Time-domain identification of electronic correlations in 1*T*-TaS_2

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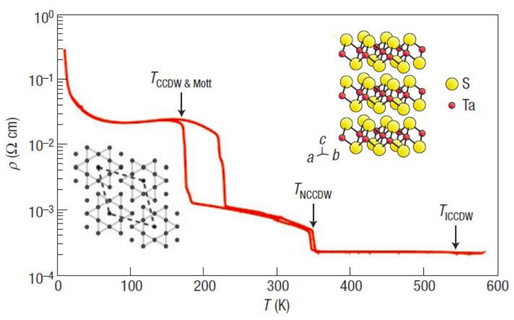
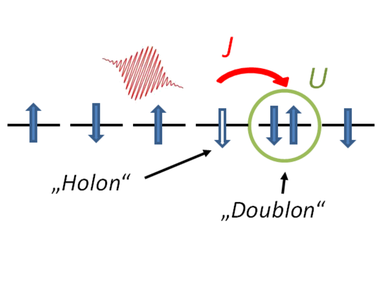
Motivation

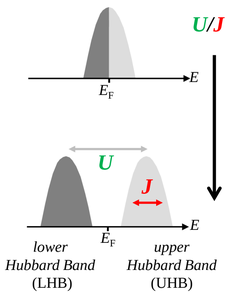
"Complex" matter is characterized by simultaneously strong interaction between different microscopic degrees of freedom, often resulting in rich phase diagrams where tiny variations of controllable parameters can lead to significant changes of the macroscopic material properties. These interactions can occur on comparable energy scales and, thus, might be hard to disentangle under equilibrium conditions. Here we used time-resolved photoemission spectroscopy to investigate the non-equilibrium electron dynamics of 1*T*-TaS_2 after optical perturbation. Despite strong electron-phonon interaction in the system, fundamental Mott physics can be isolated by monitoring the population dynamics in the upper Hubbard band.

1*T*-TaS_2

The transition metal dichalcogenide 1*T*-TaS_2 exhibits a manifold of different phases due to simultaneously strong electron-electron and electron-phonon interaction. The formation of charge density waves of different commensurability is believed to be accompanied by a Mott Hubbard transition. This widely accepted picture is still under debate and the exact role of electronic correlation effects in the metal to insulator transition is not clear [Ritschel et al., Nature Phys. **11** (2015), Yu et al., PRB **96** (2017)].



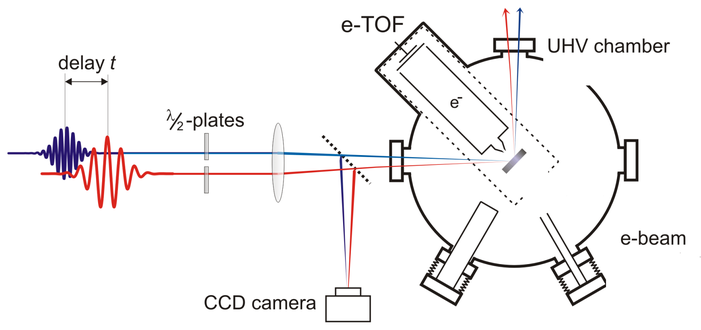


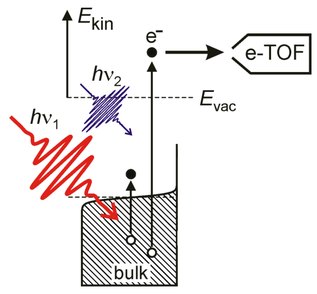


Sipos et al., *Nature Mat.* **7** (2008)

Despite its simplicity, a full theoretical treatment of the Fermi Hubbard Model is still an ongoing challenge, in particular under non-equilibrium conditions [Aoki et al., Rev. Mod. Phys. 86 (2014)]. Under equilibrium conditions, sufficiently strong on-site Coulomb interaction *U*/*J* between electrons results in a metal-to-insulator transition that is reflected by the formation of an occupied lower Hubbard band (LHB) and an unoccupied upper Hubbard band (UHB). The non-equilibrium signatures and corresponding dynamics of such systems are, however, only scarcely studied.

