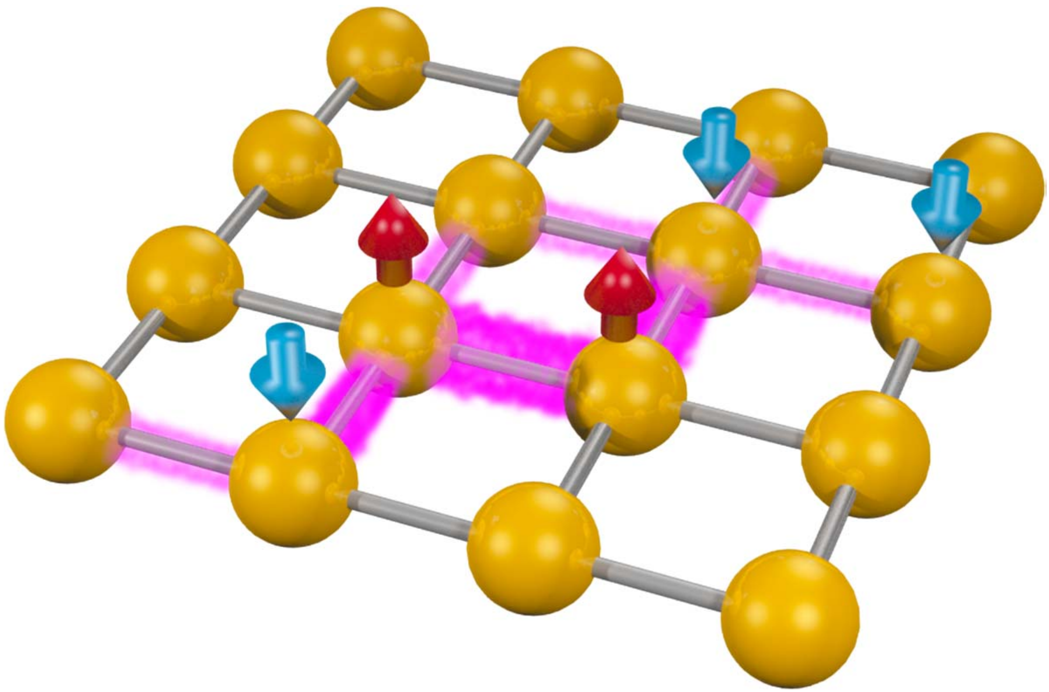
Universität Stuttgart
Institute for Functional Matter
and Quantum Technologies

Atomic-scale dynamics of collective charge and spin excitations

Sebastian
Loth

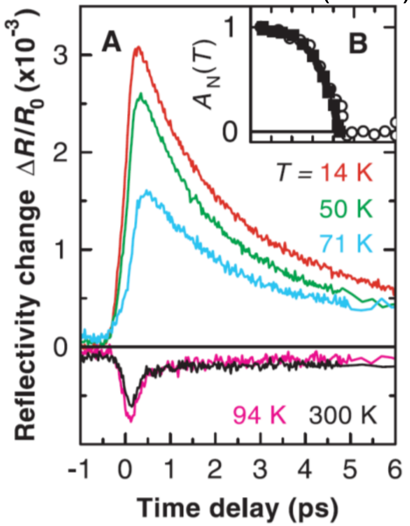
Emergence of collective phases
from interaction of electrons, spin, phonons



Correlated materials: Ultrafast dynamics

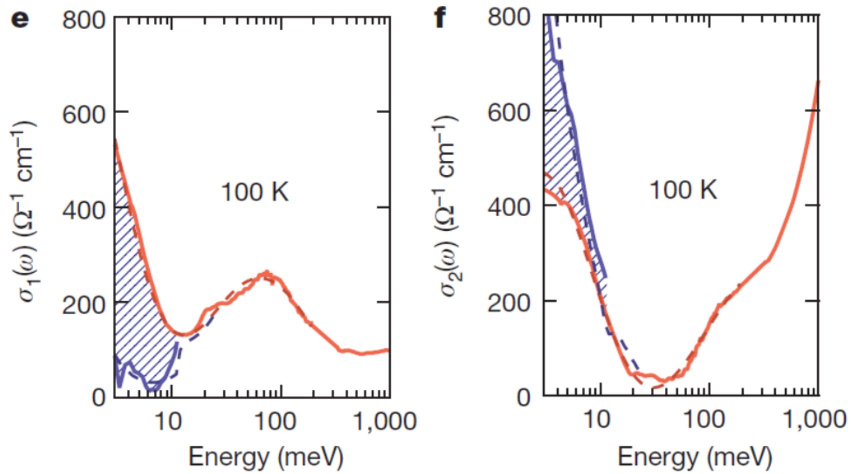
Indiscriminate pump
„Optical Heating“

Kaindl et al. 287 470 (2000)

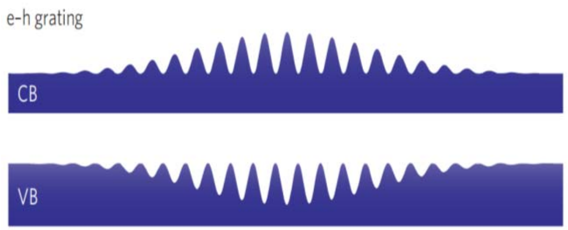


Resonant / coherent pump
Light-induced states

Mitrano et al. Nature 530 461 (2016)

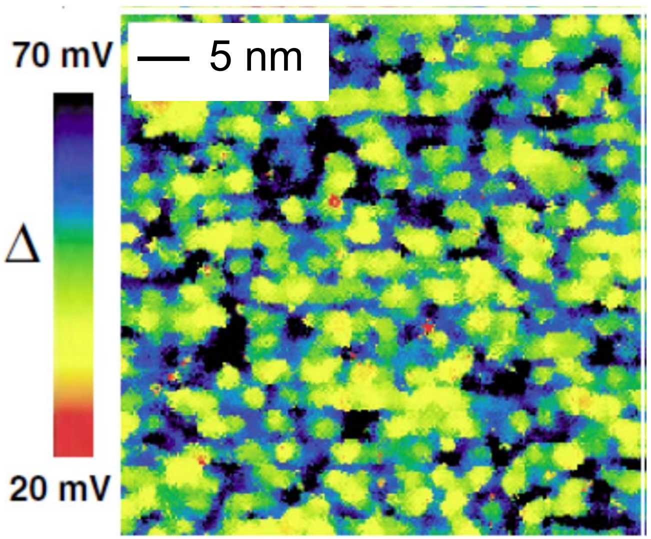


Transient grating pump
spatial diffusion

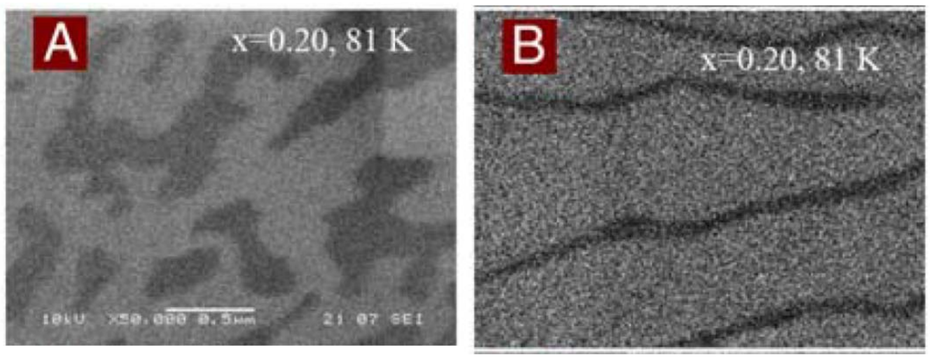


Yang et al. Nat.Phys. 8 153 (2012)

Correlated materials: Spatial heterogeneity



McElroy et al. PRL 94 197005 (2005)
also: Lang et al. Nature 415 412 (2002)
also: Parker et al. Nature 468 677 (2010)



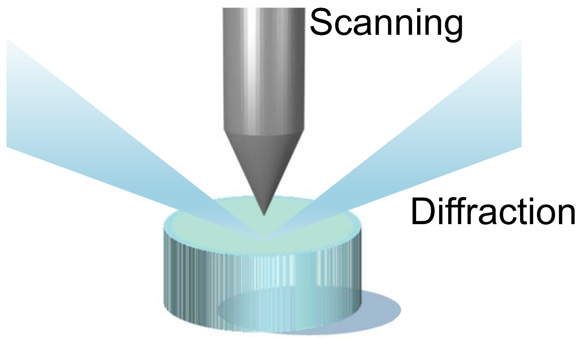
Kim et al. PNAS 107 5272 (2010)
also: Jones et al. Nano Lett. 10 1574 (2010)
also: Dagotto Science 309 257 (2005)

Order parameter fluctuations
e.g. gap disorder in SC, m,agnetization, …

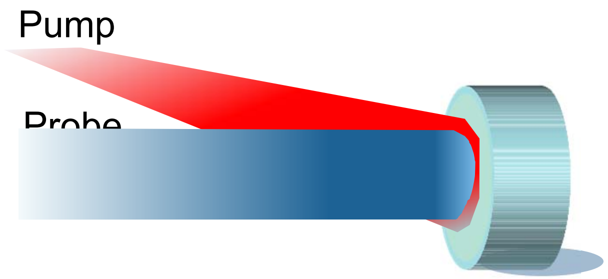
Electronic phase competition
e.g. metal & insulator, charge order & SC, …

