place on femto- to picosecond timescales, a regime accessible by a wide variety of stroboscopic pump/probe approaches. When applied to actual devices, however, functionality usually also arises from nanoscale structuring or an interplay of different materials. This calls for experimental approaches capturing optically induced dynamics in terms of a spatially dependent order parameter.

In this work, we demonstrate a new experimental approach enabling real-space phase imaging in a charge-density wave (CDW) model system with 5nm spatial and femtosecond temporal resolution. By means of a tailored dark-field (DF) approach, we obtain maps of the CDW order parameter in an ultrafast transmission electron microscope [1].

Ultrafast transmission electron microscopy (UTEM) is an emerging approach to study ultrafast processes in heterogeneous materials with nanometer spatial resolution [2]. Using the imaging, diffraction, and spectroscopy capabilities of such an instrument, we investigate structural, electronic, and magnetic dynamics in a laser pump/electron probe scheme. To this end, the Göttingen UTEM is equipped with a nanoscopic tip emitter, delivering highly coherent electron pulses by linear photoemission with down to 200fs pulse duration, 0.6eV energy width, and a sub-nm spot diameter [3].

These exceptional beam properties allow for a versatile use of the Göttingen UTEM, as demonstrated in recent years [4-10]. However, some of the most interesting possibilities are connected to the investigation of various kinds of structural and electronic phase transitions in correlated materials, such as transition metal dichalcogenides, e.g., 1*T*-TaS_2. This quasi-2D material favors a periodic modulation of the electronic density, i.e., the formation of CDWs coupled to a periodic lattice distortion (PLD) [11]. Various ultrafast electron diffraction experiments have elucidated the optically induced dynamics of transitions between several CDW/PLD phases [12-15].

In our experiments, a free-standing, single-crystalline 1*T*-TaS_2 thin film prepared by ultramicrotomy [16] is pumped out of the nearly commensurate (NC) CDW phase at room temperature towards the high-temperature incommensurate (IC) CDW phase (see Fig. 1A for a schematic of the experimental setup). We employ ultrafast DF imaging using a tailored DF aperture array (Fig. 1B) to obtain an image contrast that directly reflects the local order parameter of the NC CDW. Figure 1C shows the effect of the DF aperture array in the back-focal plane (BFP) of the microscope.

In the resulting DF micrographs (Fig. 2A), we observe the formation, stabilization, 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. The inhomogeneous suppression of intensity directly after time-zero is governed by a spatially structured laser field distribution (Fig. 2E), triggering the spatiotemporal dynamics in this experiment.

The DF micrographs contain considerably more information than spatially averaged diffraction data alone. While the practically binary contrast allows us to segment the images into NC and IC CDW regions (Fig. 2B)

and determine the area fractions occupied by both CDW phases (Fig. 2C, top), we can also individually analyze the dynamics in weakly and strongly pumped regions (green and orange curves in Fig. 2C, bottom) and compare this to the overall signal (black curve). Additionally, we observe a sharpening of NC/IC domain walls over time (Fig. 2D).

From a complementary steady-state heating experiment, we also obtain a precise thermal characterization of the specimen structure. On this basis, we model the spatiotemporal domain evolution using the time-dependent Ginzburg-Landau framework. We demonstrate that this approach reproduces the most prominent features of the ultrafast experiment, elucidating relaxation pathways and domain wall dynamics.

In conclusion, we have reported on the first ultrafast real-space imaging of CDW dynamics. Combined with static specimen characterization and theoretical modeling, ultrafast DF imaging allows for intriguing insights into the interplay of order parameter dynamics and thermal transport on nanometer length and femtosecond timescales. Allowing for sensitivity to further degrees of freedom in complex materials, these results will hopefully pave the way for novel types of ultrafast investigation.

