

Ultrafast Nanoscience Productions
presents

Starring:
Photon Exciton Pentacene

The tale of an exciton

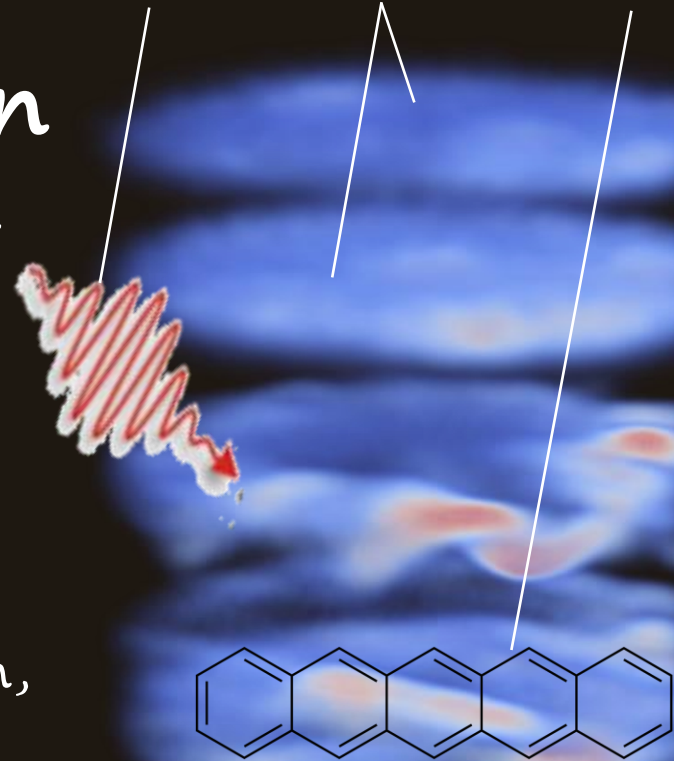
Eine Anregung. Eine Wechselwirkung.
Alles kann passieren ...

Drehbuch:

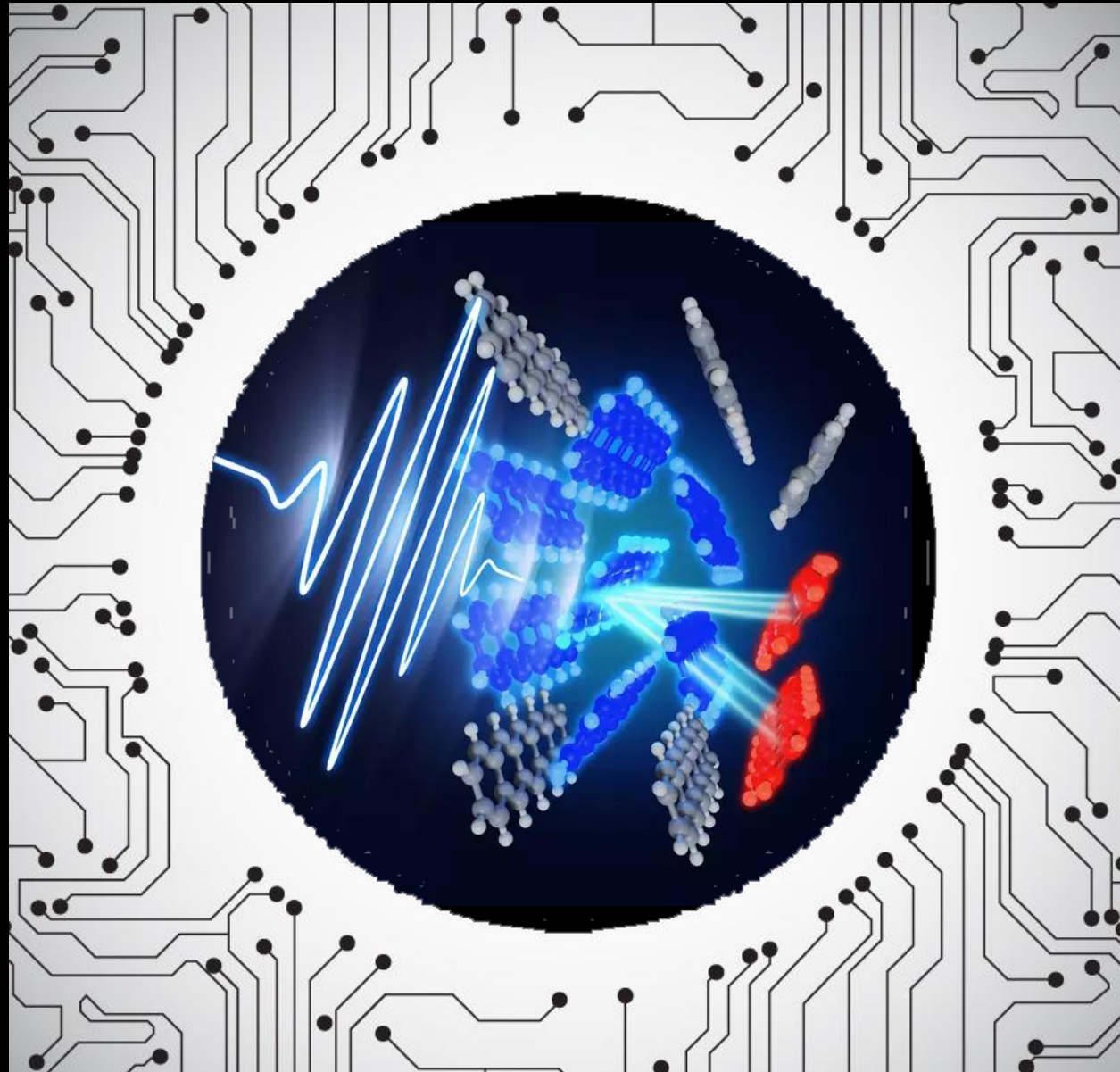
A. Neef & R. Ernstorfer

(Elektronen-)Kamera:

A. Neef, S. Beaulieu, S. Hammer, S.
Dong, J. Maklar, T. Pincelli, R.P. Xian,
M. Wolf, L. Rettig, J. Pflaum &
R. Ernstorfer

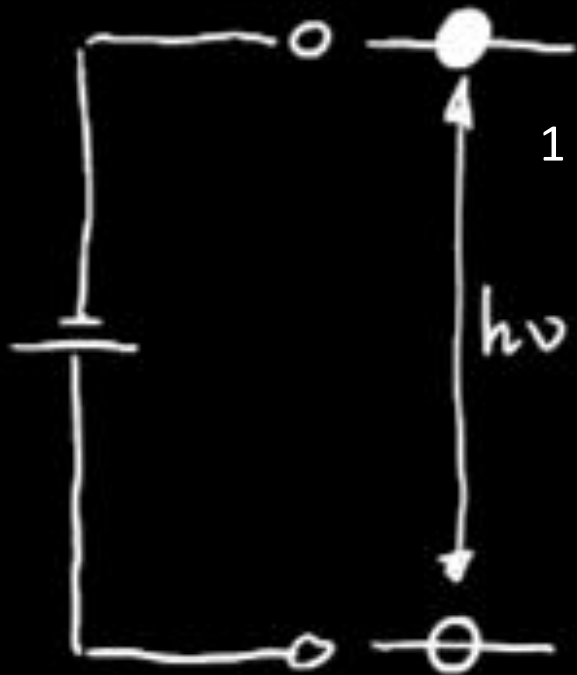


Ultrafast Nanoscience – goal: watch materials in action*!



***the fundamental processes governing device functionality**

Power from the sun



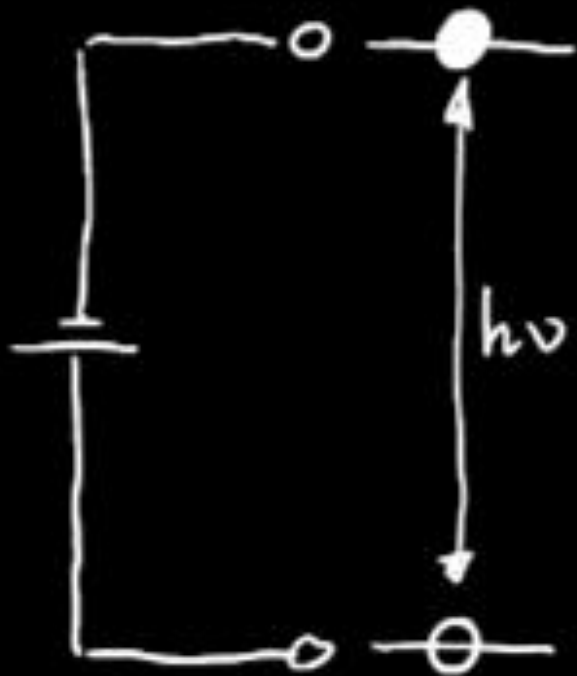
1 photon \rightarrow 1 electron

For Si
cells

$$\eta_{\max} = 29\%$$

Power from the sun

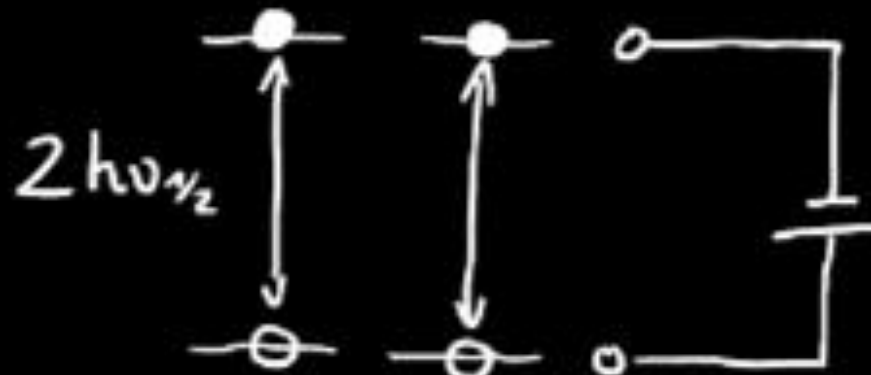
Does this actually work?
Let's look at the life cycle ...