Atomic-scale dynamics of many-body states

Spatial Probes

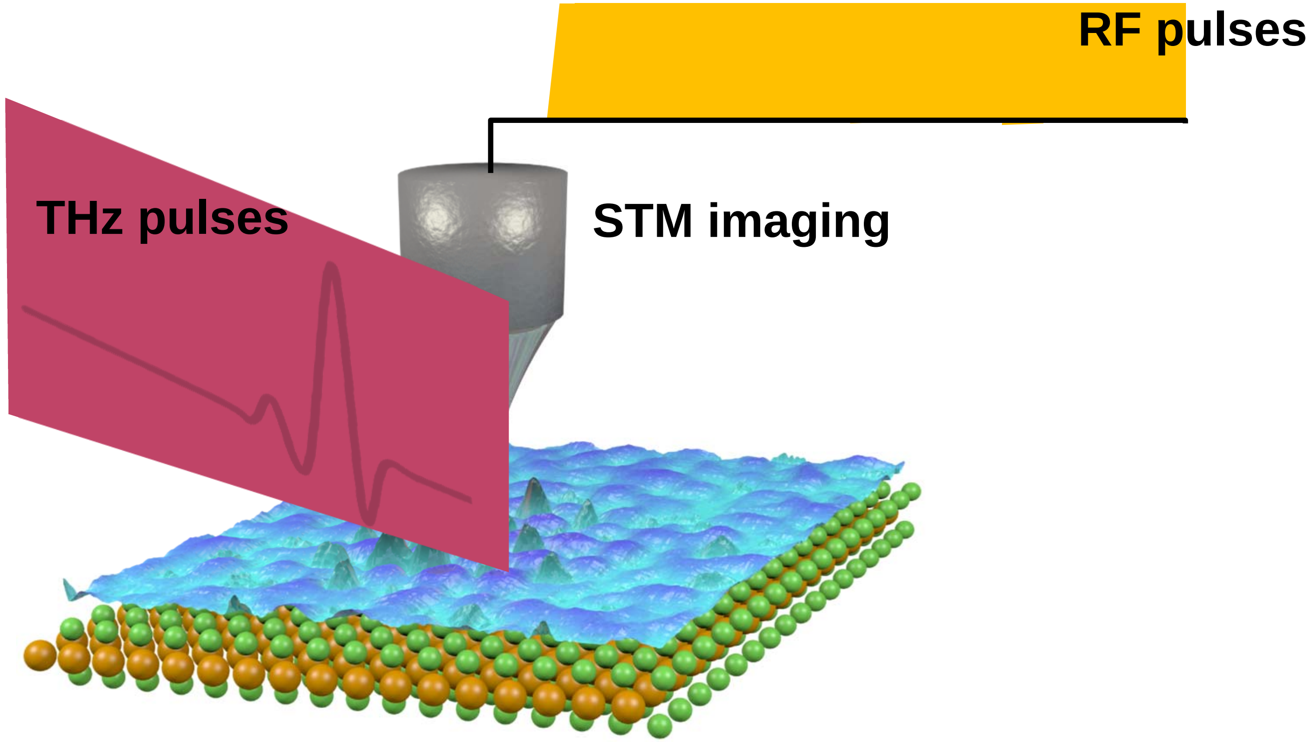


Ultrafast Probes



Timescale	Length scale	Timescale	Length scale
ms	**atomic**	**fs**	**μm**

Accessing atomic-scale dynamics in real space

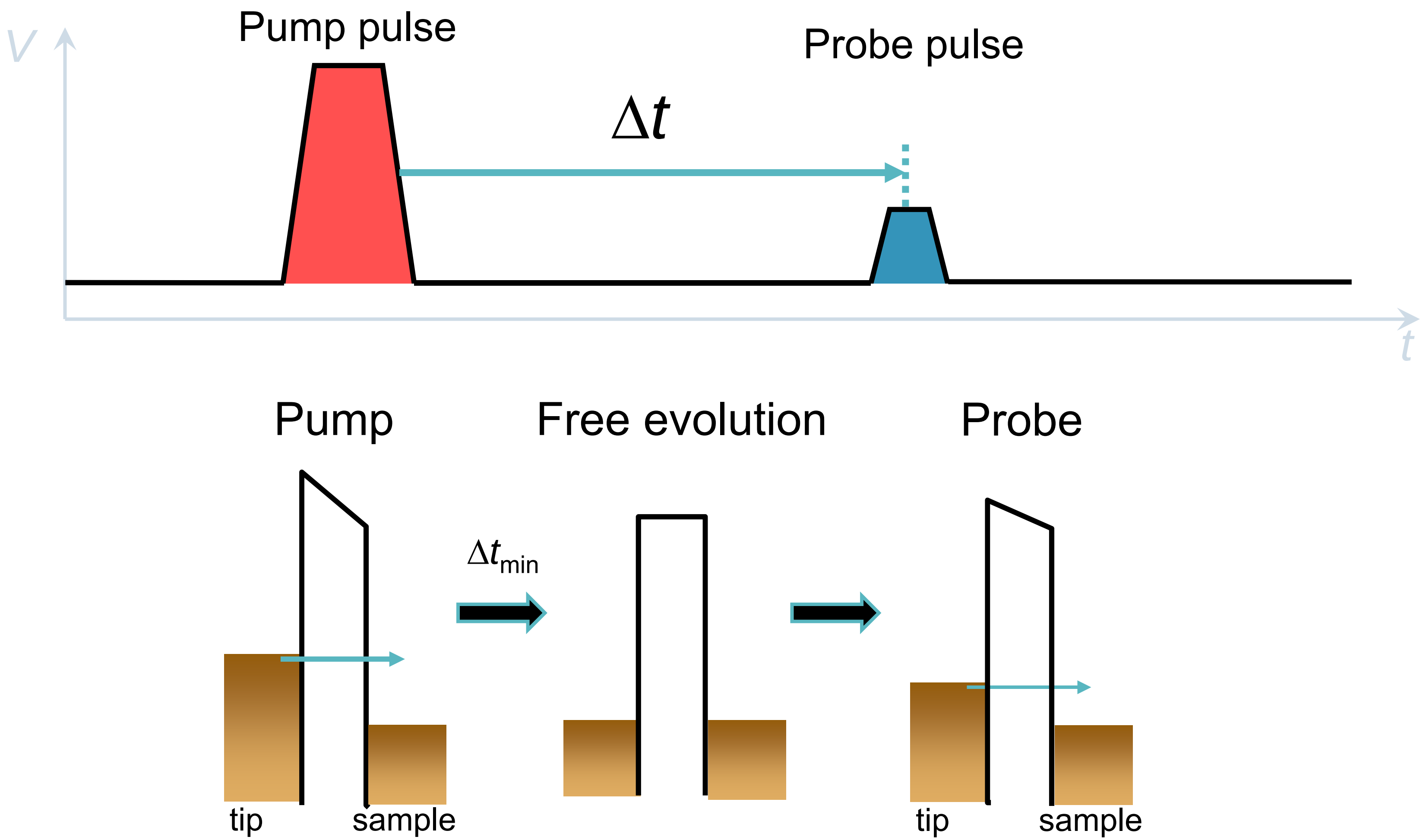


Timescale **ms**

Length scale **atomic** & Timescale **100 fs**

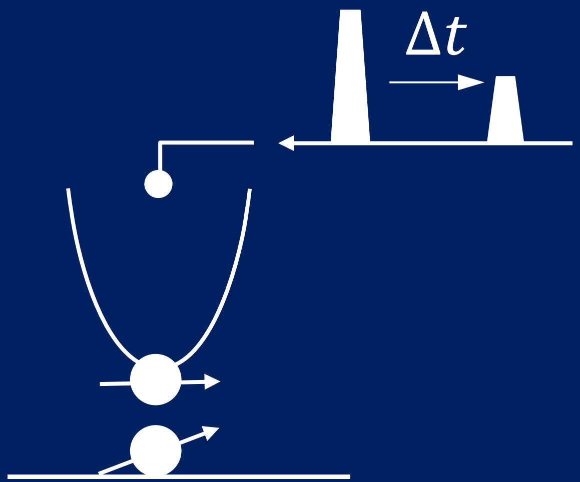
Length scale **µm**

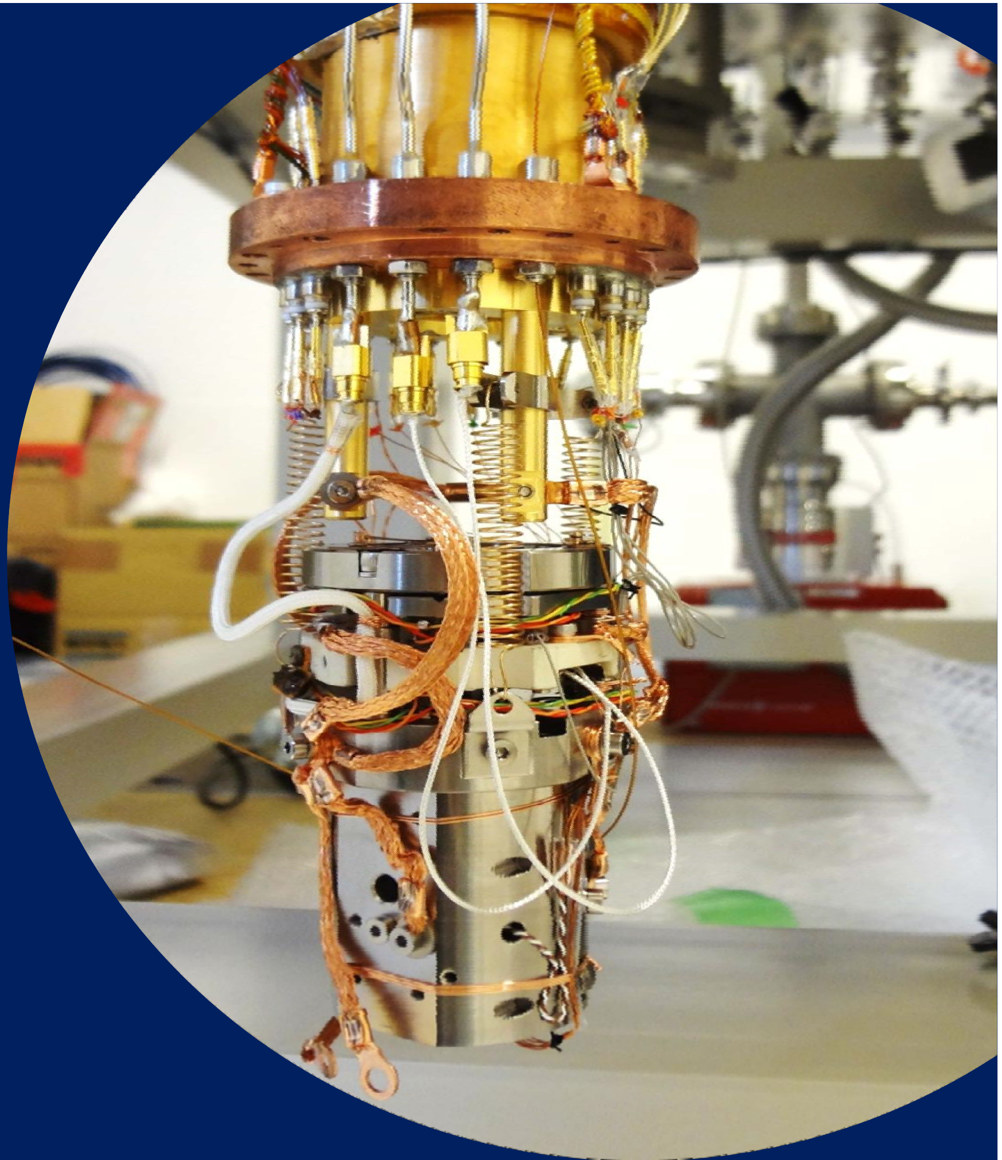
How fast can you be?



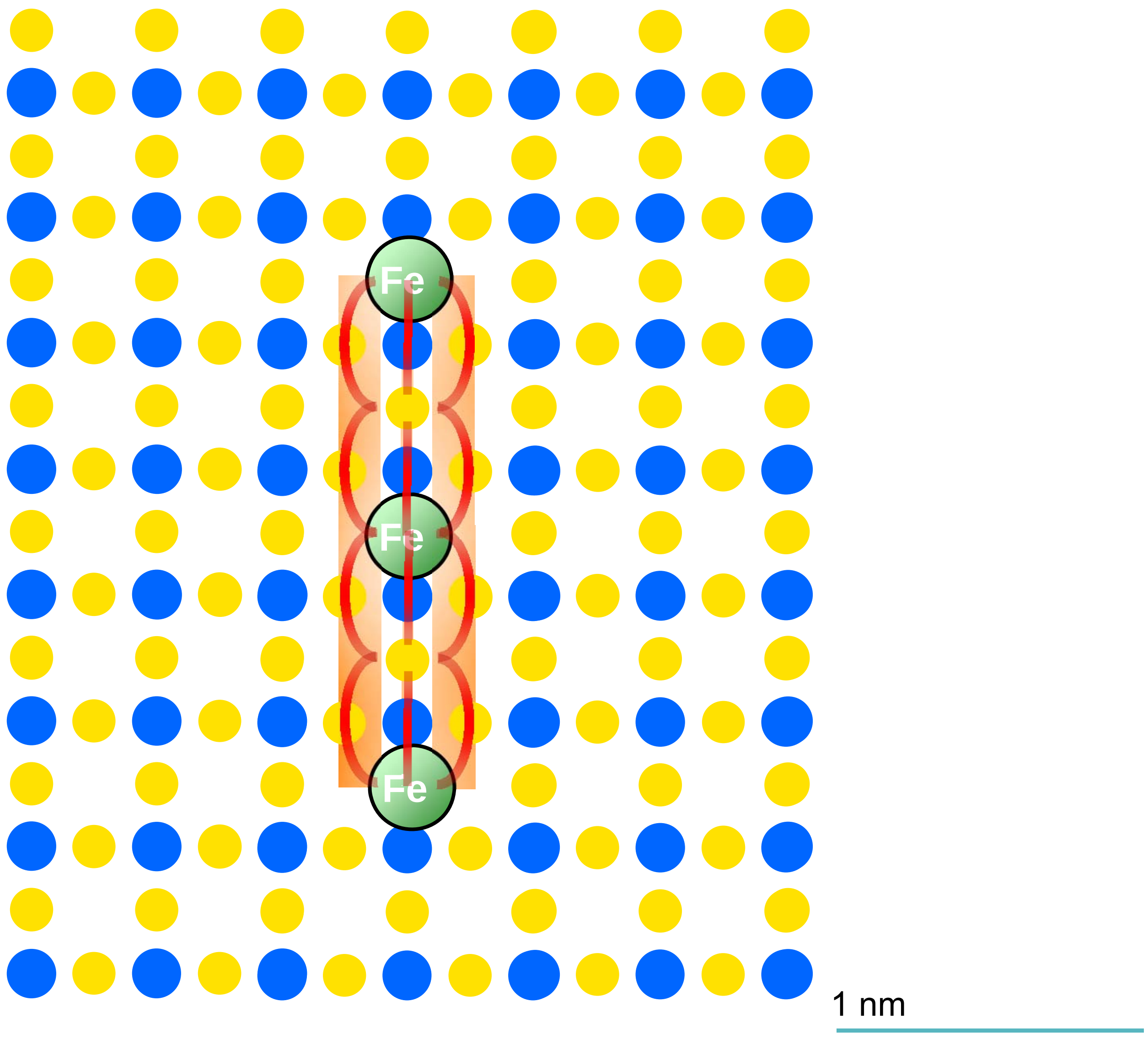
Speeds reached: t_{min}~ 50 ps (electronic) t_{min}~ 100 fs (optical)

Spin dynamics with atomic resolution

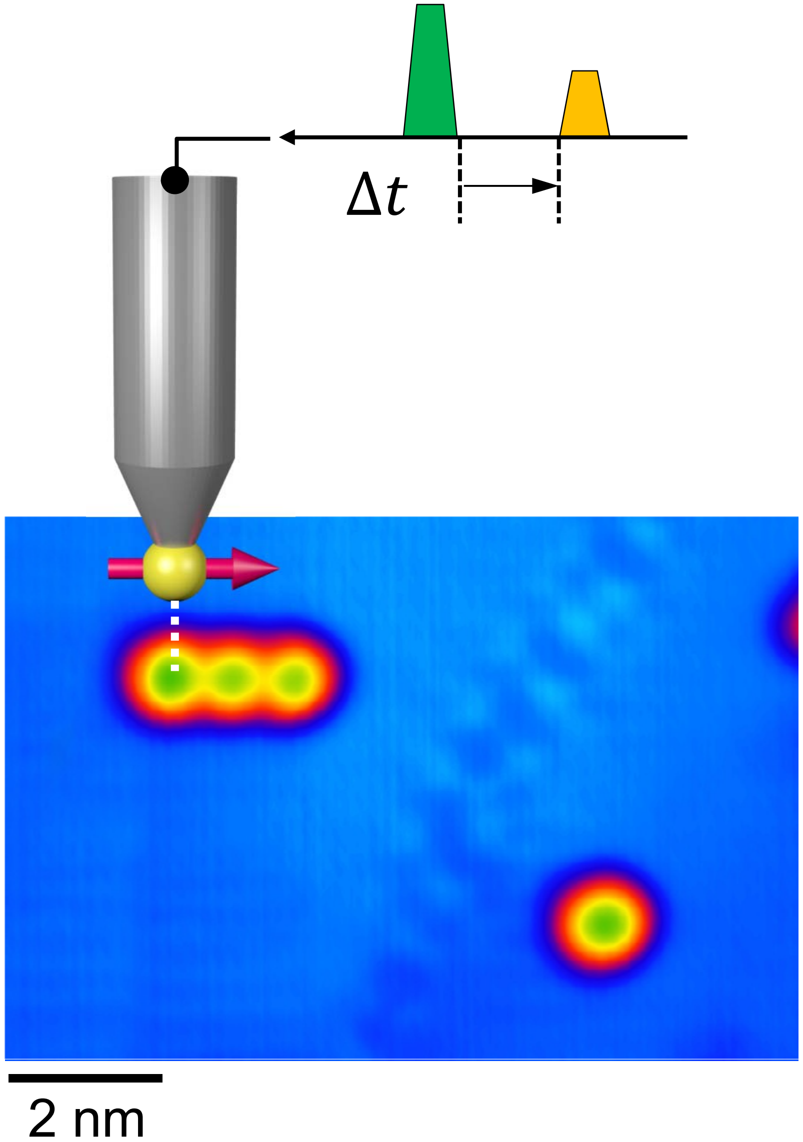


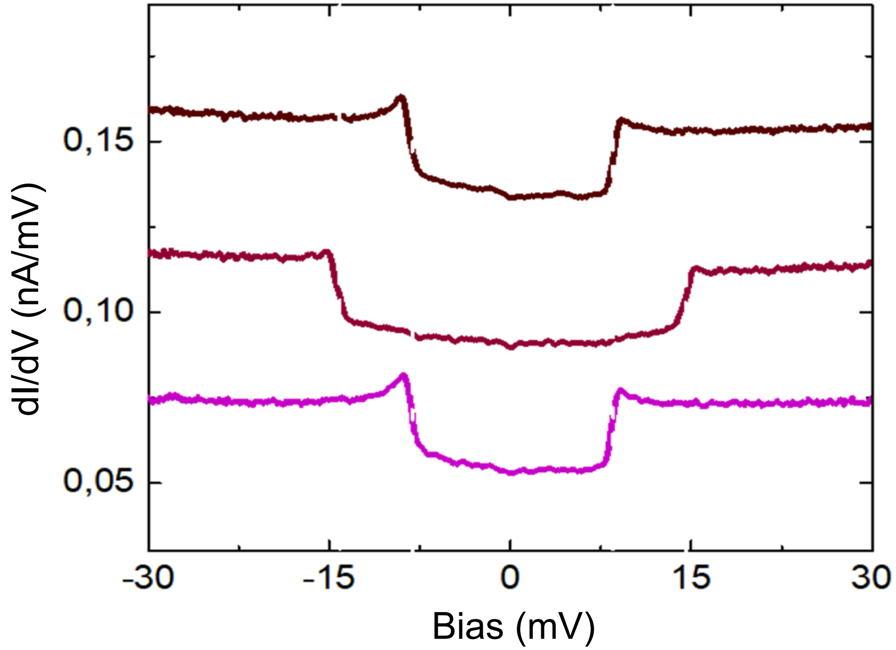


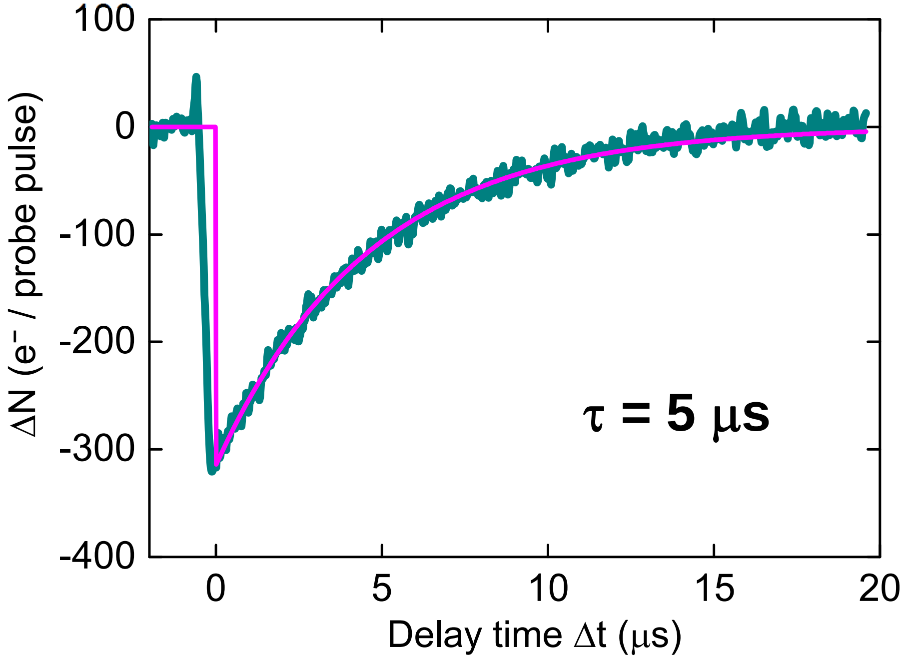
Fe_3 spins sensor



Few-atom spin systes $(Fe_3)_{Cu2N}$

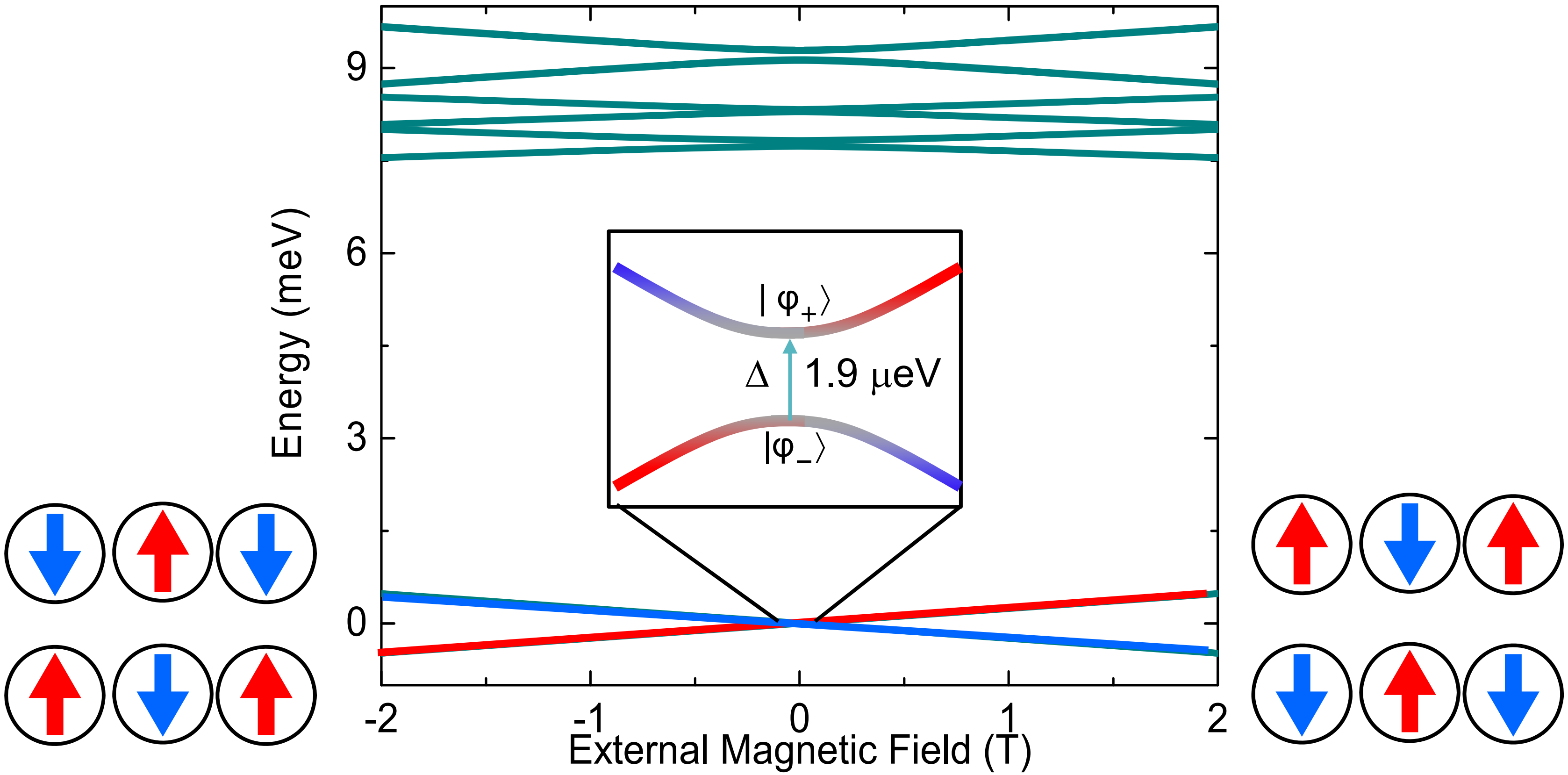






technique see: Science 329 1628 (2010).

