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Hot electron driven charge transfer dynamics at the MoS_2/gold interface

SFB 1242

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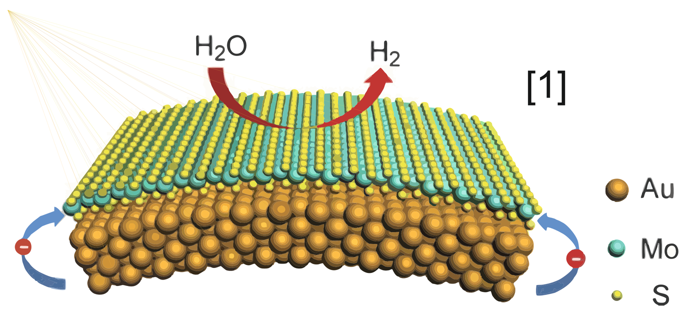
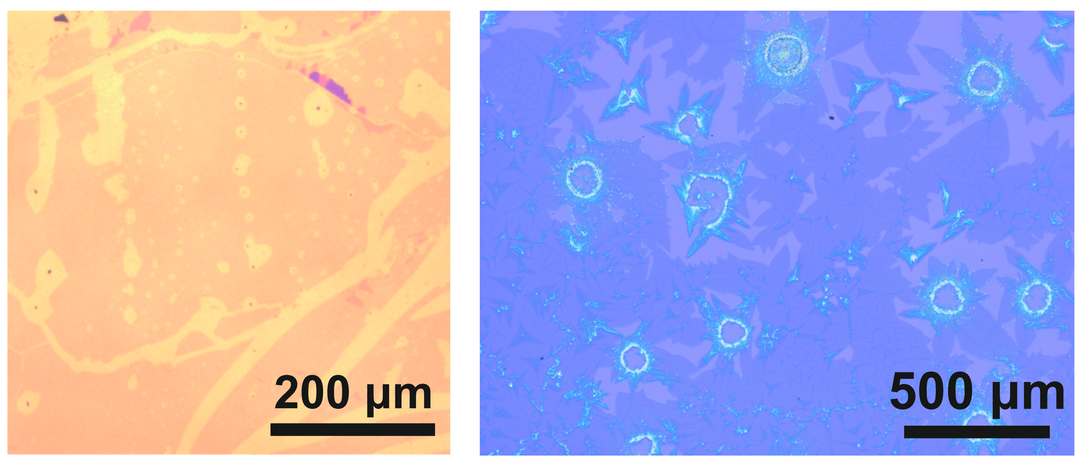
Introduction

- The function of transition-metal dichalcogenide (TMD) based optoelectronic devices is determined by the electron transfer across the TMD/Metal interface.
- However, the inter- and intraband excitation and relaxation within the metal and TMD, the screening effect of the metal, and the different channels of charge transfer between the substrate and adsorbate make the interpretation of the experimental observations challenging
- Below optical band gap pump and final state sum frequency generation (SFG) probe allows us to disentangle the different contributions to the complicated dynamics.



Sample

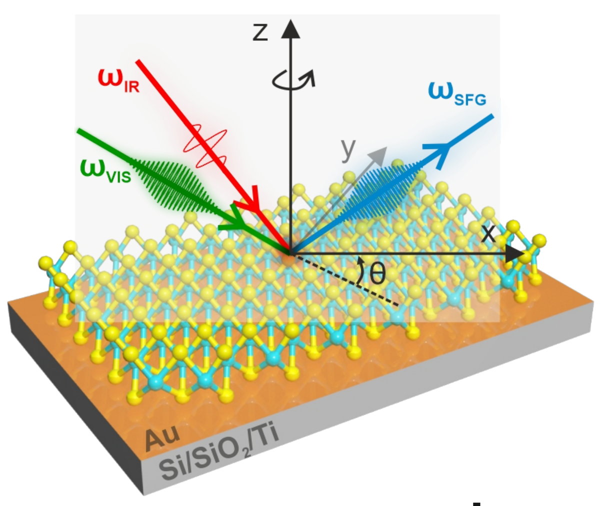
Optical images of exfoliated MoS_2/Au and CVD-grown MoS_2/SiO_2



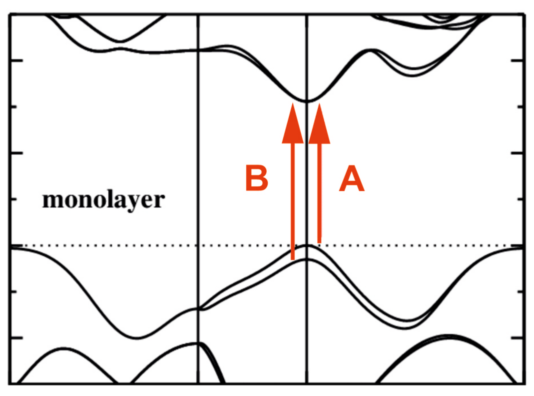
- Large flakes of monolayer MoS_2 can be obtained by both methods!