Time-resolved Photoemission

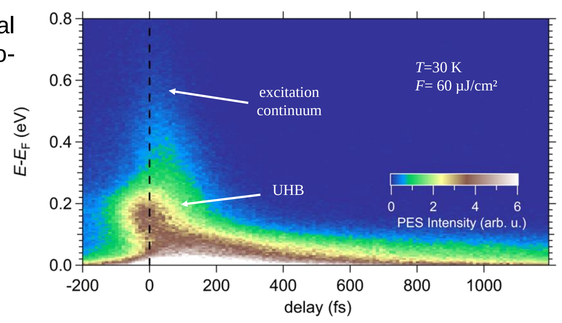




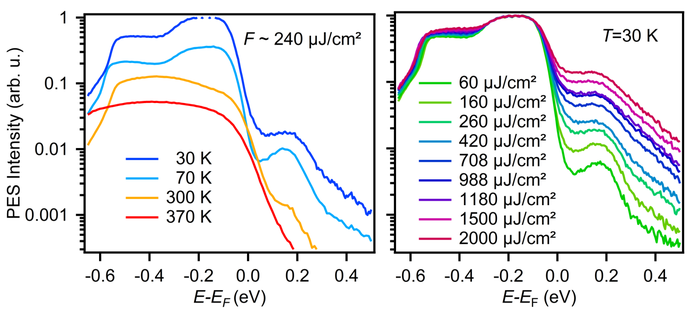
In agreement with earlier work, three distinct spectral responses can be observed upon **intense** photo-excitation of the material:

(i) the generation of hot charge carriers
(ii) a depletion of the lower Hubbard band (LHB)
(iii) oscillations of the spectral weight due to the excitation of coherent optical phonons.

Perfetti et al., PRL **97** (2006)
Petersen et al., PRL **107** (2011)
Hellmann et al., Nature Comm. **3** (2012)



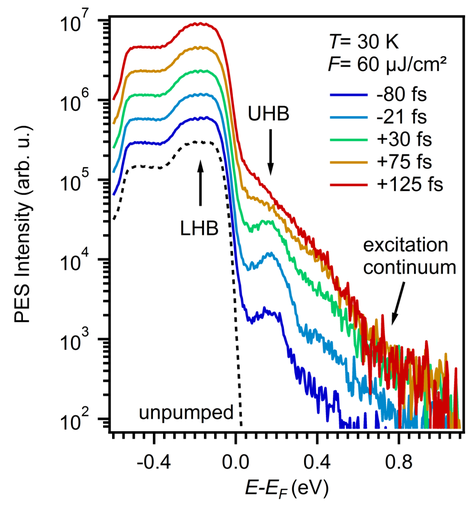
In the particular case of **weak** excitation, we also observe the appearance of a spectroscopic line at E-E_F≈200 meV., e.g. where the upper Hubbard band is expected. This spectroscopic signature is in particular visible for low excitation densities and at low sample base temperatures (in the correlated phases of the material).

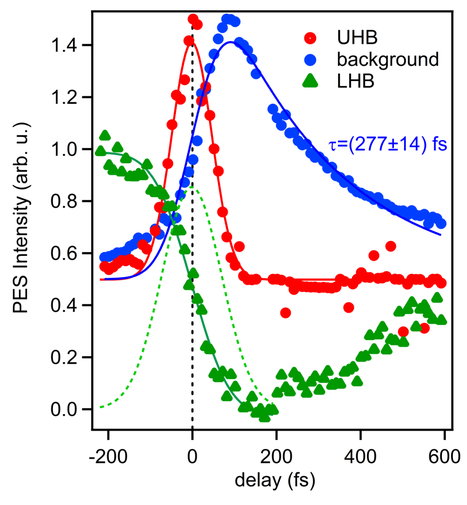


Time-averaged photoemission spectra (Δt=-125..-65 fs) as a function of sample temperature (left) and excitation fluence (right).

In agreement with recent inverse photoemission and scanning tunneling microscope studies, we identify the unoccupied electronic signature as the upper Hubbard.

Upper Hubbard Band Dynamics



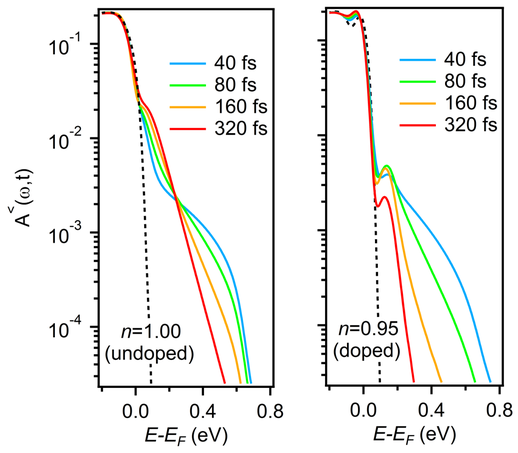


The UHB dynamics occur quasi-instantaneously and represent the fastest process found in the system. The dynamics furthermore coincide the collapse of the lower Hubbard band that was previously shown to occur on time scales faster than 20 fs [Petersen et al., PRL **107** (2011)]. We conclude that the relevant time scales are of the order of one or few hopping cycles ħ/*J*=14 fs, significantly shorter than, e.g., in simple metals,. Furthermore, this timescale is shorter than a quarter-period of the highest frequency vibrations found in 1*T*-TaS_2 (21 fs) [Gasparov et al., Rev. Mod. Phys. **68** (2002)], indicating a decoupling of CDW and Mott physics on ultrashort time scales.