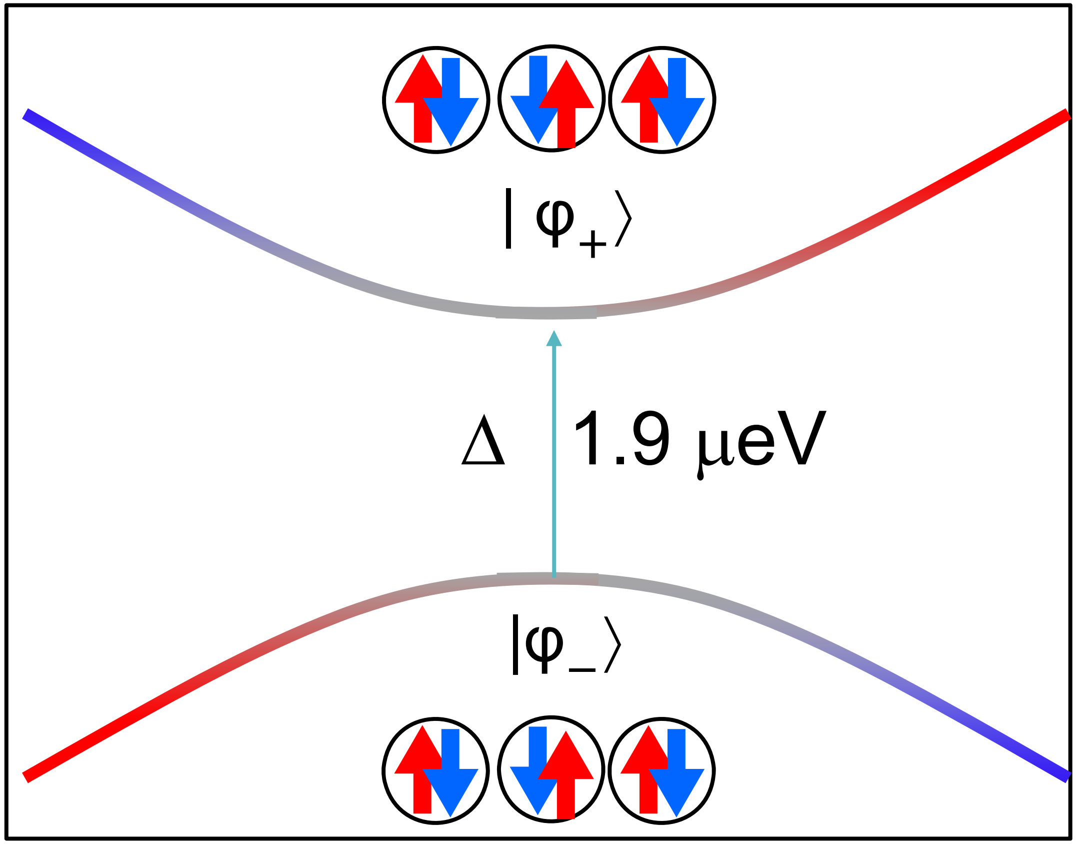
Spin state spectrum of Fe trimer



Entangled ground state and avoided level crossing at B = 0

$$|\varphi_+\rangle = |+2 - 2 + 2\rangle + |-2 + 2 - 2\rangle$$
$$|\varphi_-\rangle = |+2 - 2 + 2\rangle - |-2 + 2 - 2\rangle$$

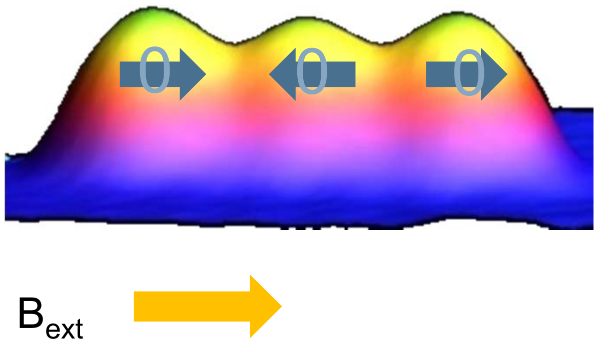
Entangled ground state and avoided level crossing at $B = 0$