For Si cells

$$\eta_{\max} = 29\%$$

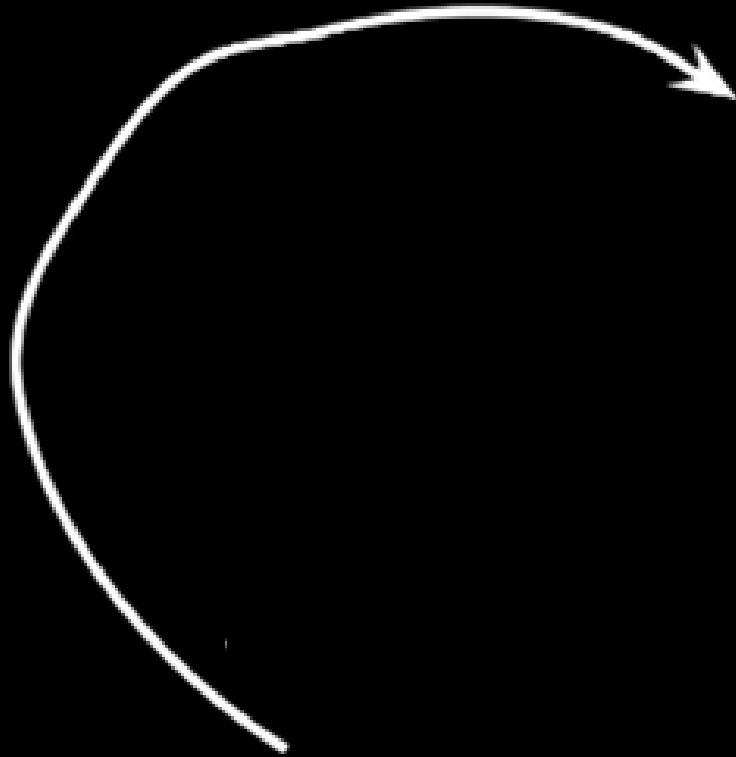
1 photon \rightarrow 2 electrons!!



$$\eta_{\max} = 39\%$$

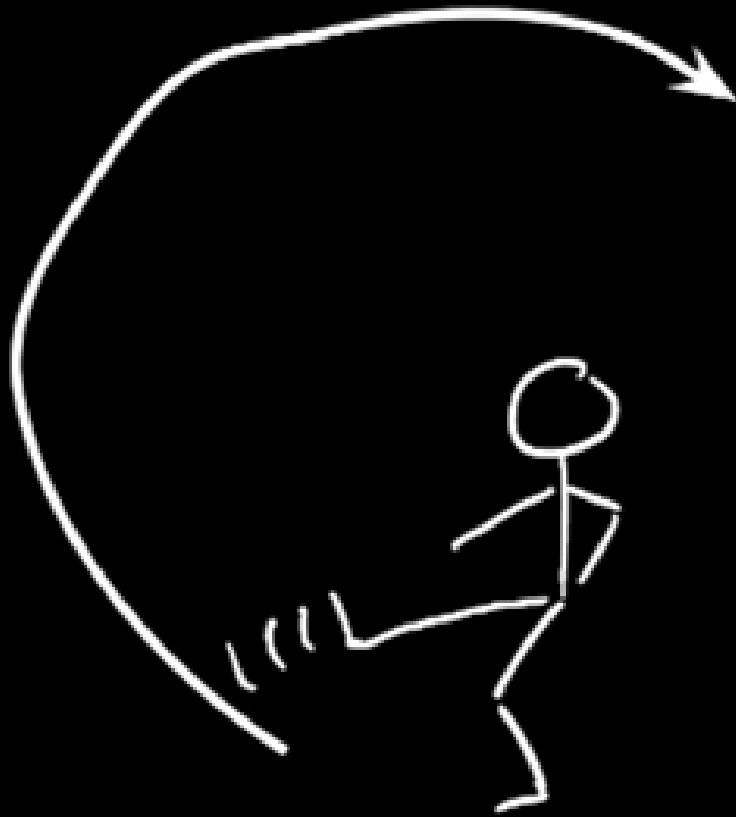
Peter Würfel,
Physics of
Solar Cells

The human life cycle



HOME
=lowest-energy state

The human life cycle



HOME
=lowest-energy state

The human life cycle



exciting single
state



HOME
=lowest-energy state



The human life cycle



exciting single state



HOME
=lowest-energy state



correlated paired state



uncorrelated state



lowest-energy state

The **exciton** life cycle in pentacene

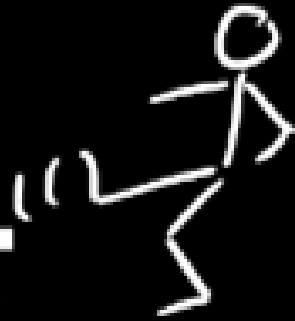


excited singlet state

LUMO, Singlet



HOMO



identical!!!