Nonequilibrium dynamical mean field theory

We consider a purely electronic model to identify the role of electronic correlations in the ultrafast dynamics observed experimentally.

- Parameters chosen such that equilibrium spectra are reproduced (bandwidth *W*=0.36 eV)
- Pump pulse included via Peierls substitution ω/*J*=8.0, single cycle envelope, $E_0/(Je)$=2.0
- Pure electronic model (no coupling to bosonis)
- Triagonal lattice geometry

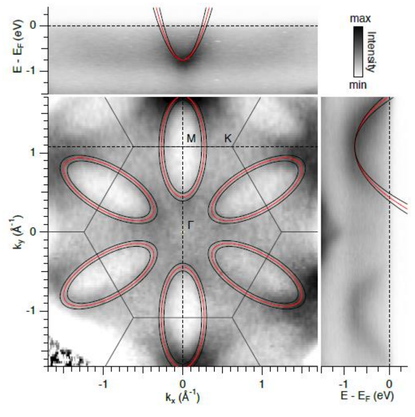


$$H = \sum_{i\delta\sigma} J c^{\dagger}_{i+\delta,\sigma} c_{i+\delta,\sigma} + \mu n_i + U \sum_i (n_{i\uparrow} - \frac{1}{2})(n_{i\downarrow} - \frac{1}{2})$$

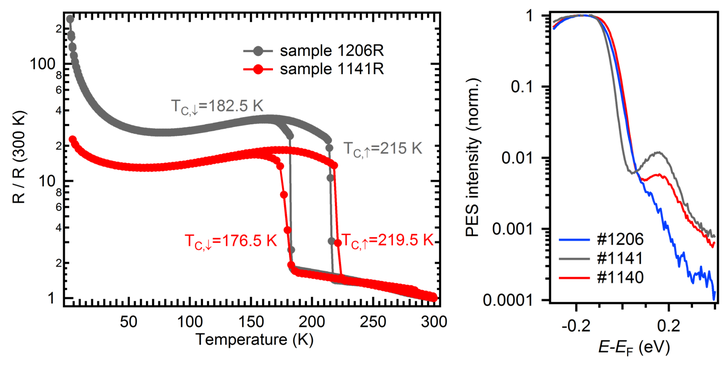
The results obtained for a half filled band (*n*=1.00) do not reproduce the experimental findings (slow population build-up at the low energy UHB edge). However, the assumption of a slightly hole-doped system (*n*=0.95) results in significantly different dynamics that qualitatively reproduce the experimental results. We conclude that the presence of holes dominates the relaxation and thermalization dynamics on ultrashort time scales.

1*T*-$Ta_{(1-x)}S_2$

Further experiments suggest that our samples are indeed effectively hole doped due to non-perfect stochiometric ratios (1*T*-$Ta_{(1-x)}S_2$ with *x* of the order of 0.01). This deficit of Ta atoms can result in band fillings ranging from half-filled (*x*=0) to quarter-filled (*x*=0.01).



ARPES Fermi-surface map recorded at T=390 K (hω=270 eV). Red lines indicate extracted elliptical Fermi contours. Black lines indicate the uncertainty in the analysis. An effective band-filling of 0.81±0.16 was extracted. Data was recorded at Beamline P04 of Petra III (DESY).



Comparison of different sample batches. (left) Normalized resistance as a function of temperature upon heating and cooling. Only minor differences between the differnet samples are observed. (right) Corresponding time-averaged photoemission spectra obtained from the same samples. For sample #1141, the UHB signature is clearly visible while for sample #1206, it is absent. A third sample (#1140) is also shown.

Conclusions

Our results show that electronic correlation effects in a complex system can be isolated in the time domain. Indeed, the dynamics of photo-excited doublons occur on time scales as short as one or few electronic hopping cycles ħ/*J*. We emphazise the importance of static or photo-generated holes, since they mediate the ultrafast relaxation and thermalization on hopping timescales. A preprint of this work can be found on arXiv: Ligges et al., arXiv:1702.05300

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