# Ultrafast Carrier Dynamics in Few-Layer Colloidal Molybdenum Disulfide Probed by Broadband Transient Absorption Spectroscopy

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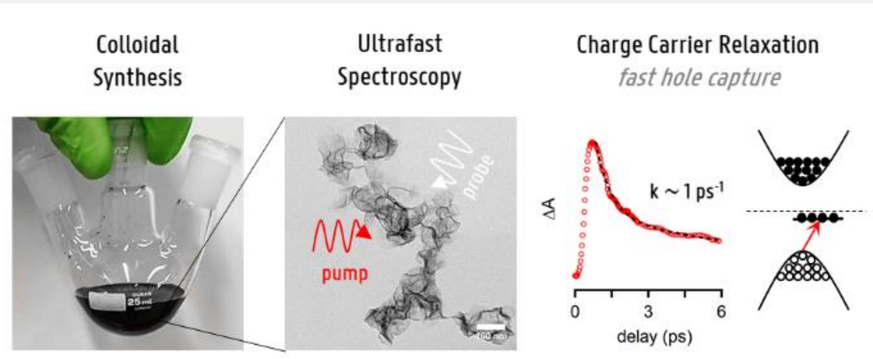
## Introduction

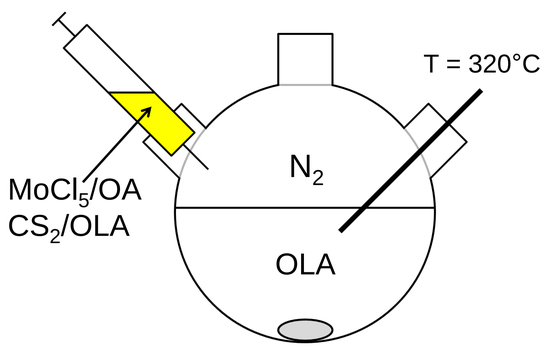
Insights into the photophysics of TMDs made by exfoliation or CVD have been pivotal for the advancement toward a broad range of applications

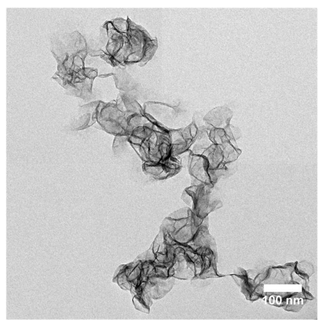
Colloidal synthesis has emerged as an inexpensive, scalable and highly-tunable alternative to exfoliated and CVD-grown TMDs

If colloidal methods have to become a true alternative, they should be benchmarked relative to CVD-grown and exfoliated TMDs



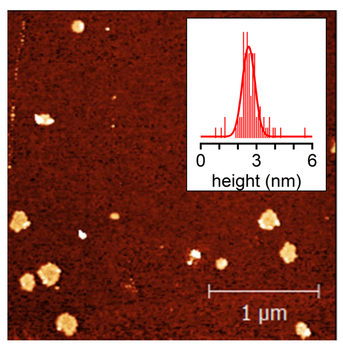

## Colloidal Synthesis of Molybdenum Disulfide

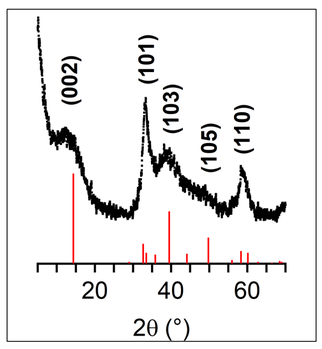


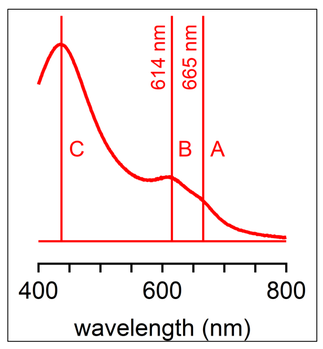




**bottom-up synthesis of $MoS_2$**

nanosheets (TEM)



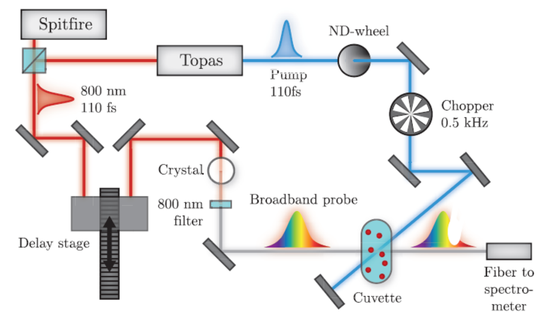



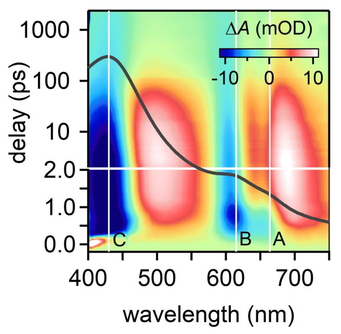

few-layered (AFM)

2H $MoS_2$ (XRD and UV-vis)

The synthesis yields few-layered colloidal 2H $MoS_2$ nanosheets

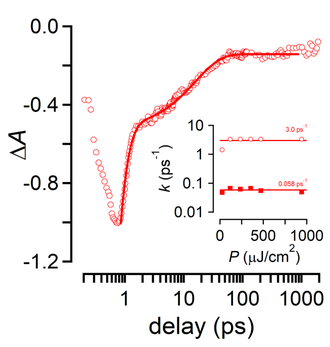
## Transient Absorption – Band Gap Renormalization