Method

Final state resonant sum frequency spectroscopy

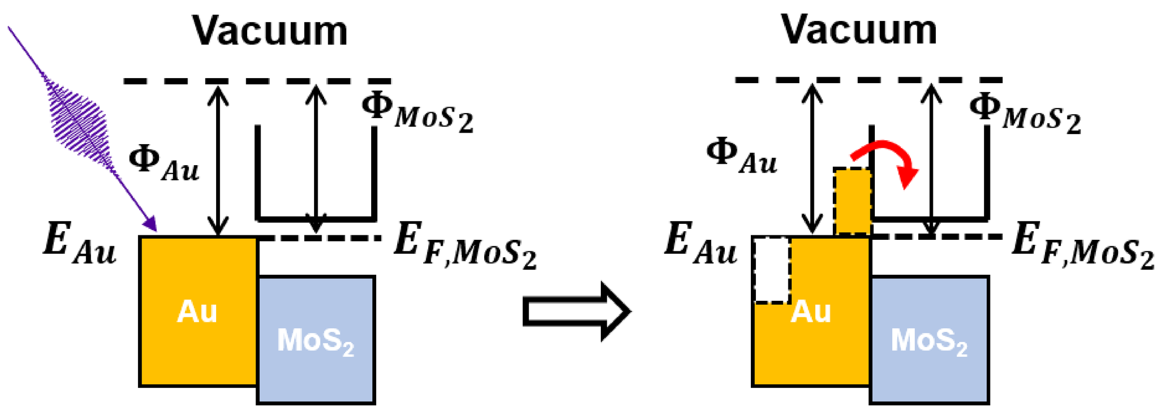


$\omega_{SF} = \omega_{VIS} + \omega_{IR}$



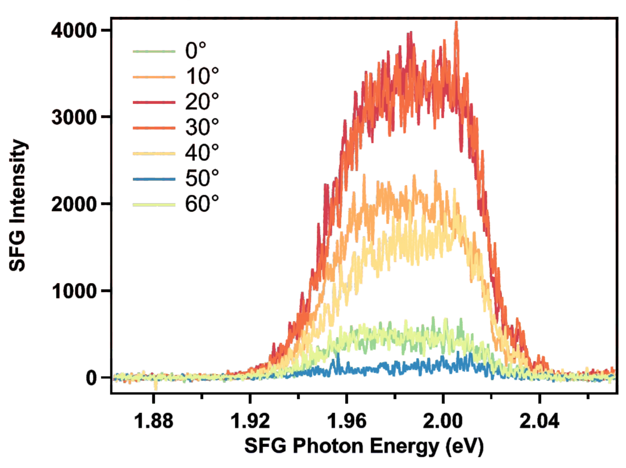
Band structure of monolayer MoS_2

Pump Au (hot electron) probe MoS_2

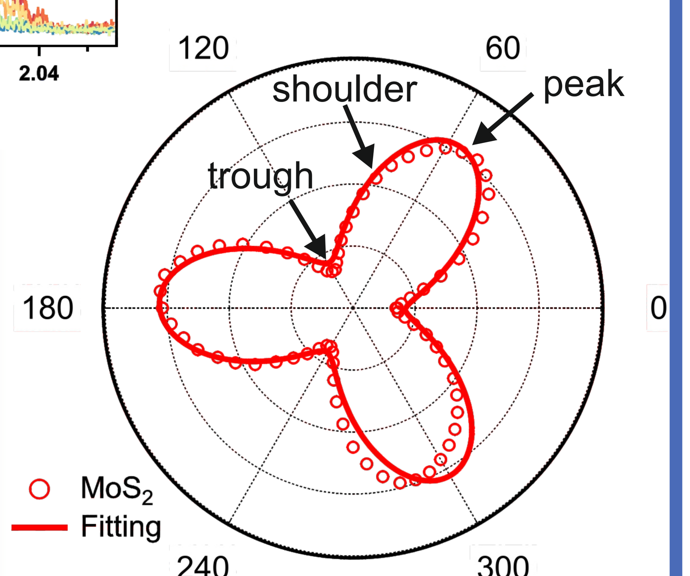


SFG spectrum

SFG spectra at different rotation angles



- The SFG intensities depend strongly on the rotation angles!