What is the **mechanism** of this process?



correlated (2x) triplet state

uncorrelated triplets



ground state

Time- and angle-resolved photoemission spectroscopy (trARPES)



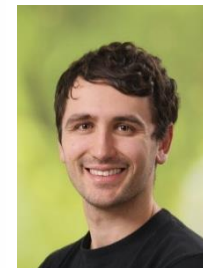
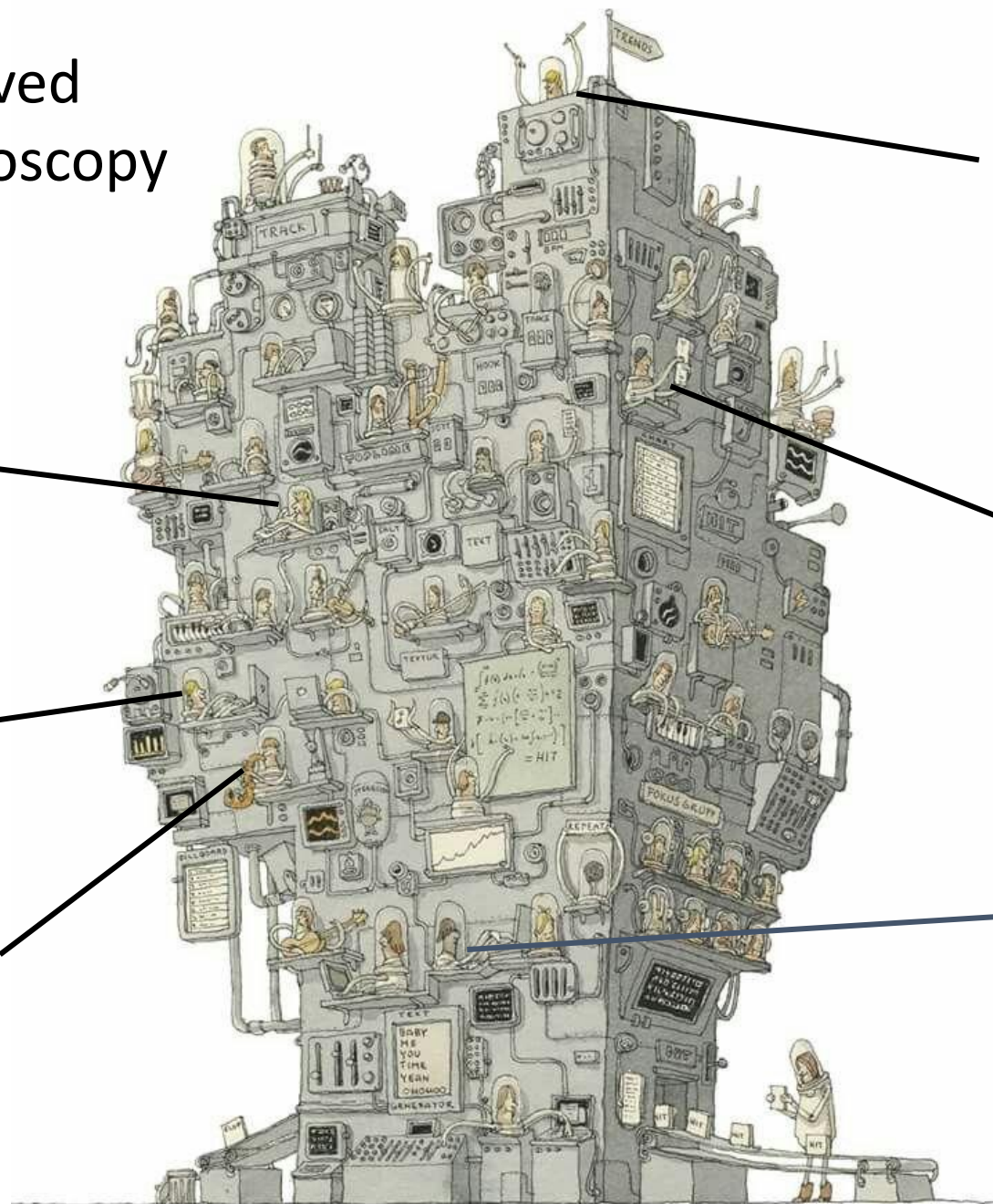
Shuo Dong