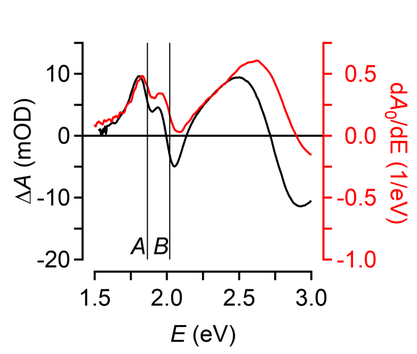


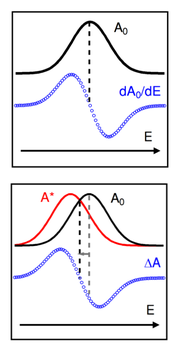




**pump-probe spectroscopy**
measure transient absorbance $\Delta A$

multiple bands of $\Delta A < 0$ and $\Delta A > 0$ around excitons



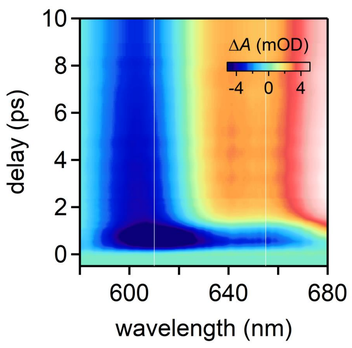




first order kinetics

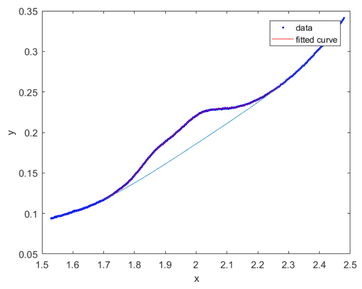
spectrum reflects a spectral shift

A spectral shift of the transition energies governs the $\Delta A$ spectrum at long pump-probe delays

## Transient Absorption – Exciton Dynamics

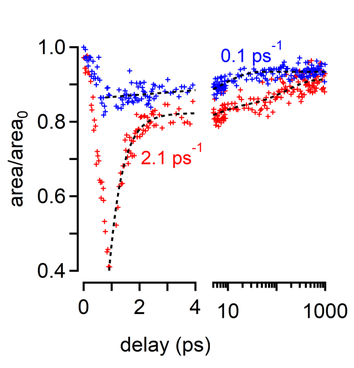


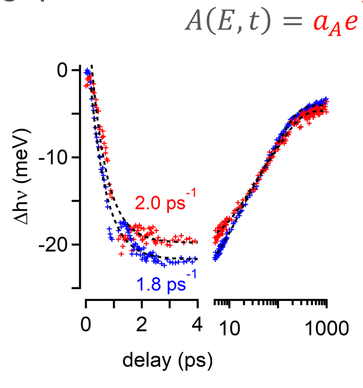

short-living bleaches around band-gap

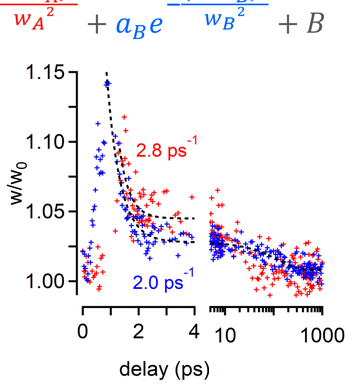



**deconvultion of excited state spectrum** $A = A_0 + \Delta A$ according to

$$A(E,t) = a_A e^{-\frac{(E-E_A)^2}{w_A^2}} + a_B e^{-\frac{(E-E_B)^2}{w_B^2}} + B$$



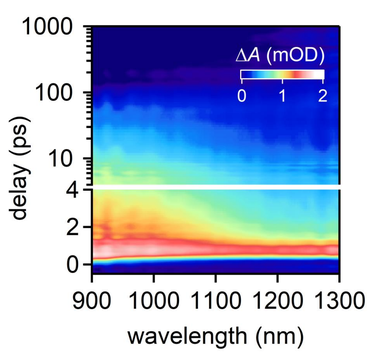



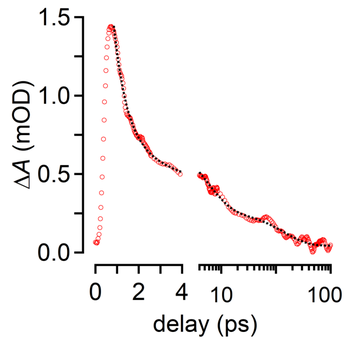

netto bleaches due to state filling
rapid decay of *A* exciton bleach

dynamics of the spectral shift and broadening happen on a remarkably similar time scale

A time-dependent spectral deconvolution of $\Delta A$ highlights simultaneous rapid changes on the state filling, spectral shifting and broadening dynamics

## Transient Absorption – Intraband Absorption

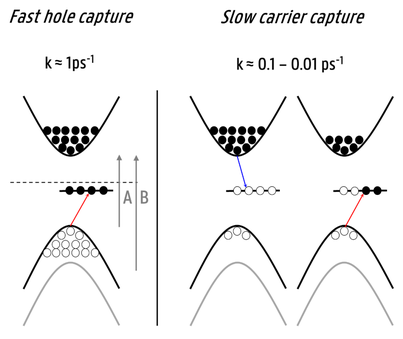



photoinduced absorption related to intraband transitions

similar dynamics above gap and sub-gap

## Conclusion – Model for Charge-Carrier Decay




**trapping of holes**
decay net *A* bleach
decay sub-gap photoinduced abs.
first-order decay

**slower non-rad. capture events**
relaxation of band-gap renorm.

A comparison between the colloidal materials as measured here and the existing literature on CVD-grown TMDs points toward very similar photogenerated carrier dynamics, eventhough an entirely different chemistry is involved in the TMD production

## Acknowledgements

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