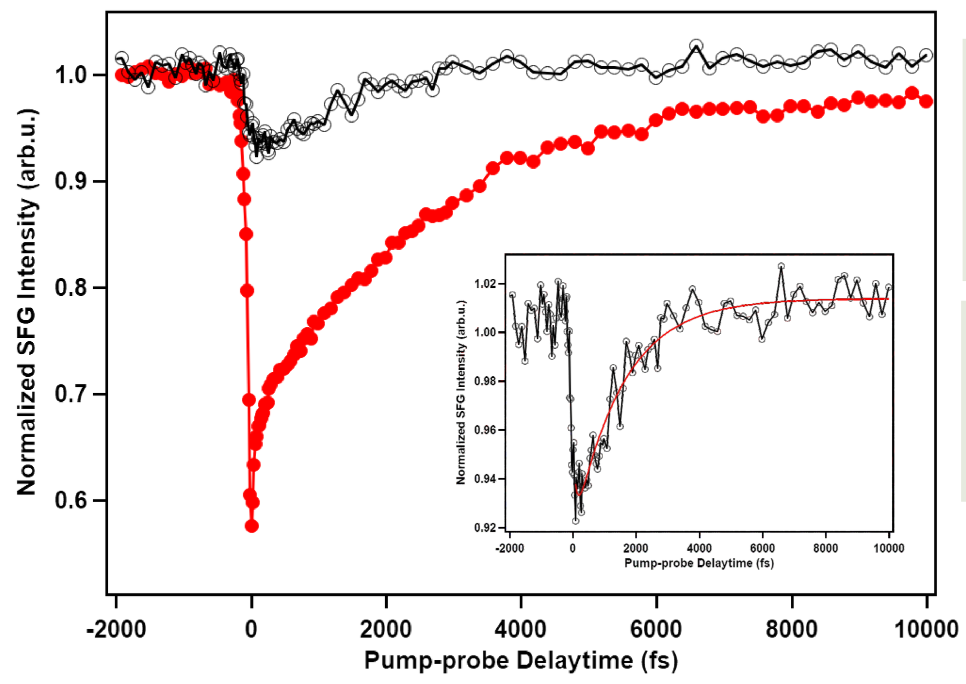


- A three-fold symmetry was first observed for MoS_2/Au compared to the well-known six-fold symmetry for MoS_2/SiO_2.

References

[1] H. Wang, et al. *Nano Lett.*, 2015, 15, 339–345.