Patrick Xian



Sam Beaulieu



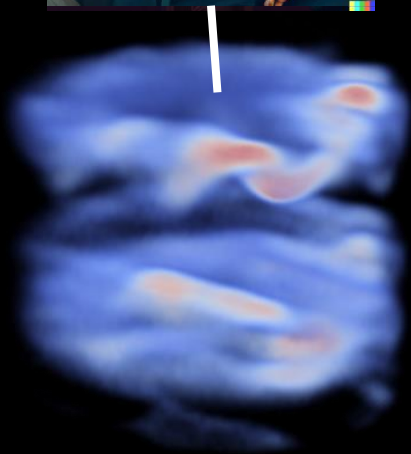
Alex Neef

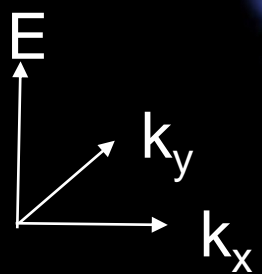
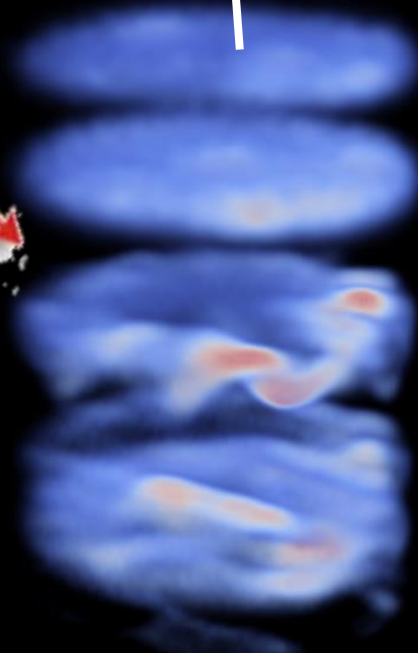
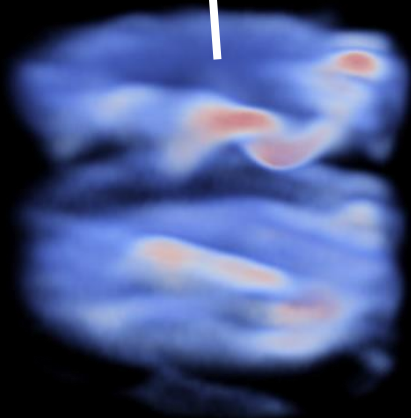
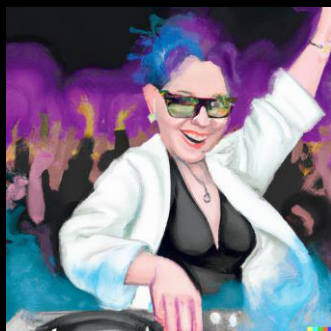


Tommaso Pincelli



Laurenz Rettig



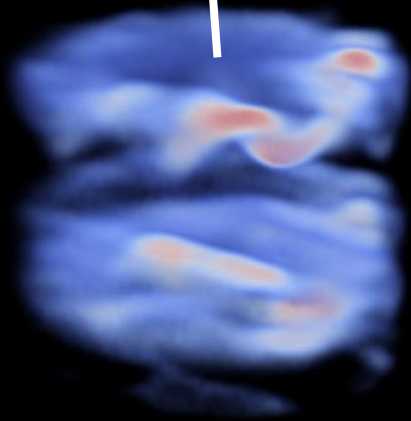
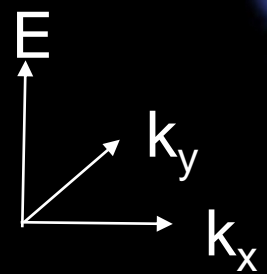


ground state

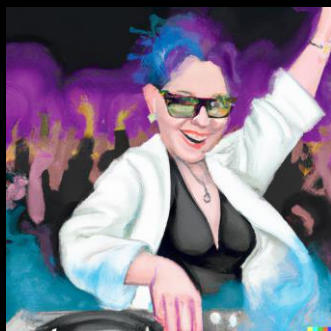
excited singlet state

0 fs

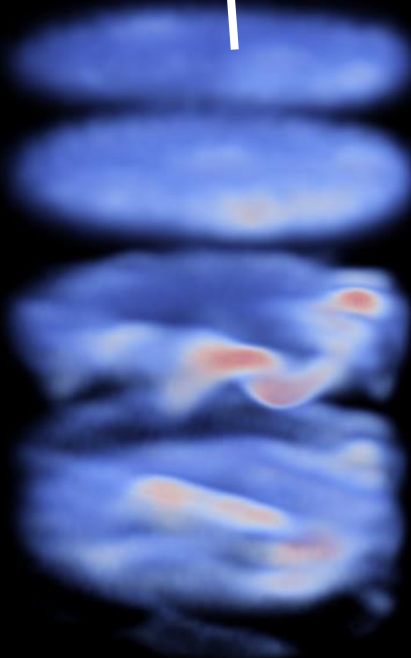
time



ground state



S
X



excited singlet state

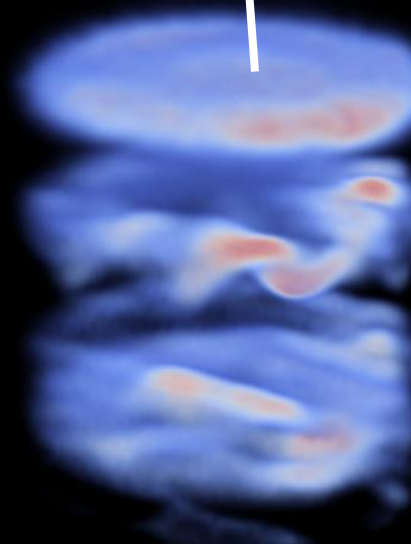
0 fs

?!?

A large white double-headed arrow pointing left and right, with the text '?!?' above it, indicating a transition or comparison between the states.