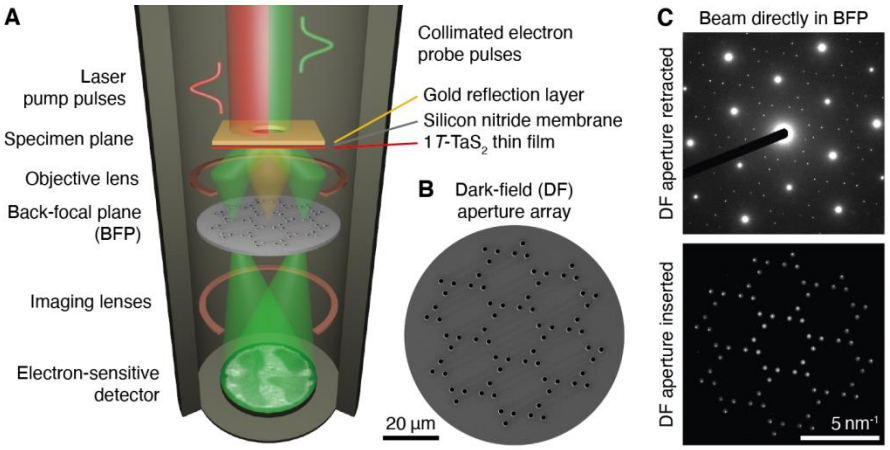

Figure 1. Dark-field imaging in the ultrafast transmission electron microscope. (A) Sectional drawing of the experimental setup. Electron (green) and optical pulses (red) are incident close to perpendicular on the specimen. (B) Scanning electron micrograph of the tailored dark-field (DF) aperture array placed in the back-focal plane (BFP) in (A). (C) Electron diffractograms of the 1T-TaS2 thin film at room temperature with the DF aperture array shown in (B) retracted from (top) and inserted into the electron beam path (bottom). Only NC CDW superstructure reflections are transmitted through the aperture array.

https://doi.org/10.1017/S1431927621010308 Published online by Cambridge University Press

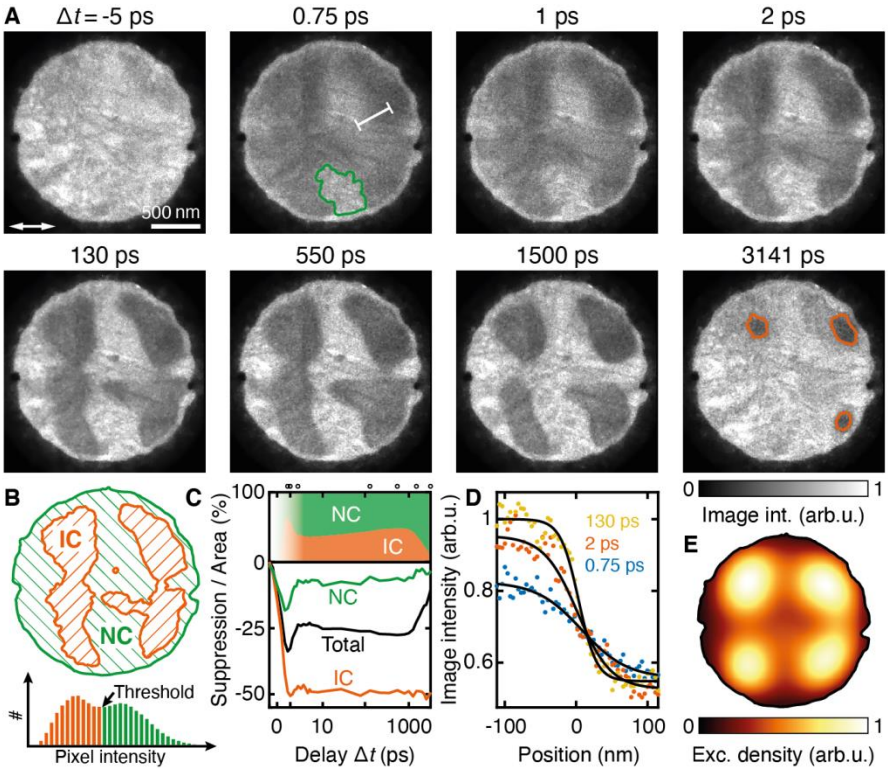

Figure 2. Ultrafast dark-field domain imaging of charge-density wave dynamics. (A) Ultrafast DF micrographs of transient domain configurations in the 1T-TaS2 film obtained in the laser pump/electron probe scheme (2.6 mJ/cm2 pump fluence, linear pump polarization indicated by white arrow). Pump/probe delay steps were chosen so as to capture all major stages of the dynamics (see black circles above (C)). (B) Top: Image segmentation at 130 ps delay time. Bottom: The segmentation threshold is determined from the intensity histogram of the full image series within the circular aperture. (C) Top: Area fractions of NC and IC regions after completed phase separation, as determined from the segmented images. Bottom: Average intensity of the image series within the entire aperture (black curve), and average intensity in weakly and strongly pumped regions (green/orange curve; evaluated regions are indicated in (A) using corresponding colors). (D) Exemplary profiles of NC/IC phase boundaries taken on the white line indicated in (A). (E) Spatial profile of the excitation density giving rise to the initial suppression pattern.

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https://doi.org/10.1017/S1431927621010308 Published online by Cambridge University Press