MoS_2/Au and MoS_2/SiO_2 charge transfer dynamic



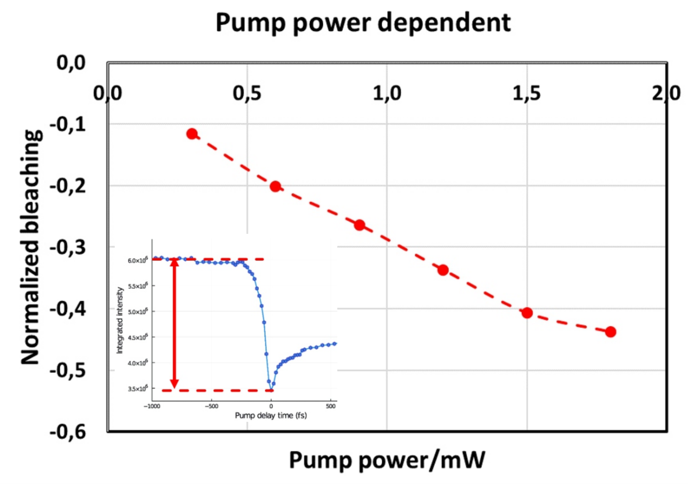
MoS_2/Au fits with tri-exponential

τ1_MoS_2 = 50 ± 5 fs
τ2_MoS_2 = 2604 ± 53 fs
τ3_MoS_2 = >5e5 fs *

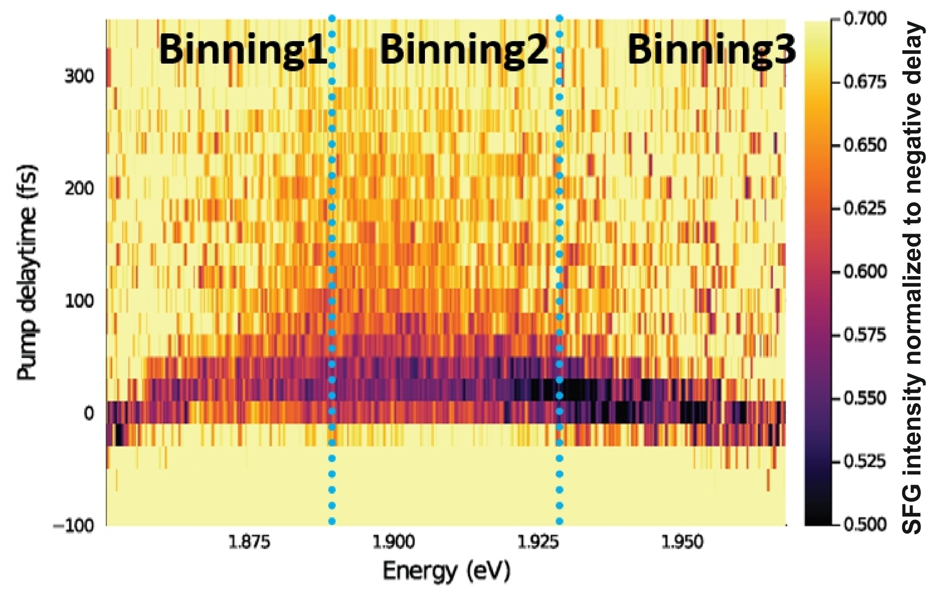
Pure Au fits with bi-exponential

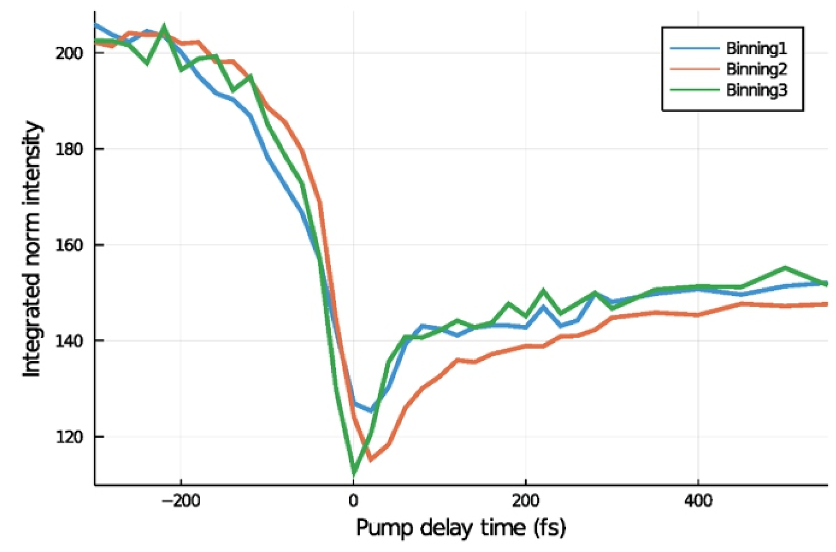
τ1_Au = 100 ± 0 fs*
τ2 _Au = 1423 ± 133 fs