T



correlated (2x) triplet state

500 fs

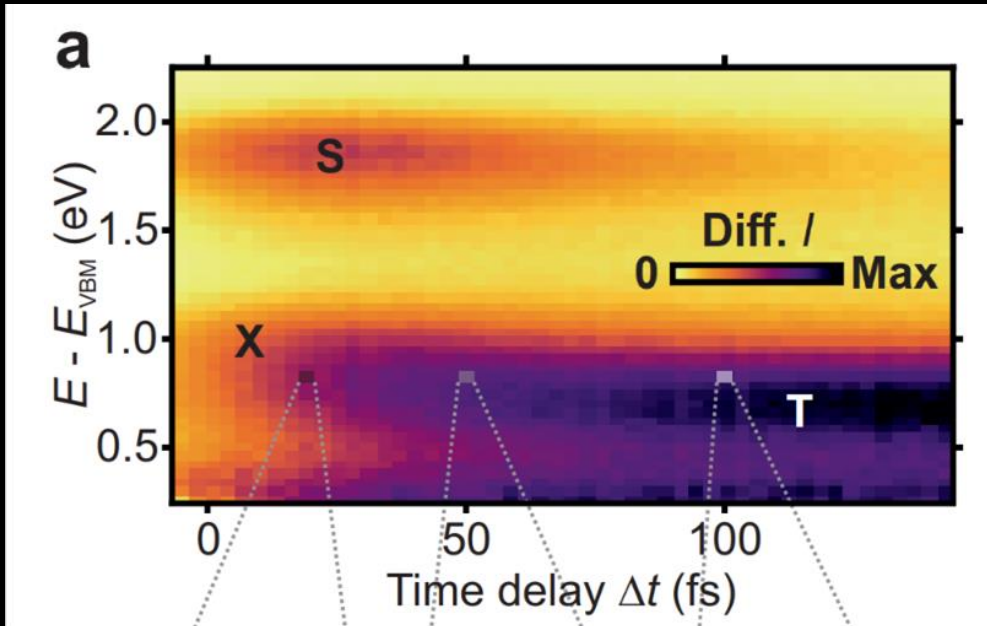
time

A horizontal white arrow pointing to the right, labeled 'time' at the end, indicating the progression of time from left to right.

The signal at the triplet energy rises simultaneously with the singlet signal!

First observed by Chan et al., Science 334, 1541 (2011) from Columbia University.

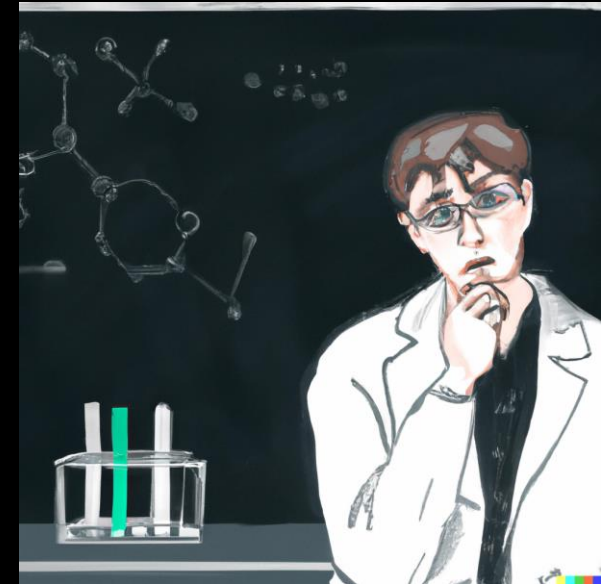
→ “**Manhattan Model**”: $X = T$
coupling between singlet and triplets is so strong that singlet fission occurs immediately!