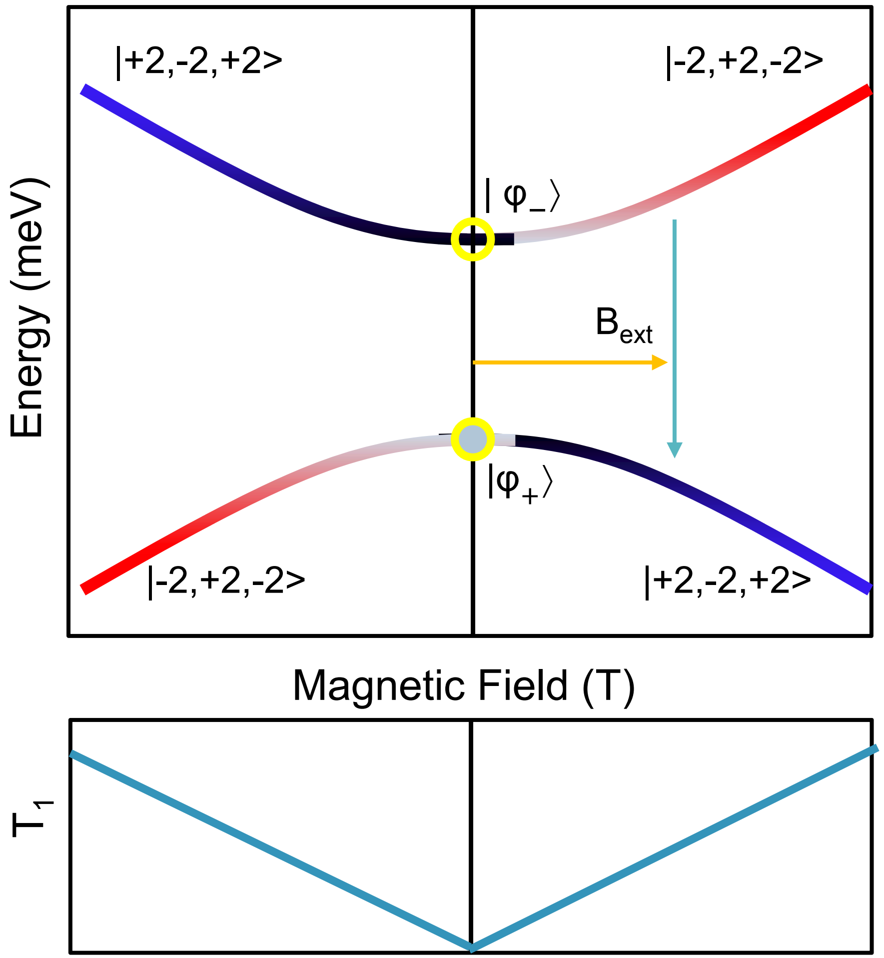


$$|\varphi_+\rangle = |+2-2+2\rangle + |-2+2-2\rangle$$

$$|\varphi_-\rangle = |+2-2+2\rangle - |-2+2-2\rangle$$

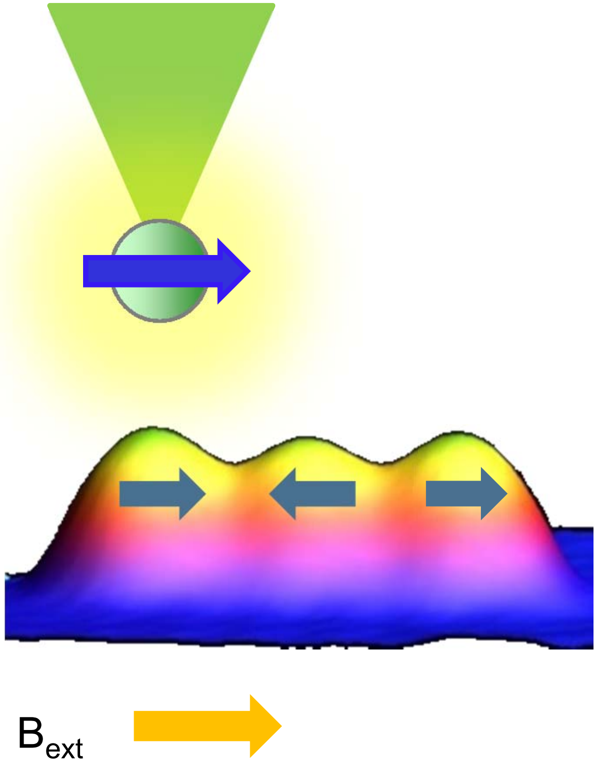
Magnetic spin-environment interaction

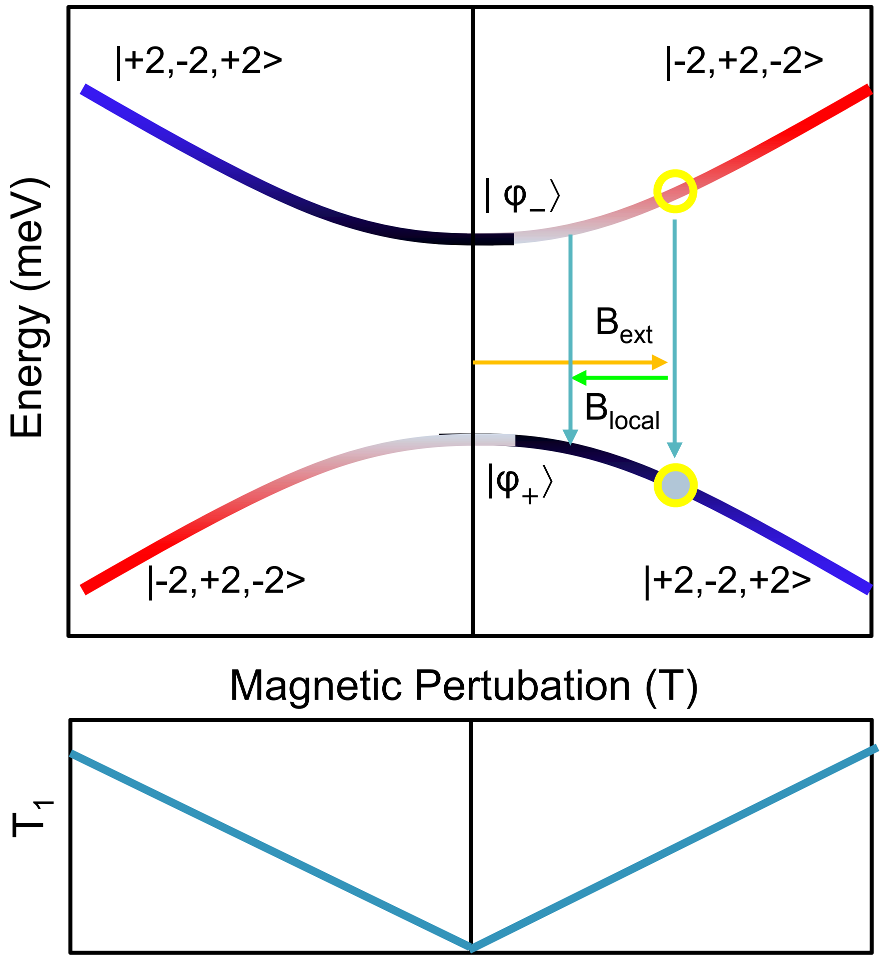




$$T1 \sim \alpha^* \Delta E$$

Magnetic spin-environment interaction

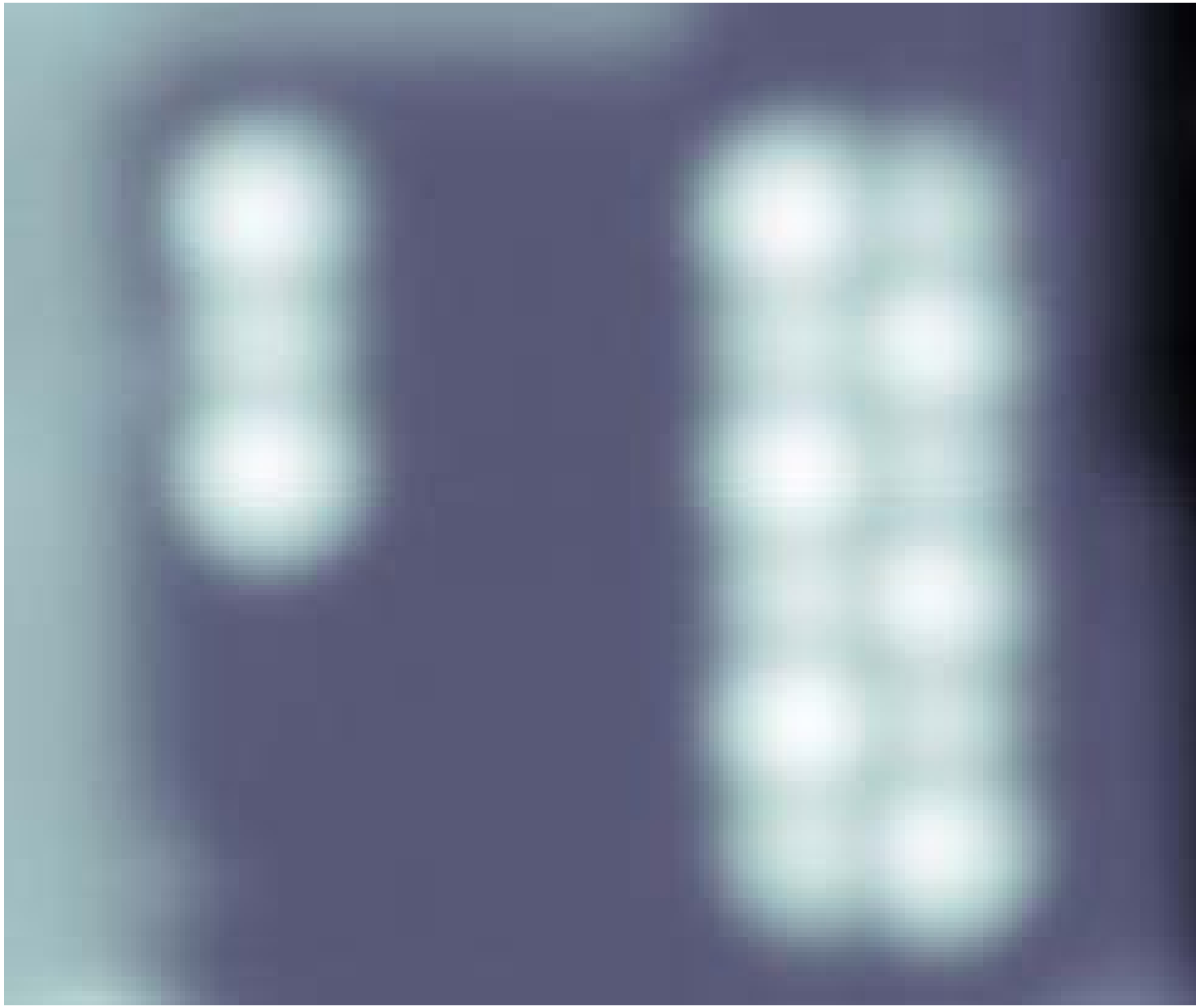




S. Yan, D.J. Choi, J.A.J Burgess, S. Rolf-Pissarczyk, S. Loth *Nature Nanotechnol.* 10 40 (2015).

Remote Spin sensing at the atomic scale

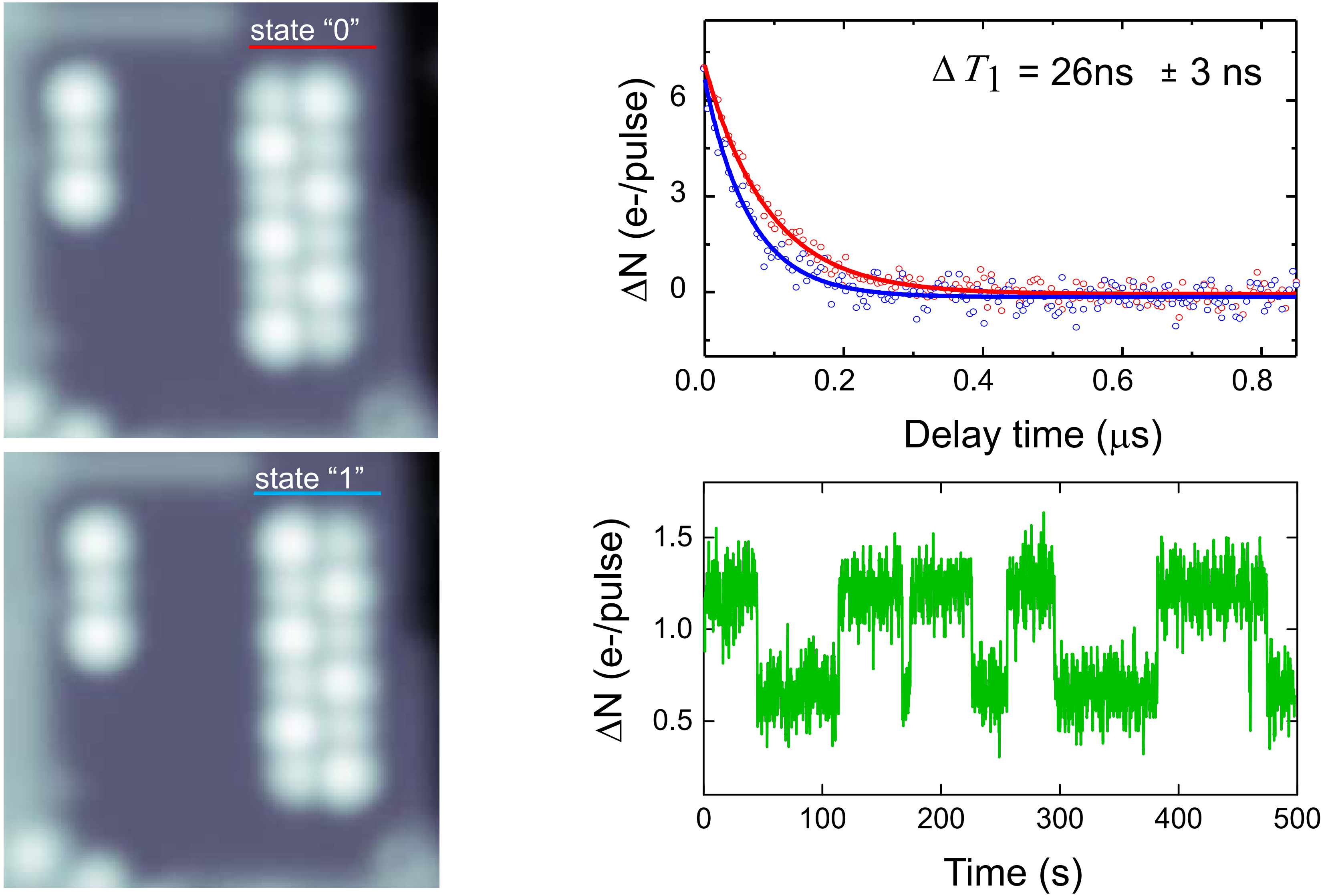
Sensor spin　　　　Bistable antiferromagnet



Bistable antiferromagnets: Science 335 196 (2012).

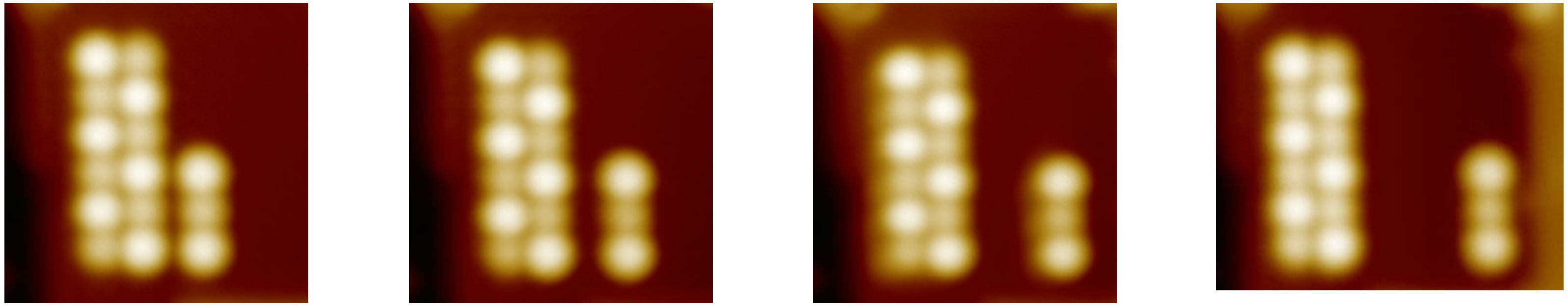
1 nm

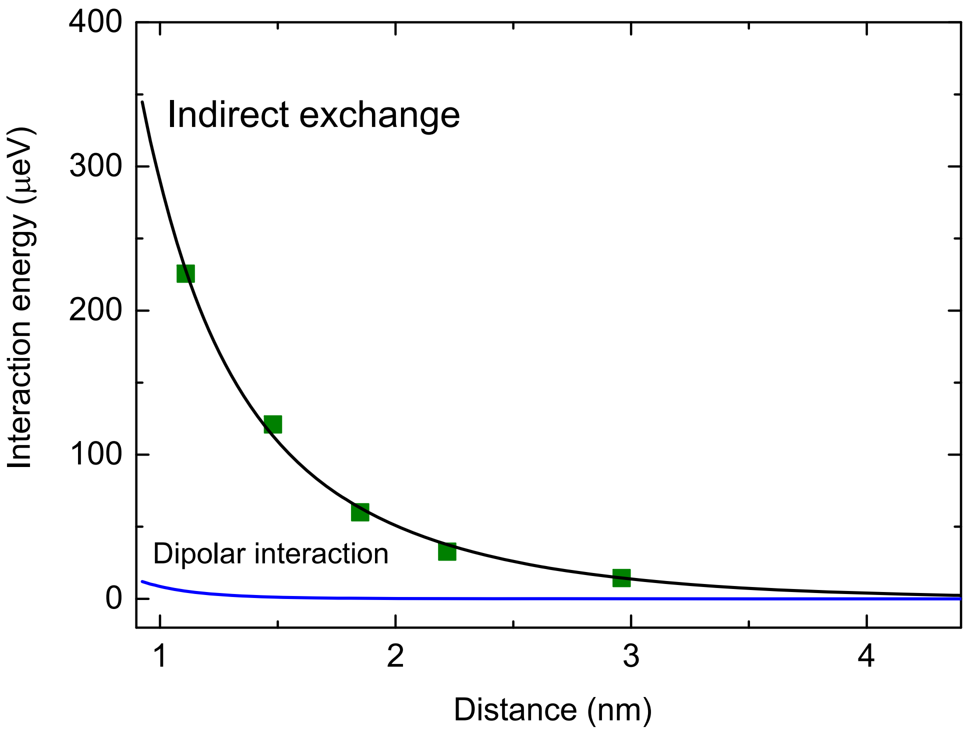
Remote Spin sensing at the atomic scale



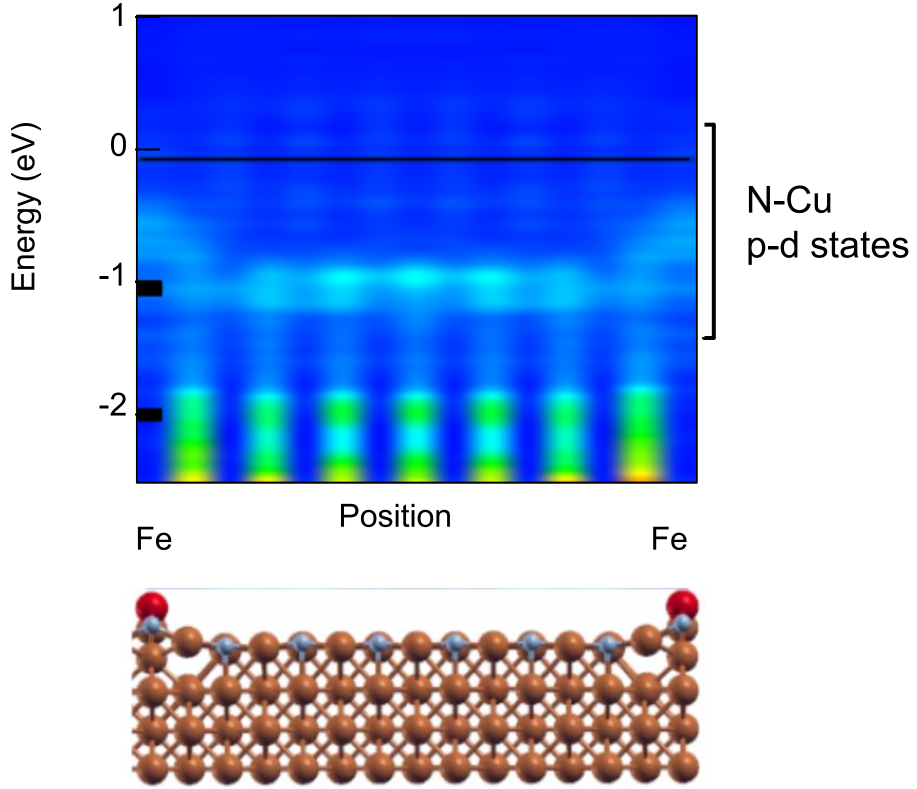
S. Yan, L. Malavolti, J. Burgess, A. Droghetti, A. Rubio, S. Loth, *Science Advances* 3 e1603137 (2017)

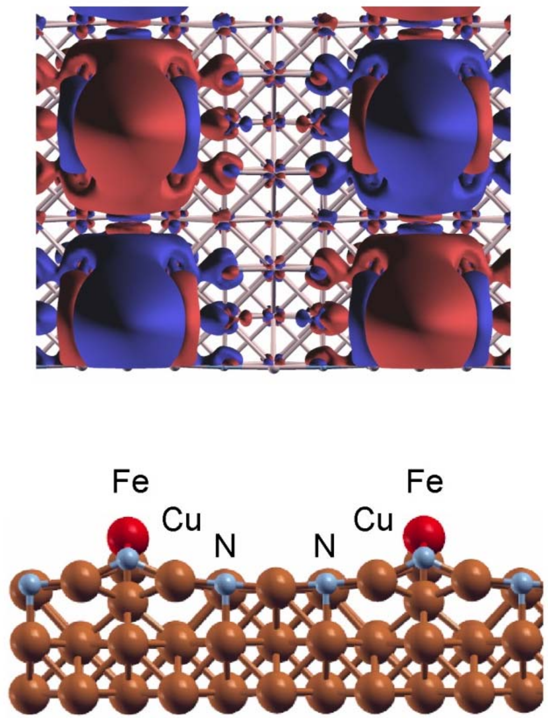
Long-range p-d exchange interaction





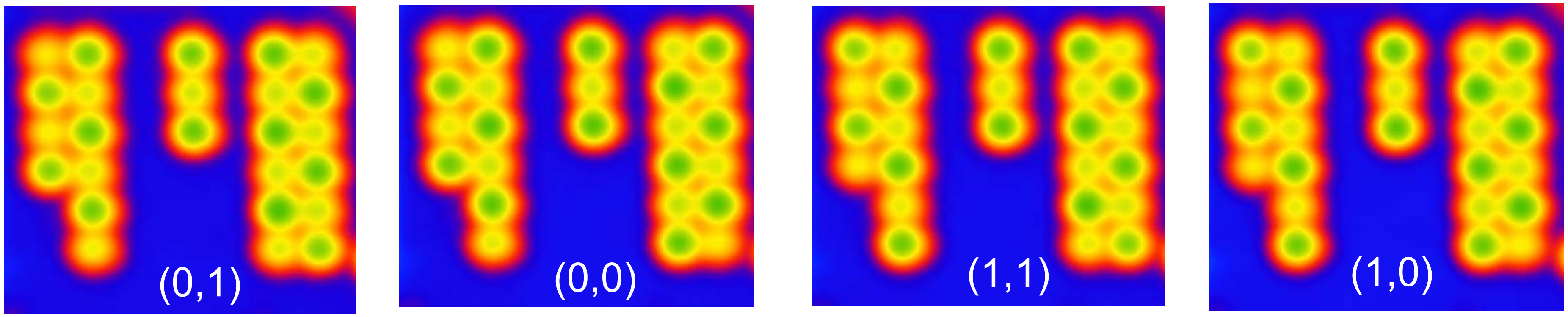
Long-range p-d exchange through Cu_2N network

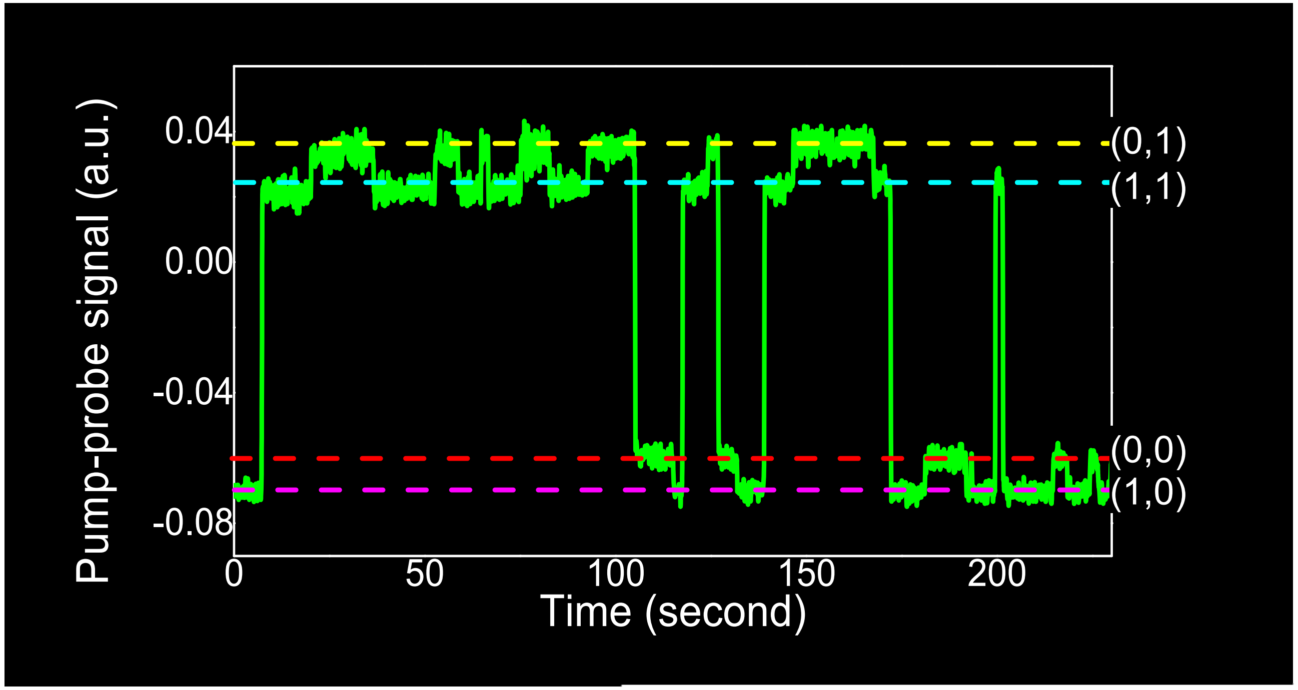




S. Yan, L. Malavolti, J. Burgess, A. Droghetti, A. Rubio, S. Loth, *Science Advances* 3 e1603137 (2017)

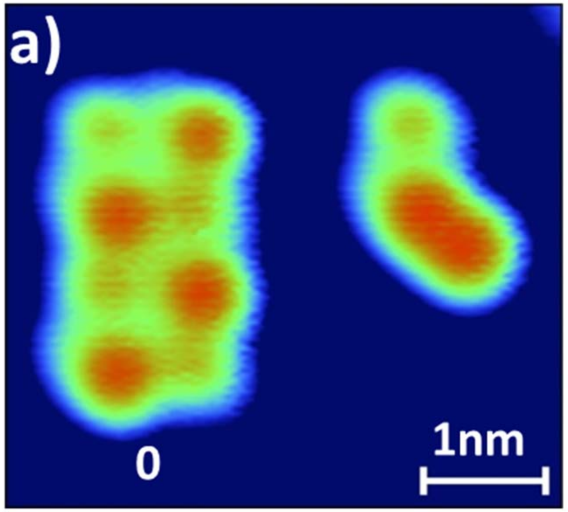
Remote sensing of correlated spin states

