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Time Resolved Electron Diffraction in the Nearly Commensurate Charge-Density-Wave Phase of 1T-TaS_2

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Femtosecond time-resolved spectroscopy has become an established new tool for studying the relaxation phenomena in strongly correlated electron systems. The ability of these techniques to distinguish various spectral components based on their different relaxation timescales has enabled researchers to obtain information about the interaction strengths between various degrees of freedom (electrons, spin, lattice etc.) in these systems.

Numerous femtosecond time-resolved experiments have been performed on charge density wave (CDW) compounds, utilizing all-optical [1] and time-resolved ARPES techniques [2]. Common to all these studies is the observation of a coherently driven amplitude-mode and two distinct relaxation timescales; one on the order of 100 femtoseconds and the second on the order of several picoseconds. In the CDW systems showing a second order phase transitions from the CDW to the normal (metallic) state, the fast process was found to exhibit critical slowing down in the vicinity of the phase transition, and was therefore commonly attributed to the order parameter recovery [1], assuming that the electron subsystem adiabatically follows the lattice. Recent studies, however, suggest that this may not be the case [3,4]. Systematic photoexcitation intensity dependence studies revealed that the non-thermal phase transition between the low temperature (T) CDW and the high temperature metallic state can be driven with an intense optical pulse [3]. However, these experiments suggest that during the melting of the electronic modulation and its subsequent (partial) recovery, both of them taking place on the timescale of ~ 100 fs, the lattice remains nearly frozen [3]. Similarly, the analysis of the order parameter dynamics in a quasi 1D CDW K0.3MoO3 with the time-dependent-Ginzburg-Landau model suggests [4] the fast process to be related to the purely electronic mode and not to the coupled electron-lattice order parameter. All of the experiments prior to this work have investigated solely the dynamics of the electronic system. The intriguing question of interplay between the electronic and lattice degrees of freedom remained an open issue. To study the dynamics of the lattice part of the order parameter and the corresponding superstructure, time resolved structural probes are required.

Here we report on first studies of structural dynamics in the quasi two-dimensional CDW system 1T-TaS2, where the dynamics of the CDW's periodic lattice distortion (PLD) has been investigated by means of femtosecond electron diffraction (FED). The results demonstrate an extremely fast suppression of the CDW modulation, faster than the period of the amplitude mode. The recovery of the CDW state is, however, found to proceed on the picosecond timescale - the longer of the two timescales observed by optical experiments.

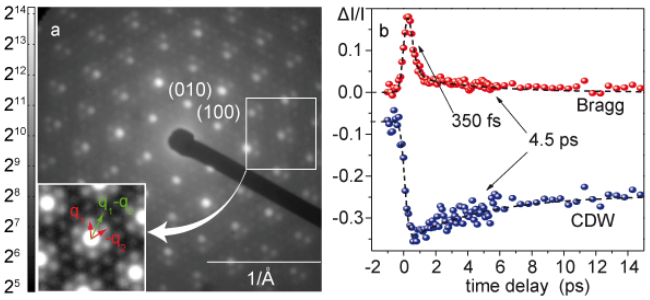

Figure 1. Panel **a** shows the equilibrium diffraction image of a 30nm thin 1T-TaS_2 film at a temperature of 200K. Two of the bright Bragg reflections are indexed and the inset shows a symmetrized and rescaled blow up of the white square. Each Bragg reflection is surrounded by six satellites, arising from the CDW period of q_i. Secondary CDW reflections are also observed. Panel **b** shows transient relative intensity changes of Bragg and CDW reflections upon excitation with a 2.4 mJ/cm^2, 400 nm laser pulse. The corresponding timescales are indicated and given by the fitting curves (dashed lines).