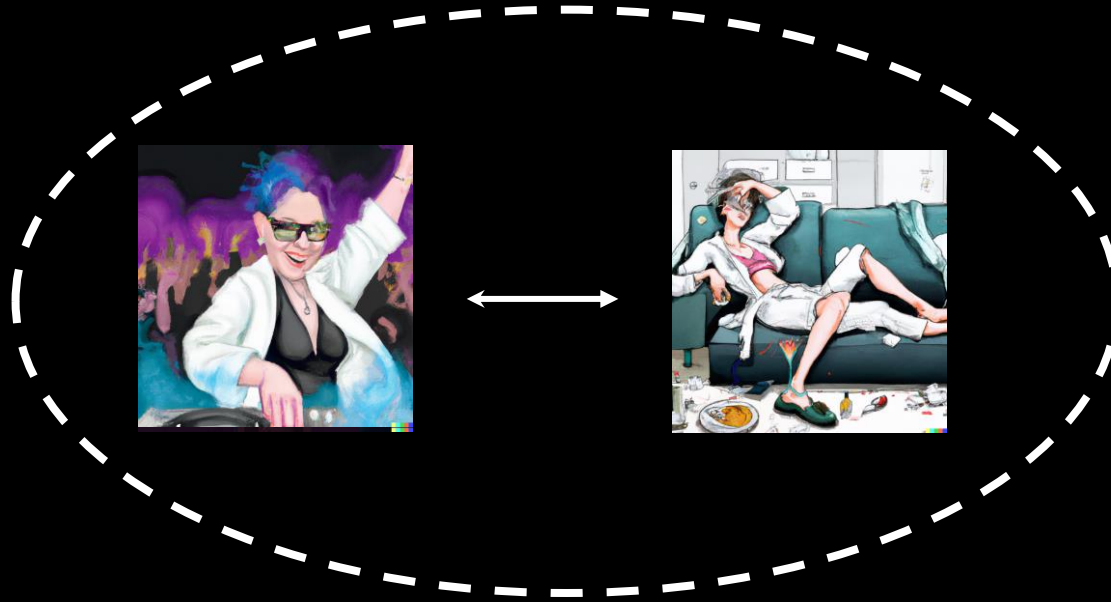
Manhattan model



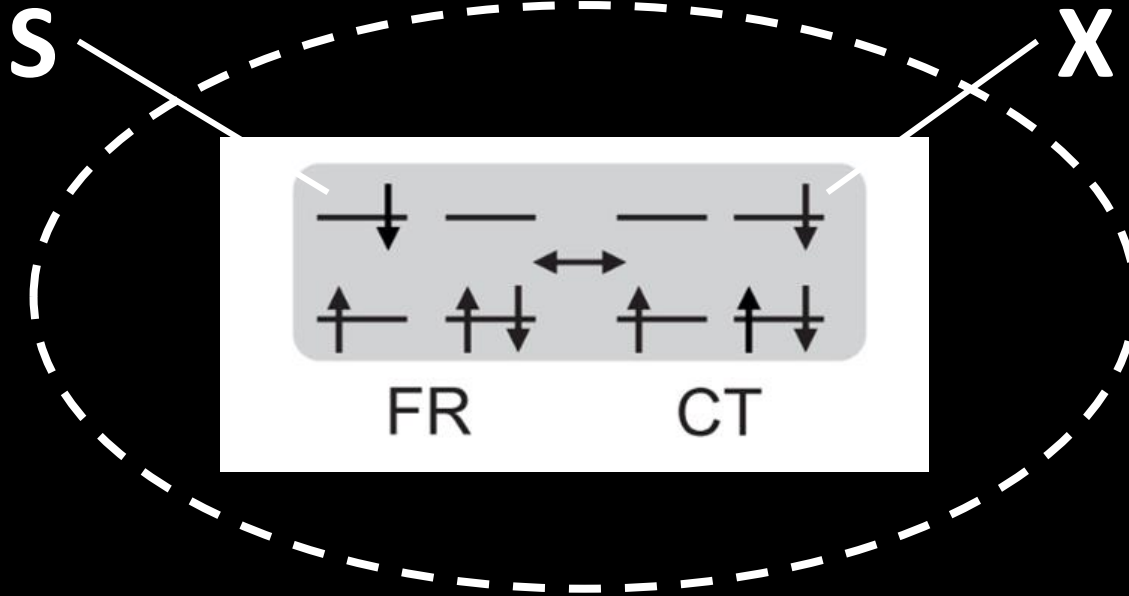
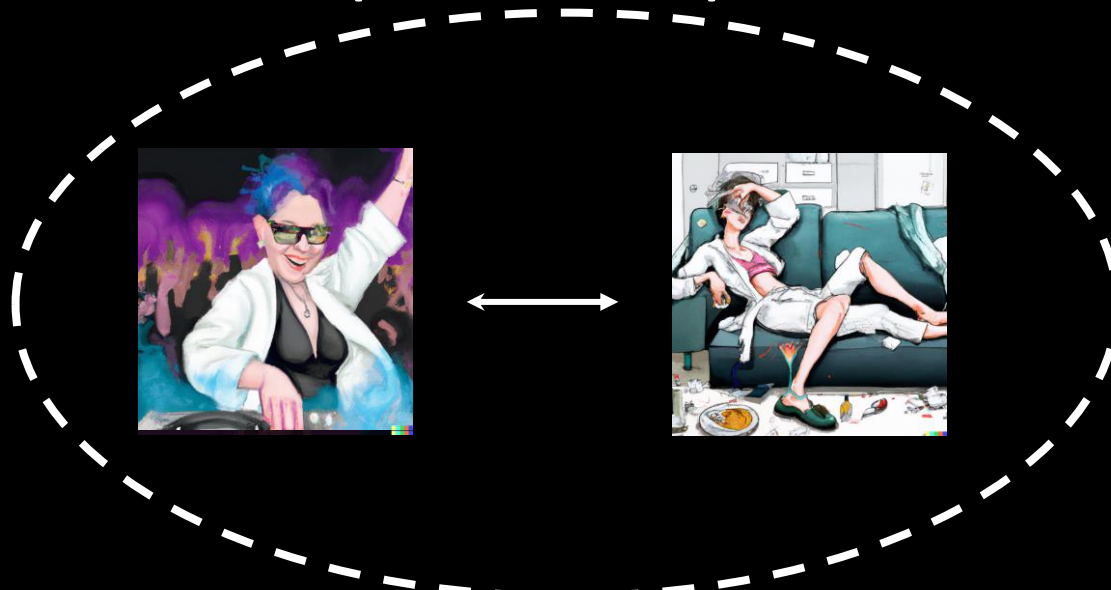
Manhattan model