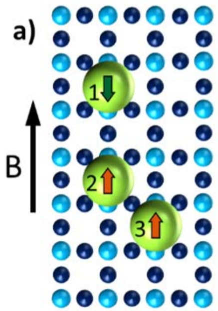


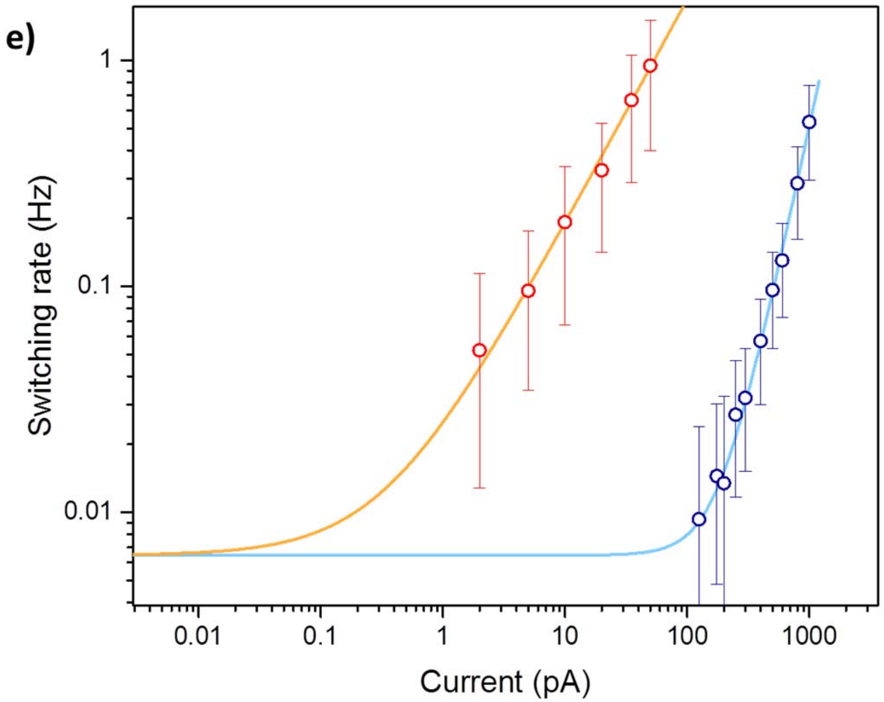


Antiferromagnetic correlation: $\dfrac{P_{(0,1)} + P_{(1,0)}}{P_{(0,0)} + P_{(1,1)}} = 1.12 \pm 0.09$

What is the merit of remote spin sensing?



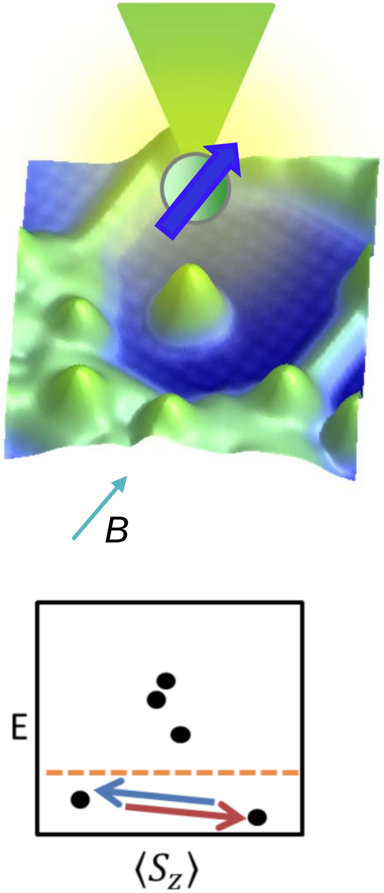


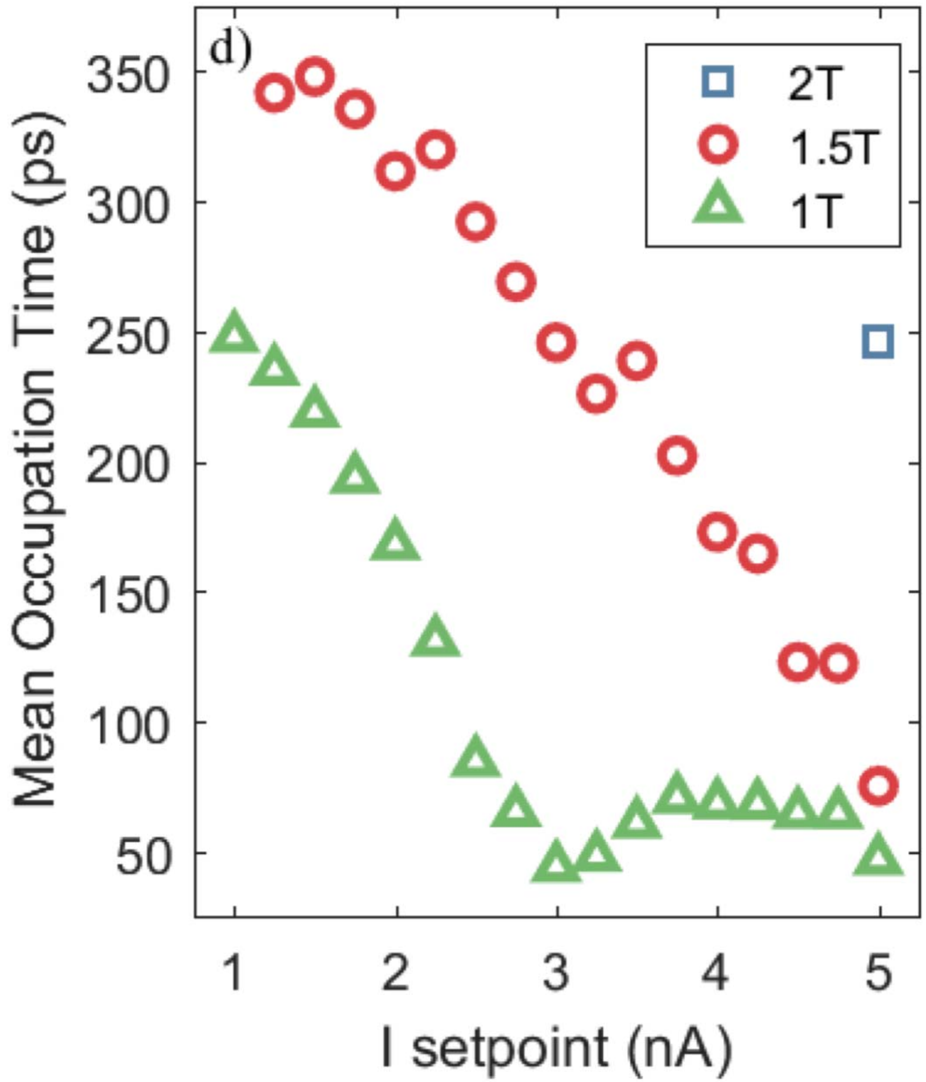


- 100x less invasive than direct measurement
- Approaches non-invasive measurement condition

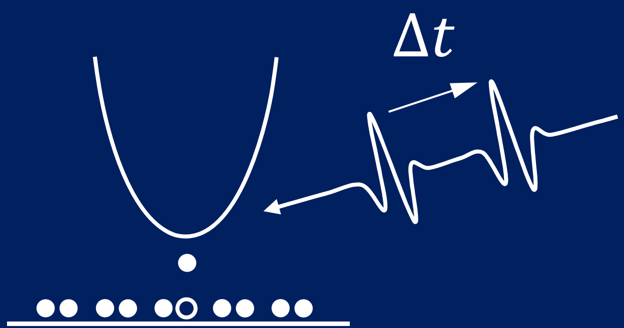
L. Malavolti et al. *forthcoming* (2019)
S. Rolf-Pissarczyk, S. Yan, L. Malavolti, J.A.J. Burgess, G. McMurtrie, S. Loth PRL 119, 217201 (2017).

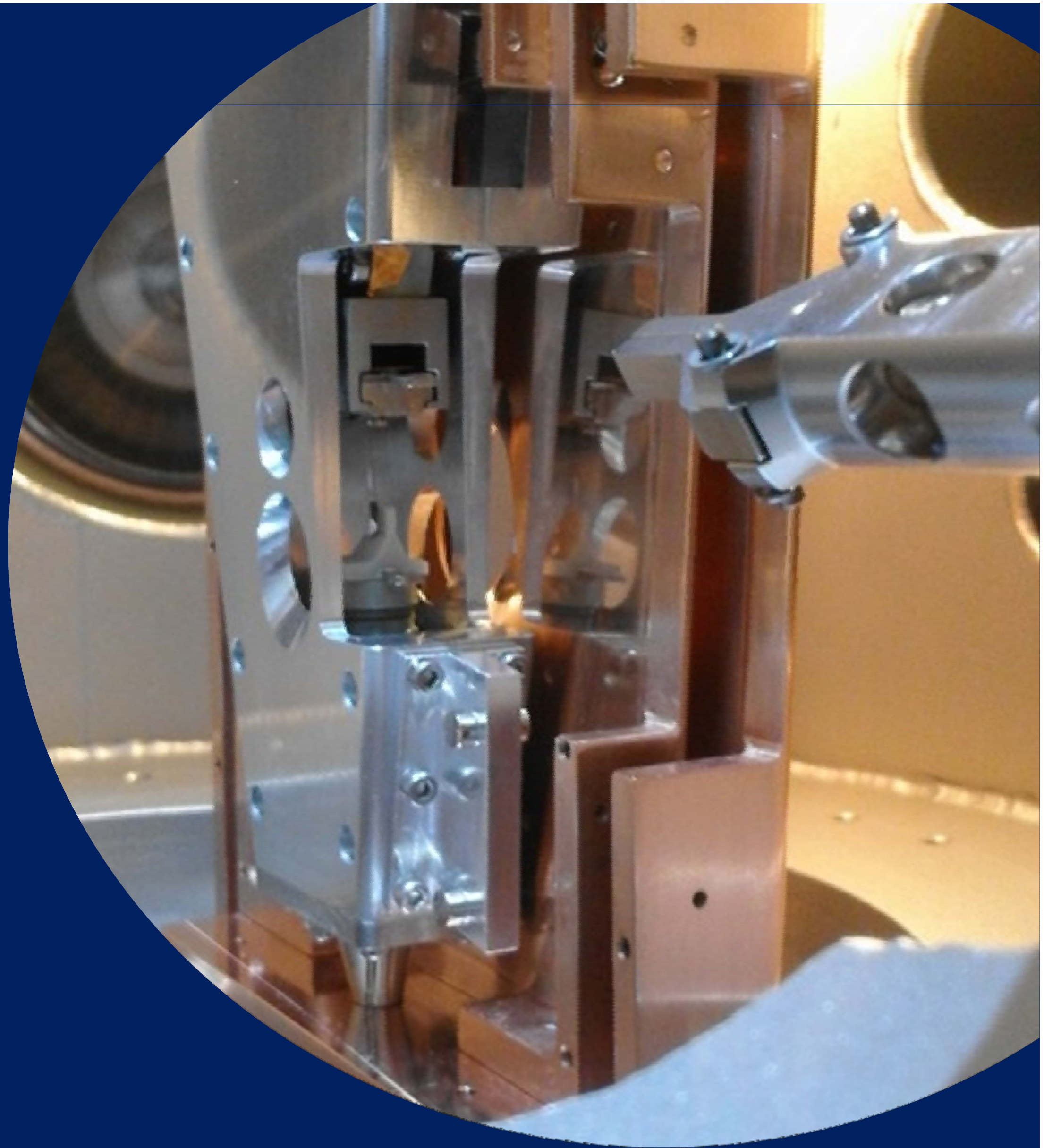
Switching speed as function of spin state composition





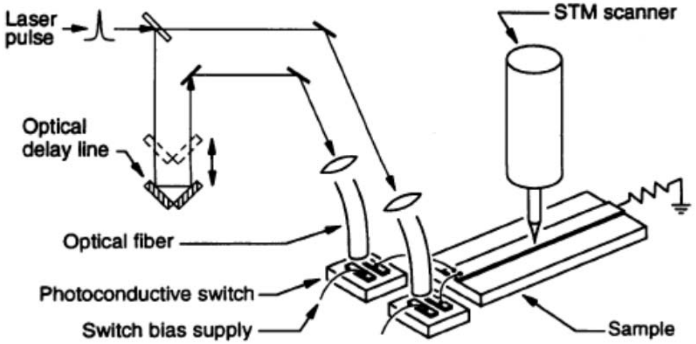
Charge density dynamics with femtosecond resolution

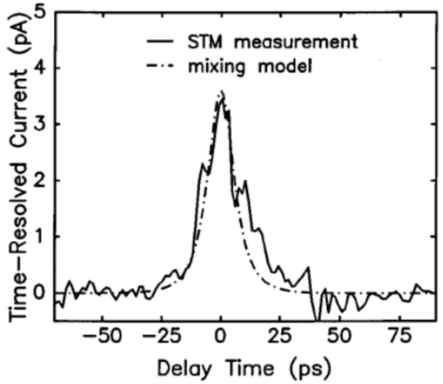




Ultrafast STM beyond 10 ps speed

Nunes, Freeman Science (1993)

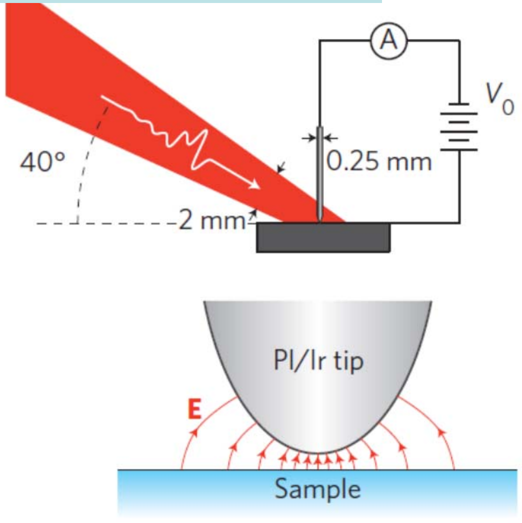




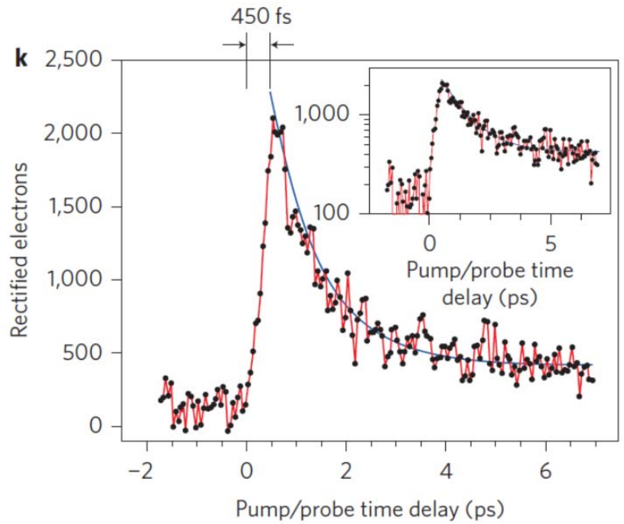
Optical switches

Ultrafast STM beyond 10 ps speed

THz-induced field emission

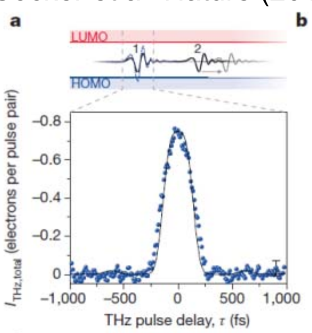


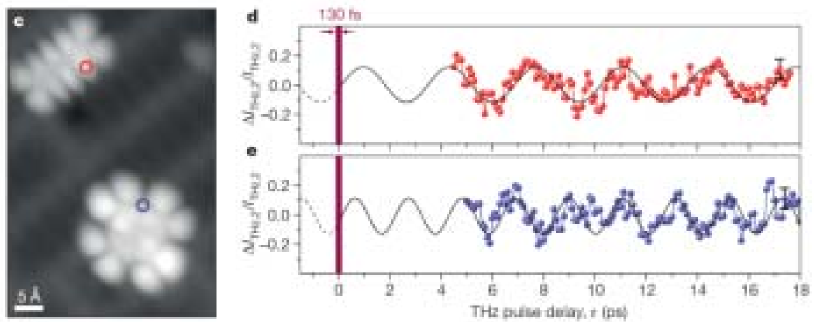
Cocker et al. Nature Photonics (2013)