The experiments have been performed on 30 nm free standing single crystalline 1T-TaS2 films in transmission geometry, utilizing the experimental set-up described elsewhere [5]. The sample was photoexcited with a 140 fs optical pulses at 400 nm, while the time evolution of the diffraction pattern was probed by a time-delayed sub 250 fs (50 keV) electron pulse. The experiments were performed in the nearly-commensurate CDW phase at 200 K applying excitation densities of 2.4 and 4.8 mJ/cm2. The equilibrium diffraction pattern recorded in this set-up is displayed in

Figure 1**a**, together with the assignment of the scattering vectors (insert). Each of the main Bragg peaks, showing 6 fold symmetry, is surrounded by six first order CDW peaks at q_i, together secondary reflections q_i-q_j. Figure 1**b** presents the time evolution of the intensity of the Bragg and the CDW peak, following photoexcitation with a 140 fs pulse at 2.4 mJ/cm^2. Strong suppression of the CDW intensity is observed, happening within hundreds of fs. This is accompanied by a rapid increase of the intensity of Bragg reflections, consistent with the suppression of the CDW order. The intensity of the Bragg peaks is again suppressed on a timescale of ~350 fs due to increase in the lattice temperature by energy transfer from electrons to q≠0 phonons.

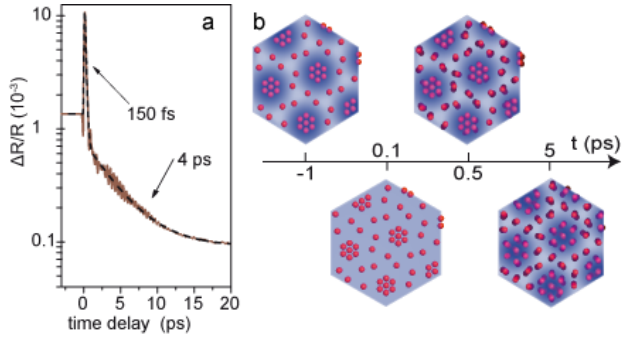

Figure 2. Panel **a** depicts the transient relative change in reflectivity at 800 nm recorded with the same excitation fluence and temperature as used for the transient diffraction measurements, together with its fit (dashed line). The oscillatory superposition on the biexponential decay exhibits among others a 2.3 THz oscillation, which is commonly attributed to the CDW amplitude mode [6]. Panel **b** shows the real-space structure evolution of the Ta plane of TaS_2 following photoexcitation (circles represent Ta atoms, the blue shading represents the density of conduction electrons). Prior to photoexcitation (t ≈-1ps), the Ta atoms are periodically displaced from their pure 1T structure, forming the nearly commensurate CDW. Intense perturbation of the electronic system gives rise to a homogenization of the electron density (t≈0.1 ps), driving the lattice towards the undistorted state. In parallel, the energy is transferred from the electronic subsystem to q≠0 phonons on the 300 fs timescale, resulting in recovery of the electron density modulation and thermal disordering of the lattice (t≈0.5 ps). The CDW order is recovered at t≈5ps, after which time the sample is thermalized at a somewhat higher temperature.

This process can also be monitored in the change of the inelastic background (not shown here), which rises simultaneously and with the same time constant as the Bragg peaks are suppressed. The CDW recovery proceeds with a time constant of ~4 ps. These data can be compared to the data obtained under the same conditions in an all-optical configuration, see Figure 2**a**. As already mentioned, the reflectivity transient has a biexponential decay, with the longer decay time matching the decay time of the CDW intensity as obtained by the FED experiment. The faster timescale of ~150 fs is not observed in the FED experiment and is therefore solely attributed to the purely electronic relaxation of the perturbed CDW. The findings of both the time resolved electron diffraction and the all optical pump probe studies are summarized in Figure 2**b** and its caption. More details can be found in the published literature [7].

In conclusion, we have demonstrated that FED in transmission can be used to study complex phenomena like the order parameter dynamics in charge density waves. The new structural information illustrates the direct optical manipulation of the order parameter that leads to a suppression of the CDW amplitude on a timescale of several hundred fs. This timescale is comparable to a half a period of the corresponding collective mode, implying that suppression of the periodic lattice distortion amplitude (order parameter) is electronically driven and of highly collective nature. The recovery of the CDW however proceeds on the timescale of several ps, implying that the ultrafast initial process, studied extensively with all-optical techniques, is related to the partial recovery of the electronic part of the order parameter alone. Clearly further experimental and theoretical efforts are required to further clarify these observations.

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