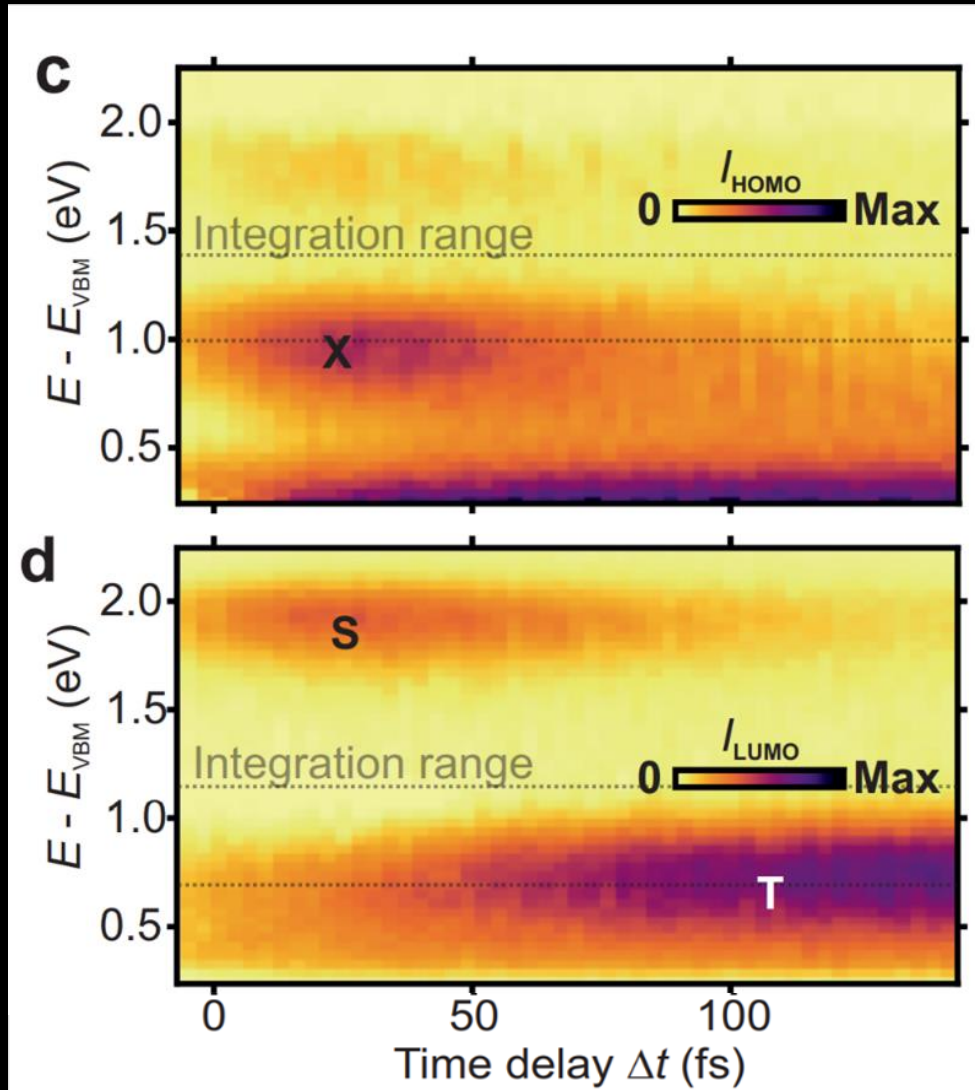


The clubbing-hangover model: $X=S$ (Berlin model)



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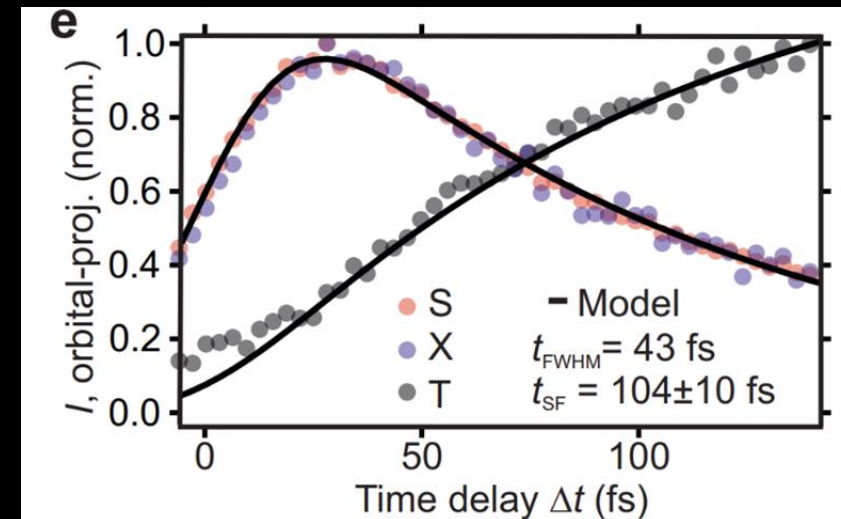


Utilizing the momentum-space distribution, we can decompose the signal into HOMO-like and LUMO-like orbitals.

X has the same dynamics as S!

T rises with the decay of the singlet!

→ Berlin model rocks, Manhattan model sucks!



original publication: [Neef et al., Nature 616, 275 \(2023\)](#)