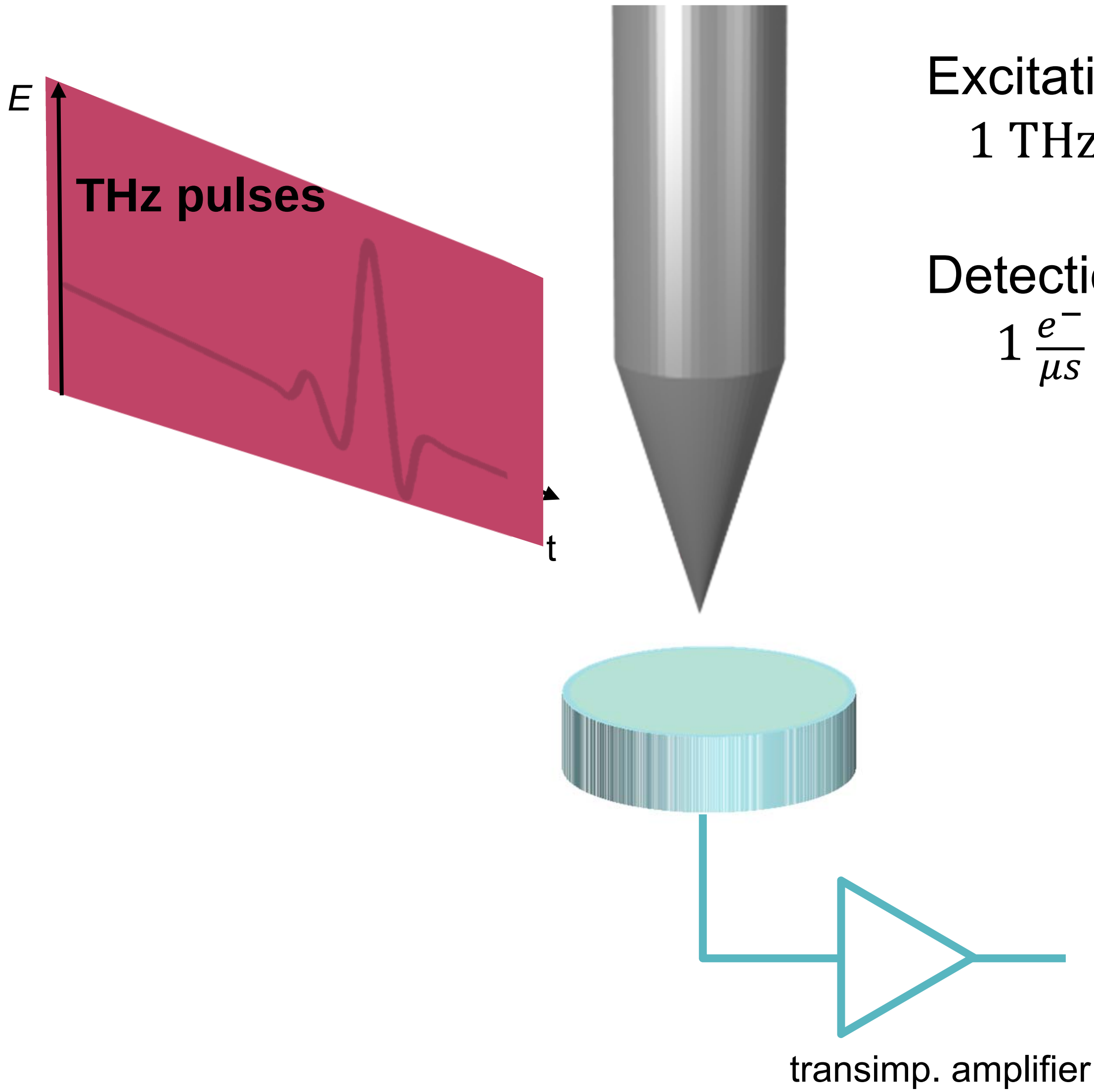
THz-induced tunneling

Cocker et al. Nature (2016).





The THz – STM principle



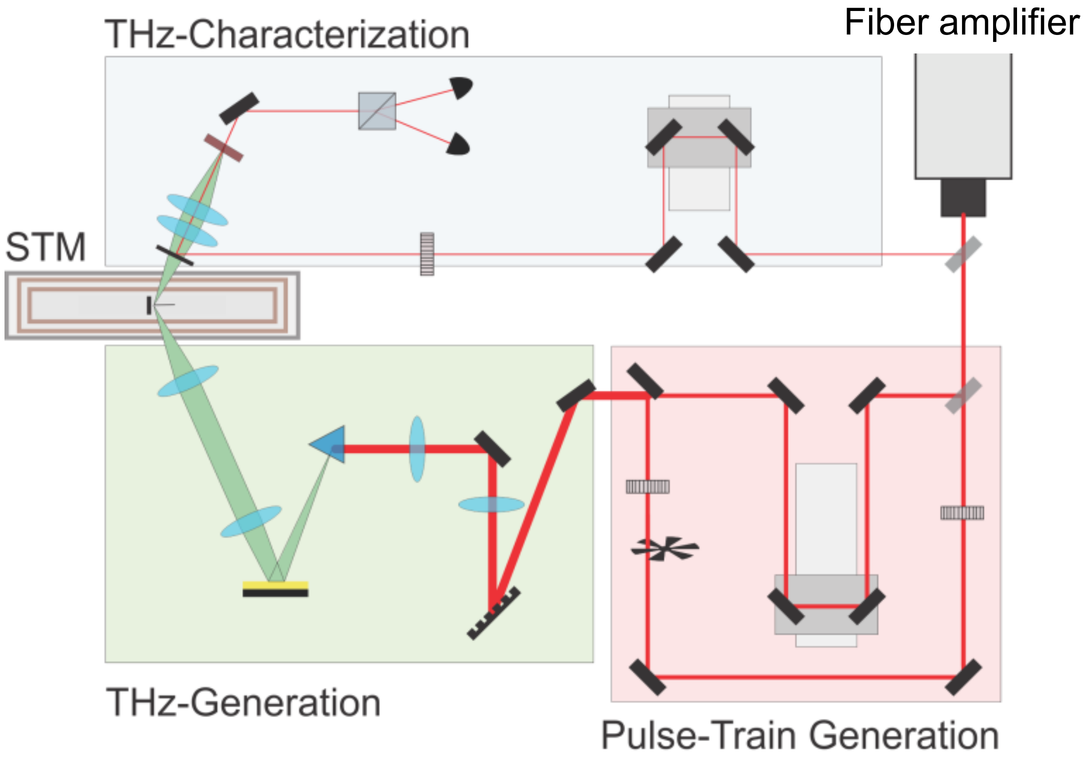
Excitation:

$$1\ \text{THz} \;\hat{=}\; 300\ \mu\text{m} \;\hat{=}\; 4\ \text{meV}$$

Detection:

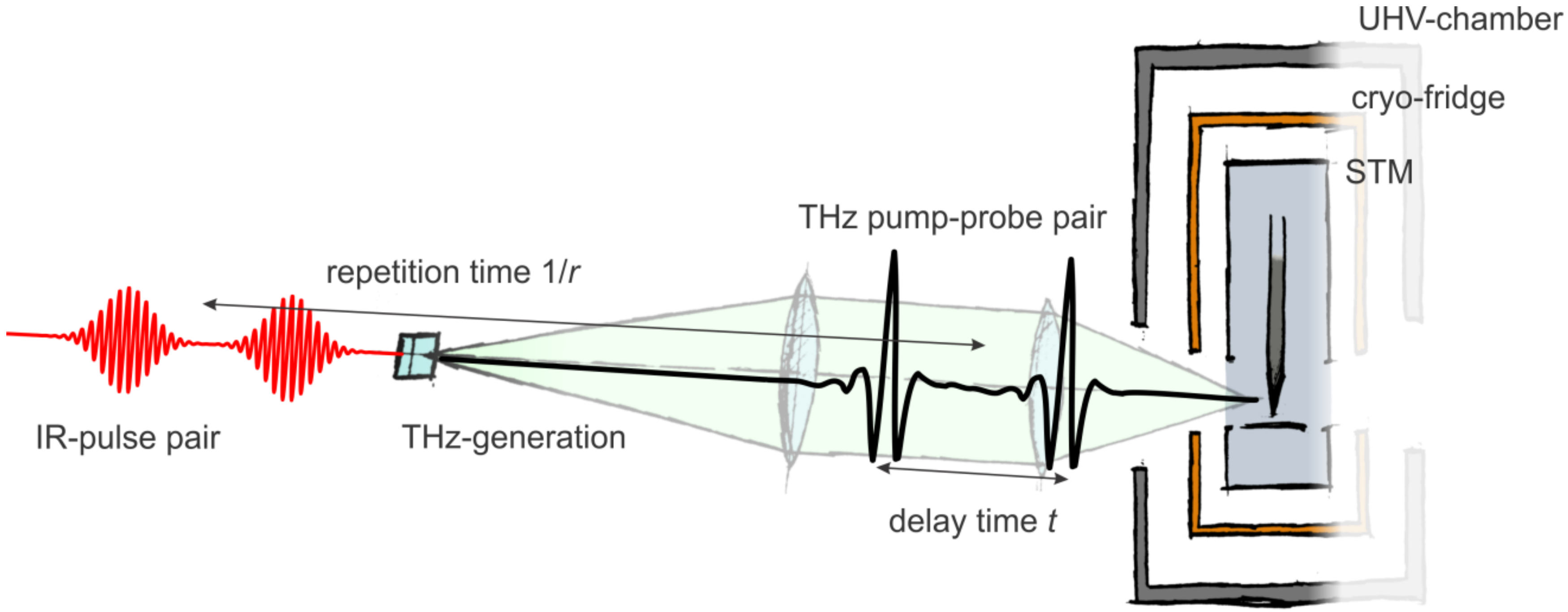
$$1\,\frac{e^-}{\mu s} \;\hat{=}\; 160\ fA$$

Ultrafast „voltage“ source

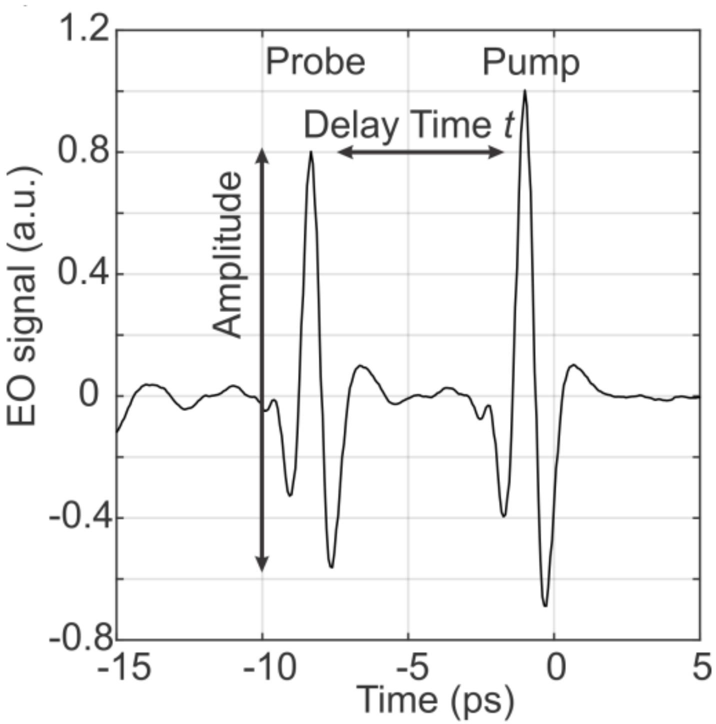


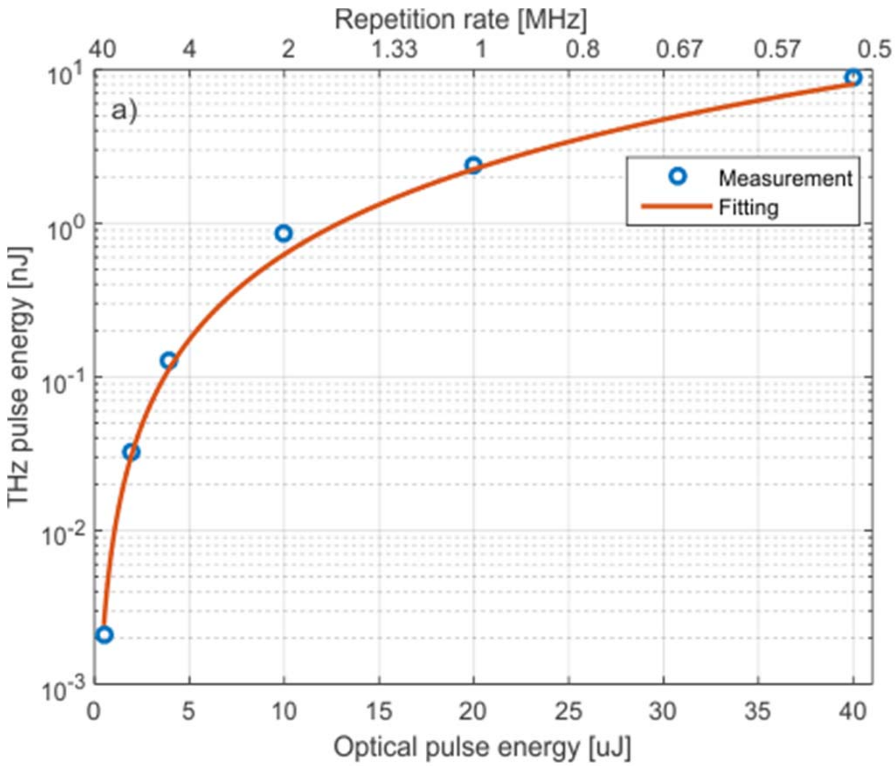
Collaboration with A. Cavalleri, S. Rajasekaran, A. Cavalieri, I. Grguras (MPI Hamburg)

THz – STM Idea


UHV-chamber
cryo-fridge
STM
THz pump-probe pair
repetition time 1/r
IR-pulse pair
THz-generation
delay time t

Ultrafast „voltage“ source



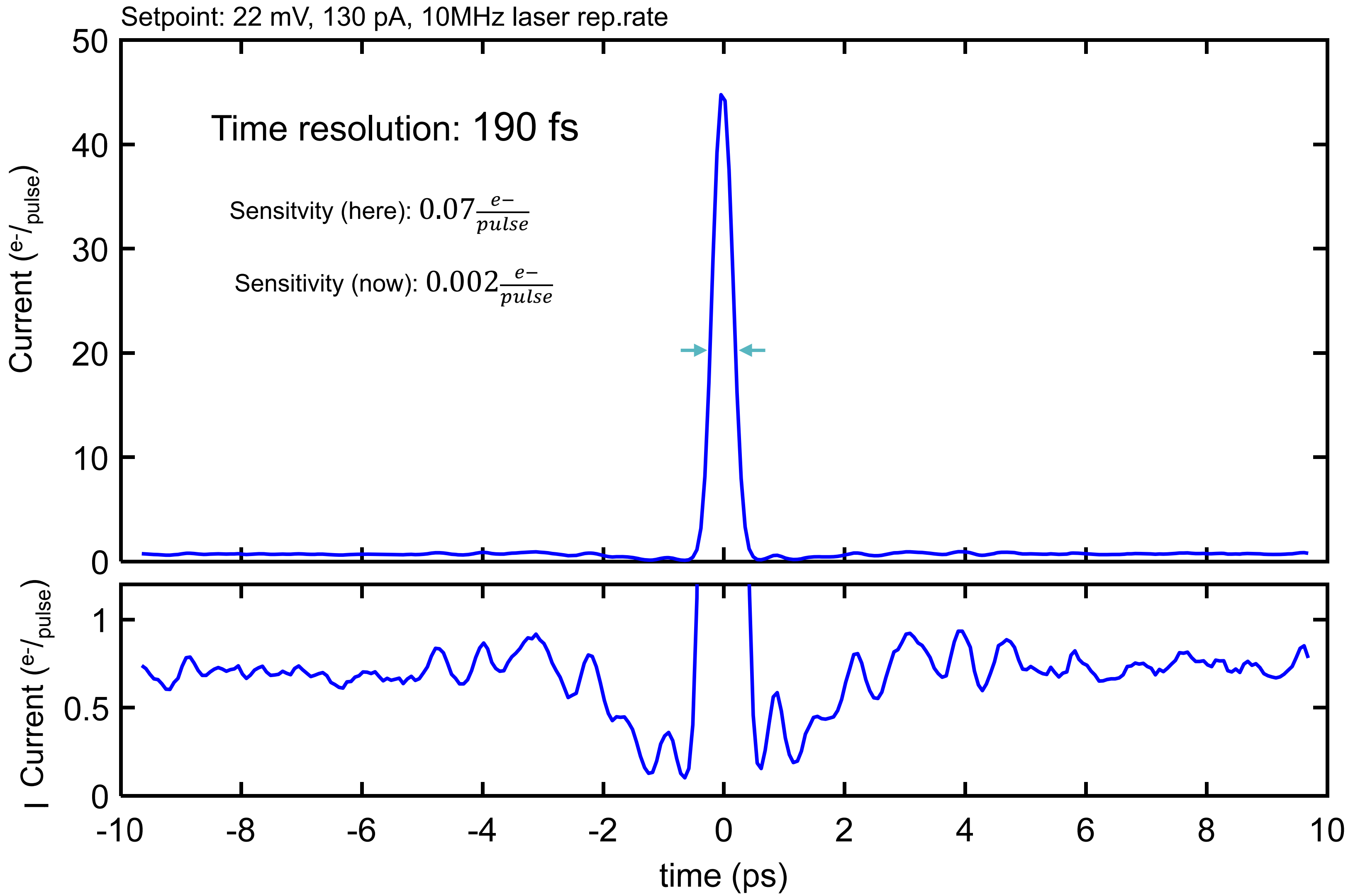


Peak junction voltage in STM

10 mV $|_{40\ \mathrm{MHz}}$ – 30 V $|_{0.5\ \mathrm{MHz}}$

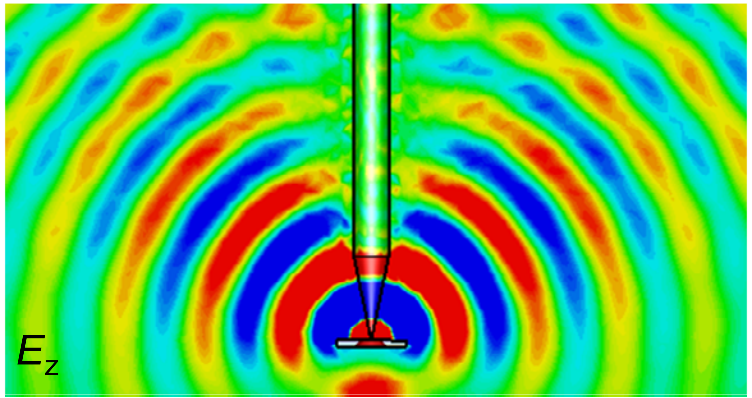
Collaboration with A. Cavalleri, S. Rajasekaran, A. Cavalieri, I. Grguras (MPI Hamburg)