Pulse FWHM: ~50fs

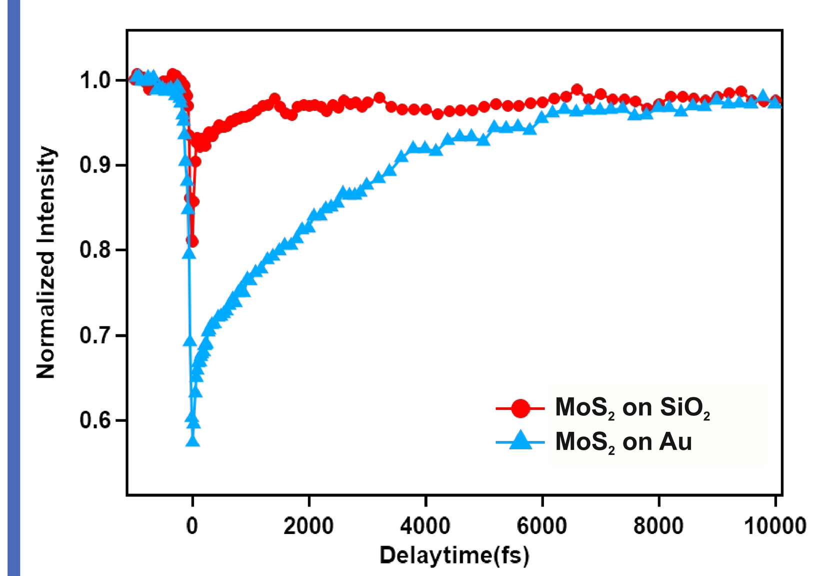


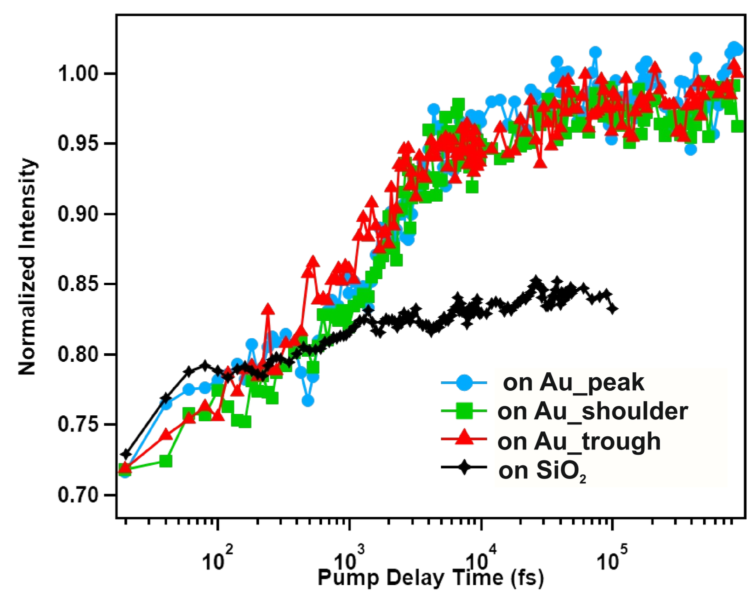
One-photon process!





Recovery time at position where is on resonance is longer than that of *off-resonance* position



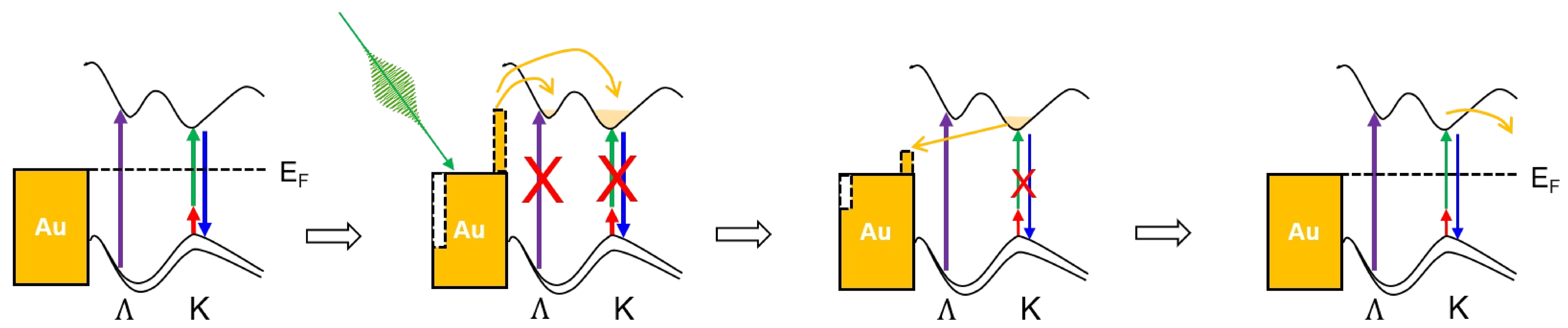


Dynamics of MoS_2/SiO_2:

- Smaller bleaching
- Similar process of fast decay and slow decay

Pump power-dependent measurements need to be done on MoS_2/SiO_2

Tentative interpretation



Before pump ⇨ **At time zero: state blocking** ⇨ **Electron transfer back to gold** ⇨ **Electron phonon scattering**

Conclusions

- Hot electrons from the gold substrate can easily tunnel to the MoS_2, resulting in a state-blocking for exciton formation.
- The intravalley relaxation of the hot carrier takes place on a time scale of 2.6 ps after a back donation of the hot electron to the gold substrate which takes place on a time scale of 50 fs.
- The slow dynamics (>100 ps) could be due to the intervalley scattering.