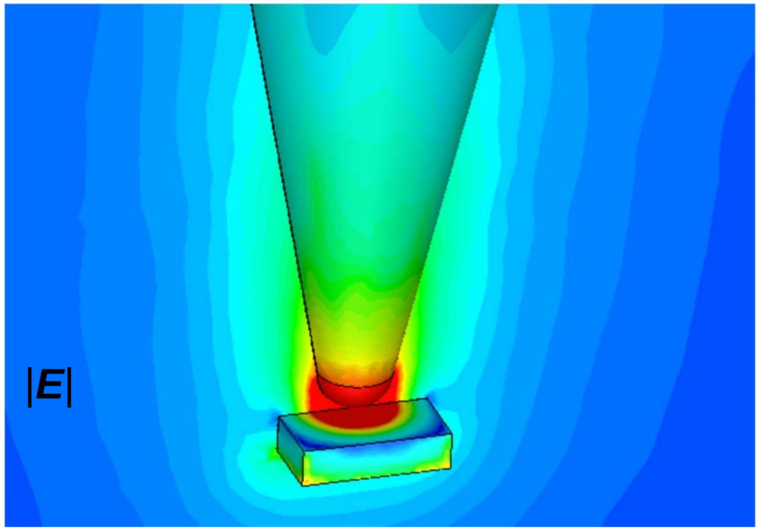
Ultrafast and ultrasensitive spectroscopy



THz coupling to STM tip

Antenna radiation pattern at 0.5 THz



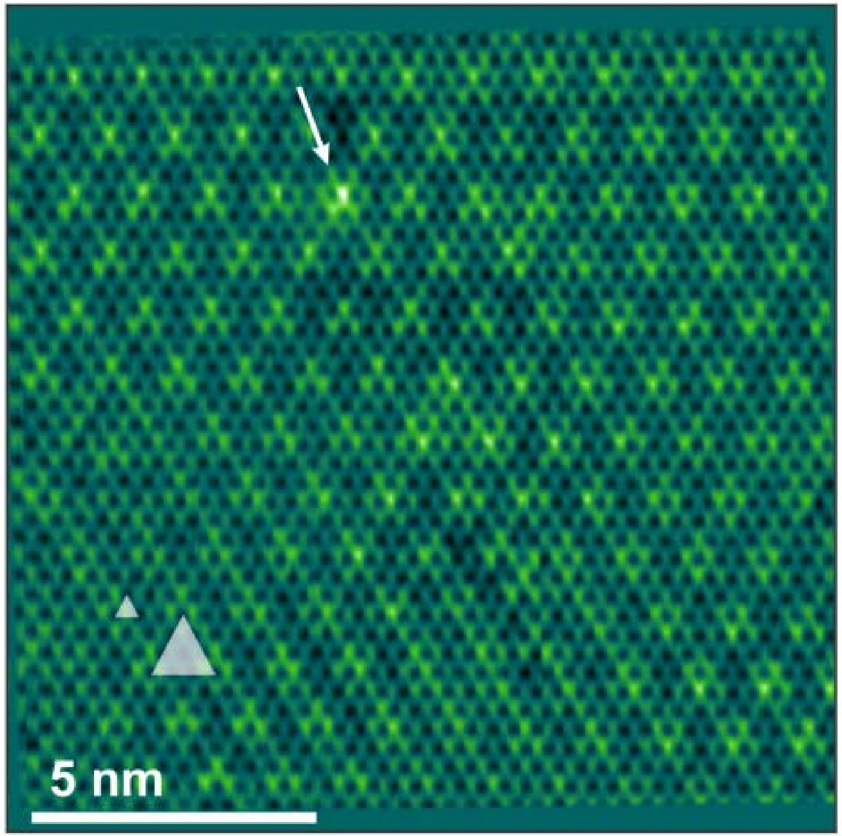


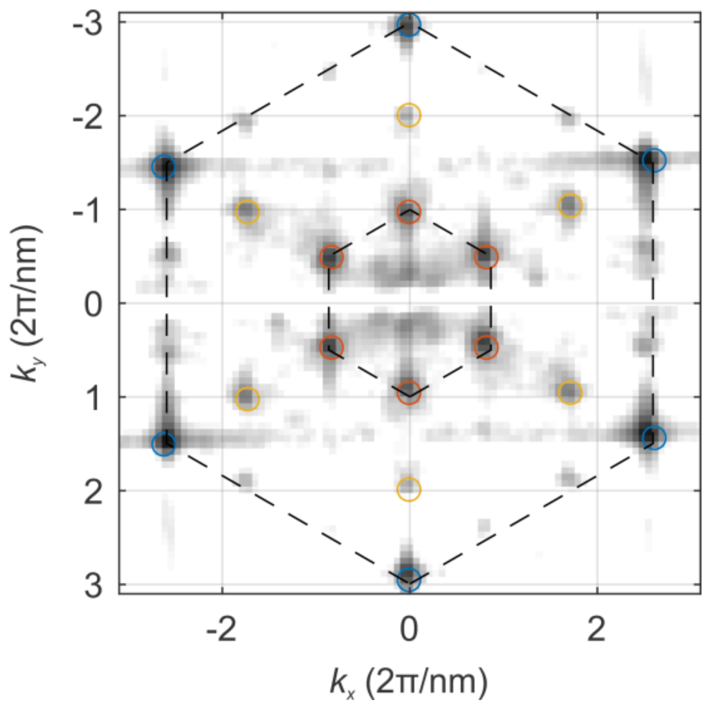
- THz E-field couples to **tip**
- Field enhancement at tip apex
  $$\frac{E_z^{tip}}{E_z^{ff}} \approx 1.000 \; - \; 10.000$$
- Scaling of enhancement
  FEM model: junction capacity
  Experiment: microapex modifications

See also: Nguyen et al. La Phys. Can. 71 157 (2015), Jelic et al. Nat Phys. Adv.online (2017).

Charge density waves in 2H-$NbSe_2$

$NbSe_2$ T = 20K, T_{CDW} = 38 K





CDW gap: 2 meV
(ARPES, Borisenko et al. PRL 102 166402, 2009)

CDW formation driven by electron-phonon interaction.

Fermi surface nesting & Phonon softening

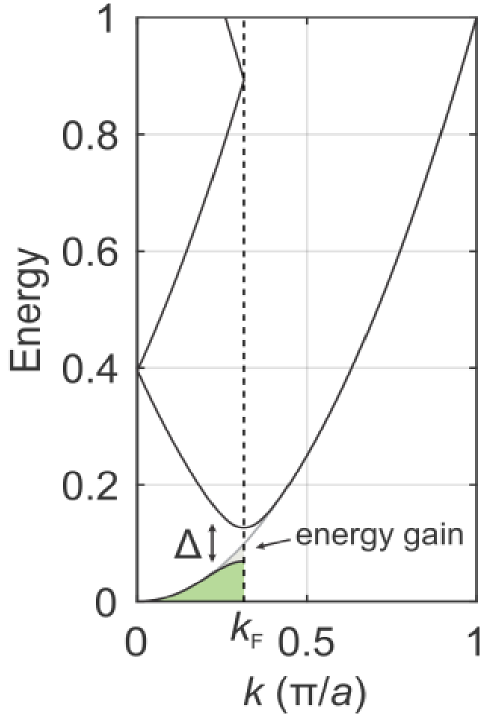
Incommensurate CDW

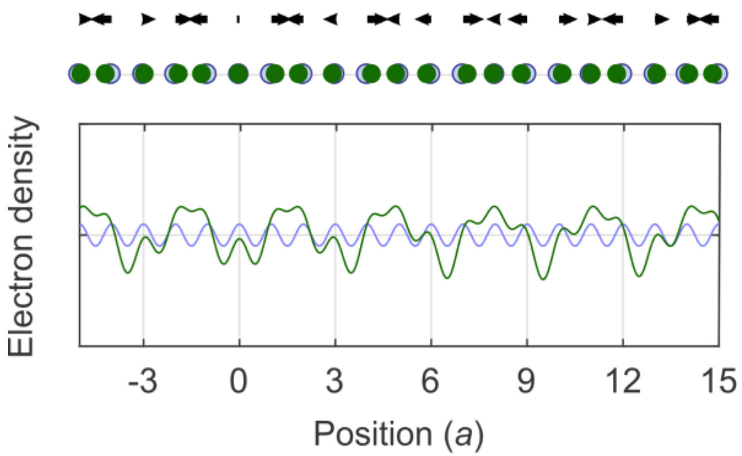
$$\vec{q}_1 = (3.1 \pm 0.05) \cdot \vec{q}_{CDW}$$

$NbSe_2$:
Flicker, van Wezel Nat. Comm. 6 7034 (2015)
Malliakas, Kanatzidis JACS 135 1719 (2013)
Inosov et al. New J. Phys. 10 125027 (2008)
Borisenko et al. PRL 102 166402 (2009)
etc., also
G. Grüner Rev. Mod. Phys. 60 1129 (1988)
W. McMillan Phys. Rev B 12 1187 (1975)

Charge-density wave formation

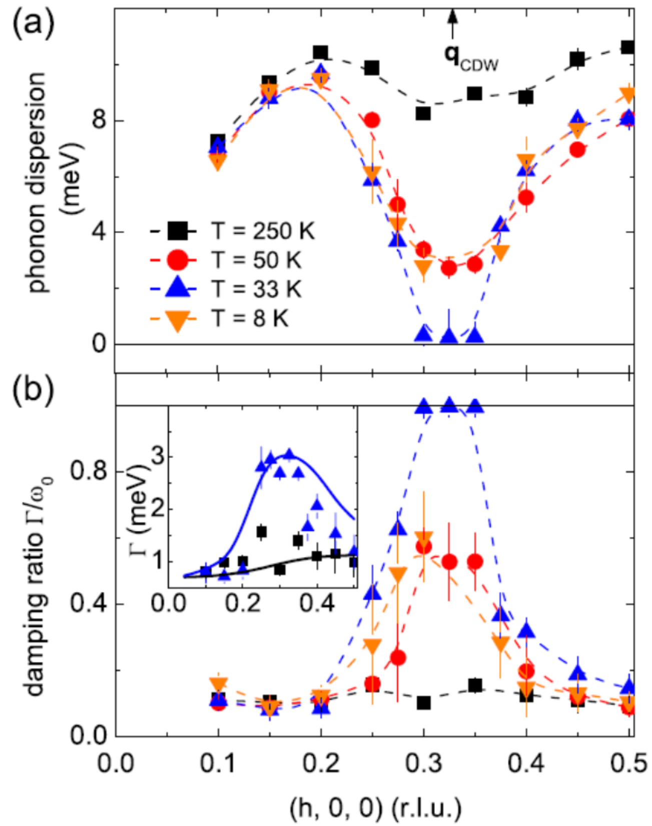
1D





2H-$NbSe_2$ (2D)

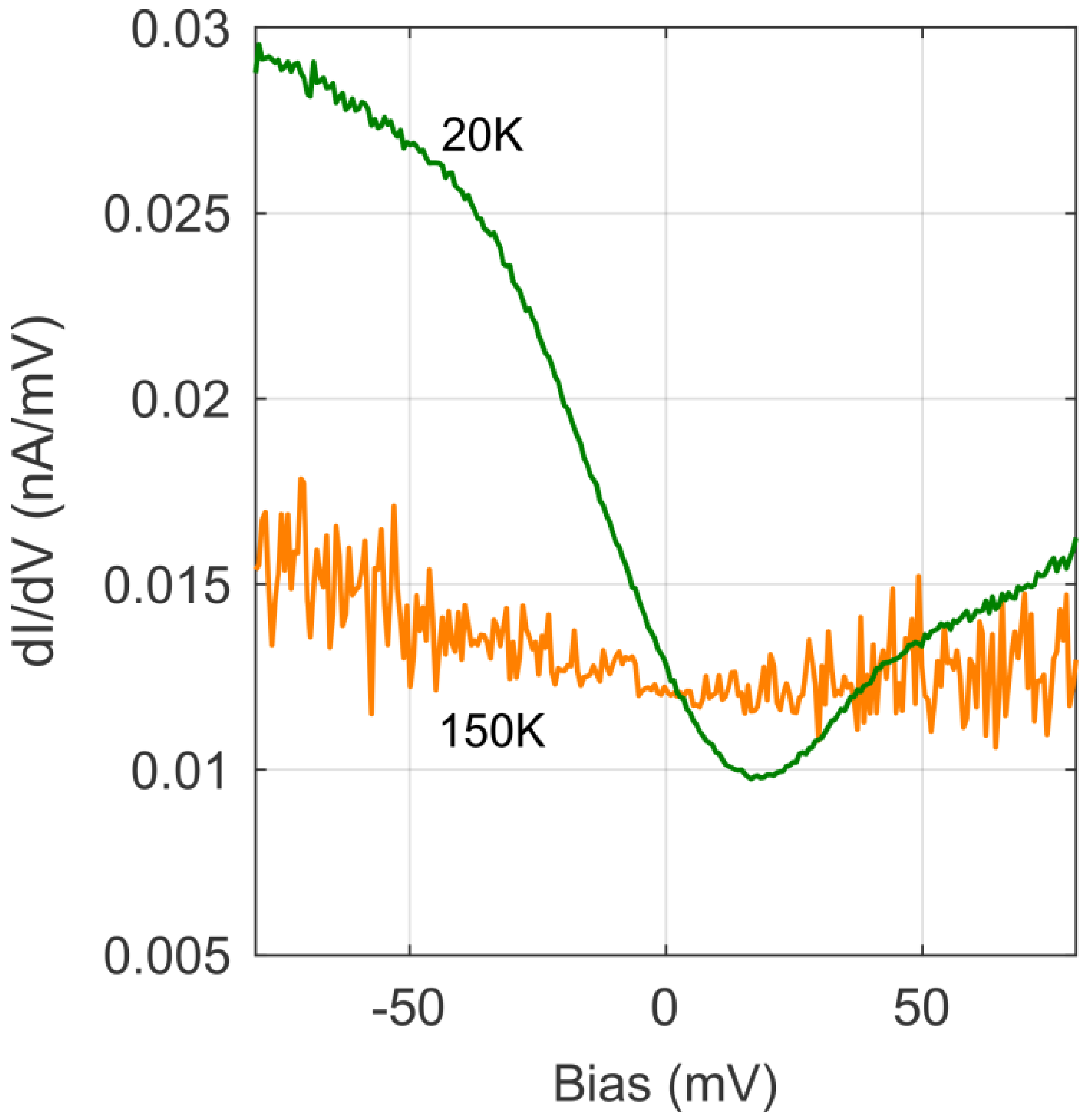
Weber et al. PRL 107 107403 (2011)



CDW formation driven by electron-phonon interaction.

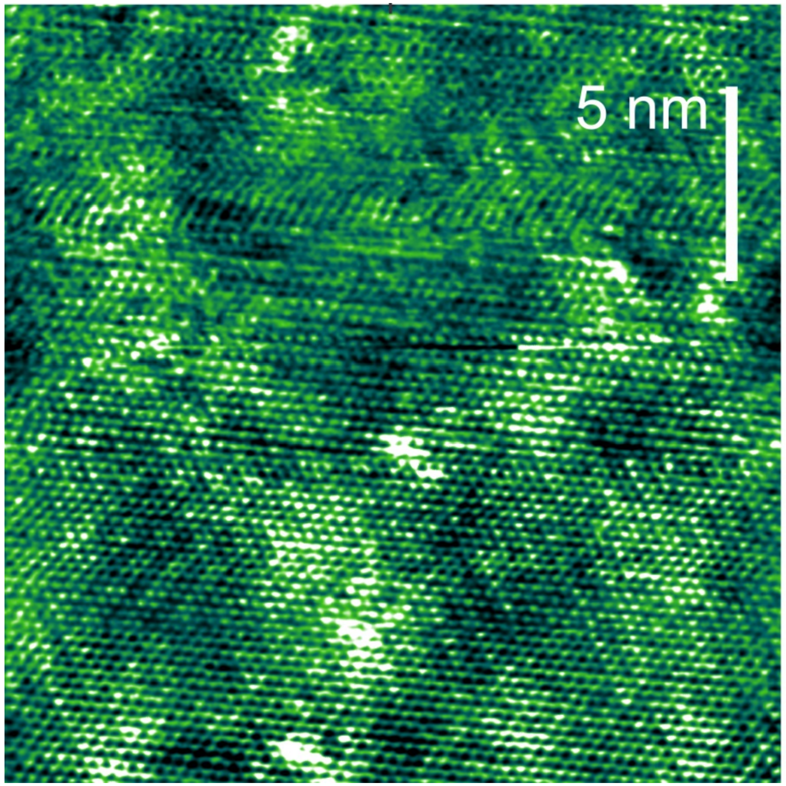
Fermi surface nesting & Phonon softening

Charge-density wave formation

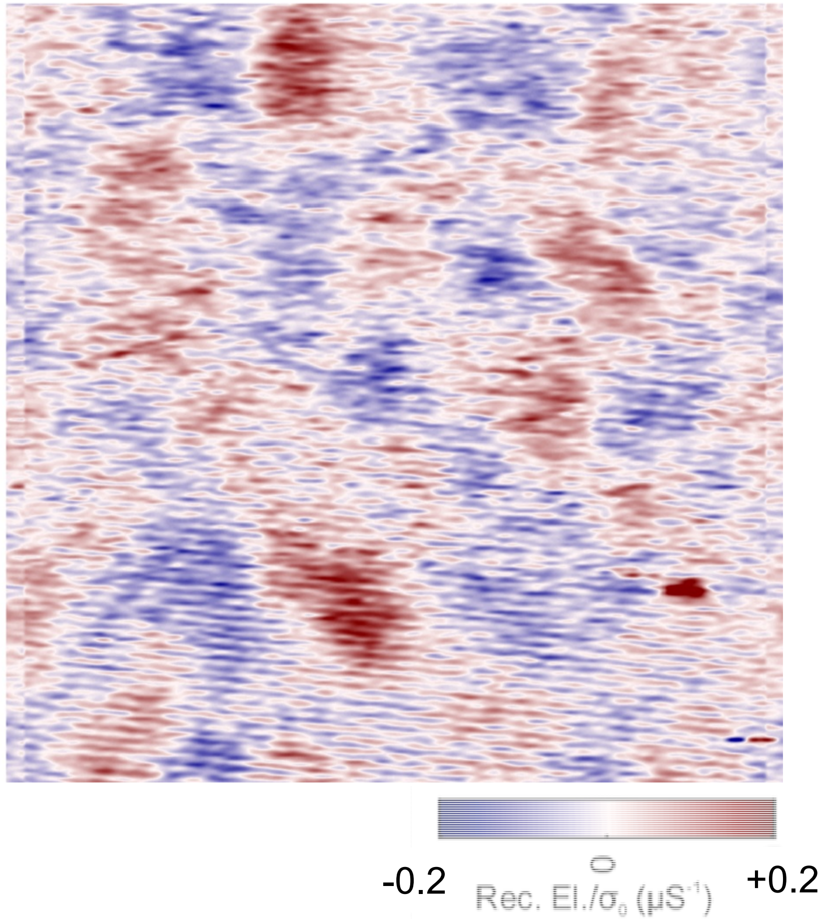


Ultrafast imaging of the CDW dynamics

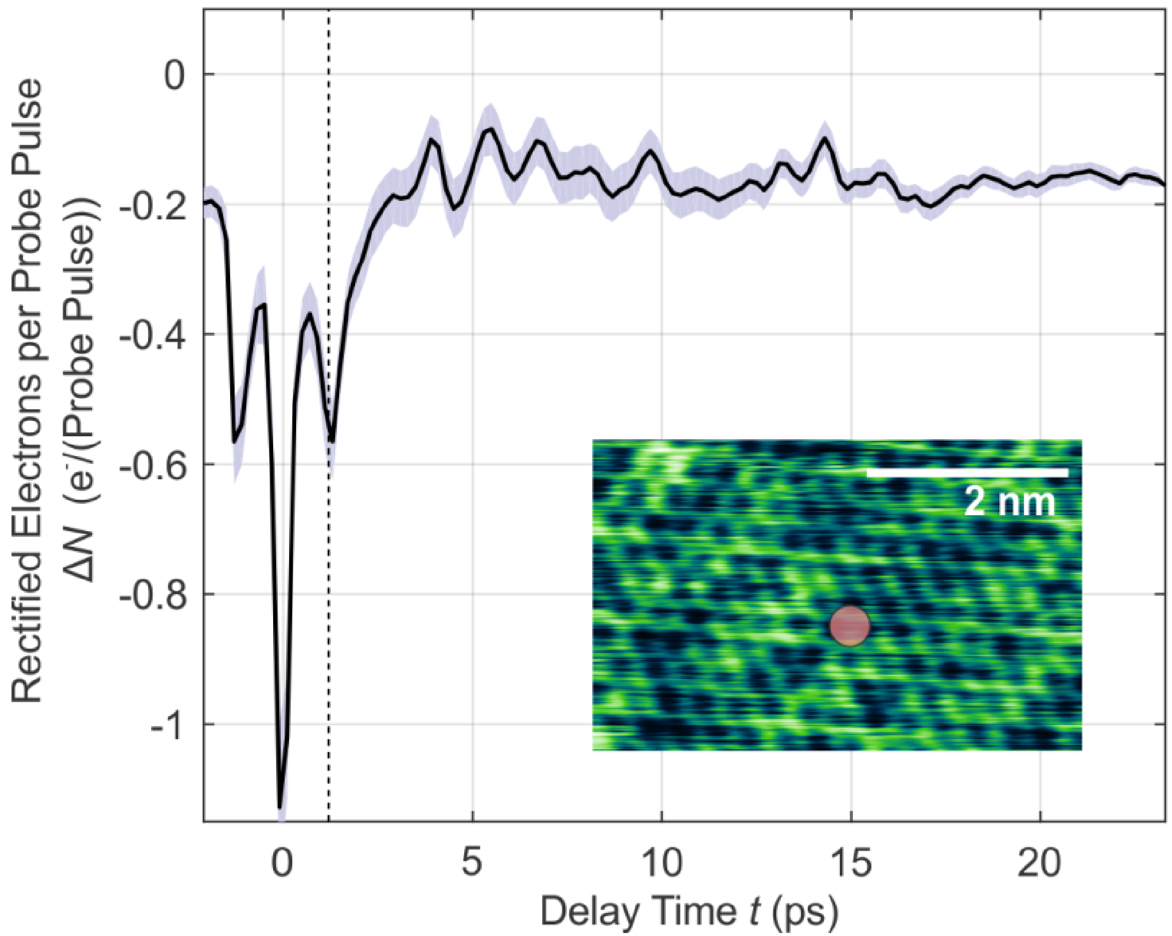
STM 1 nA / 1mV



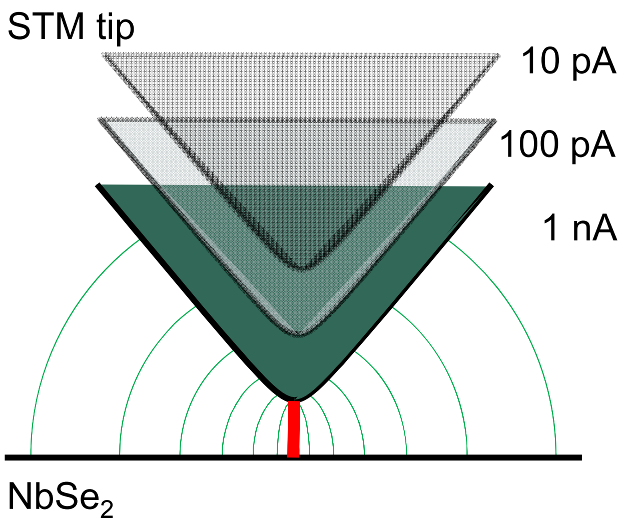
THz 40 MHz, fixed delay 300 fs

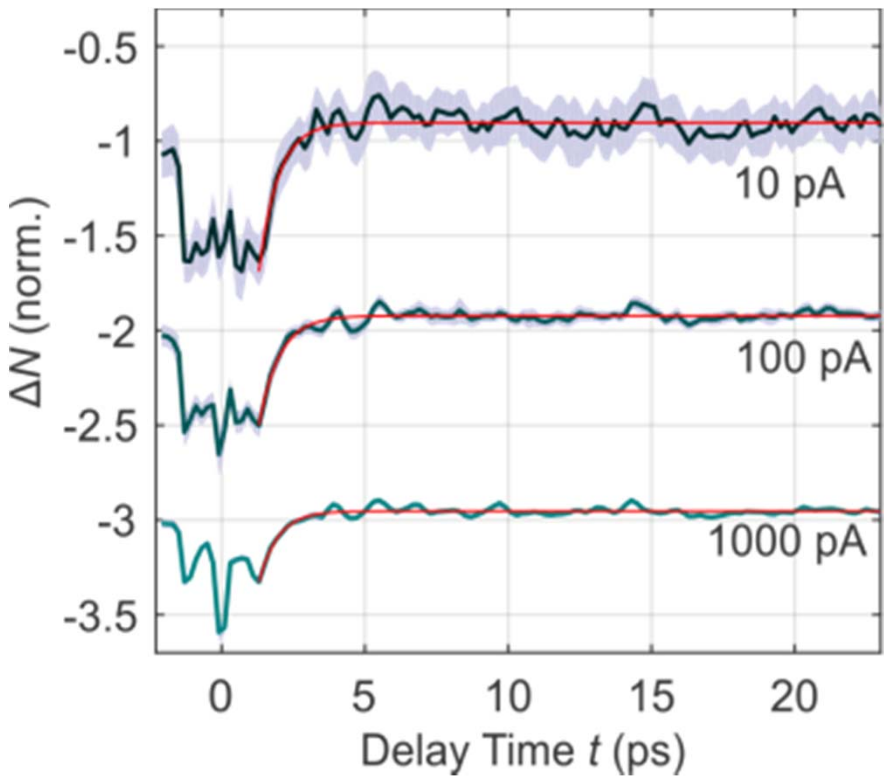


Local Pump Probe Spectrum of $NbSe_2$



What is the excitation mechanism?

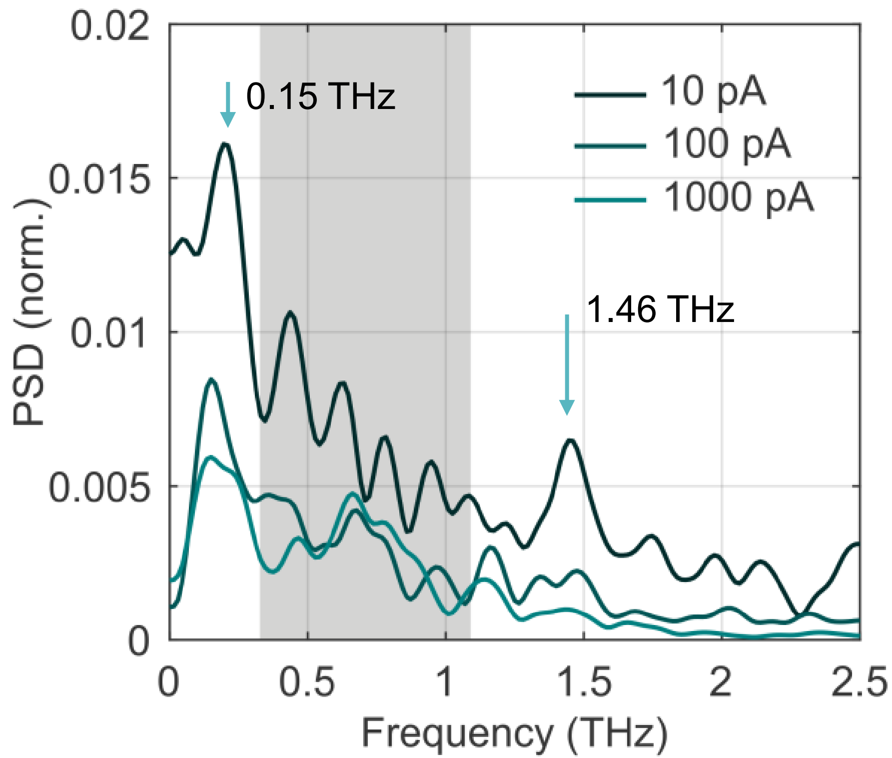




1 nA : Hot electron tunneling ($0.4\frac{e^-}{pulse}$)

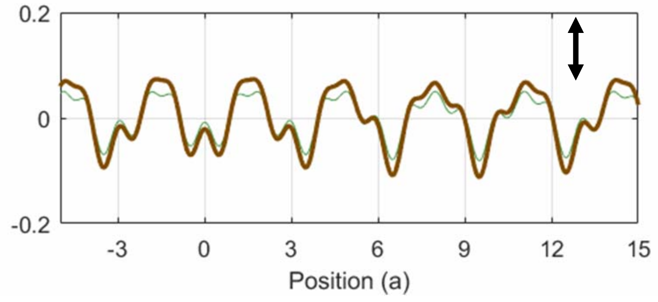
10 pA: Electric field coupling ($0.008\frac{e^-}{pulse}$)

Dynamic response of CDW after THz excitation

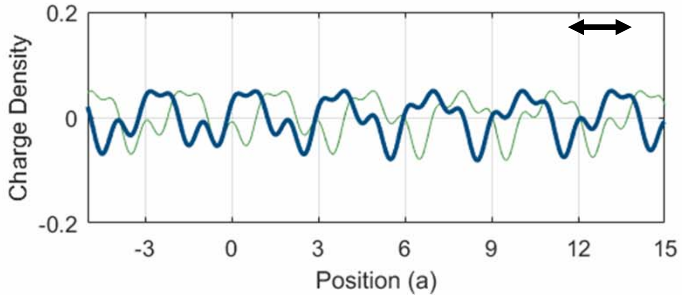


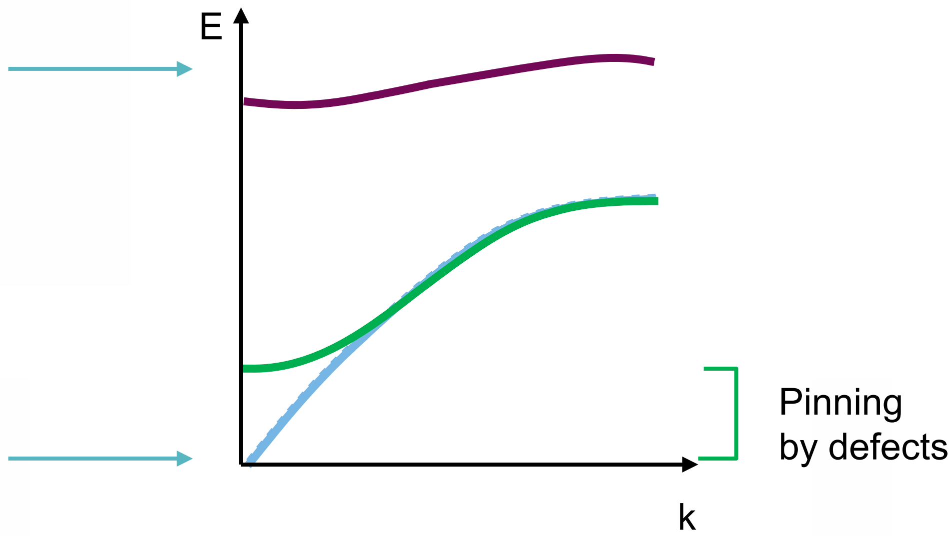
Collective excitations of a charge density wave

Amplitude mode



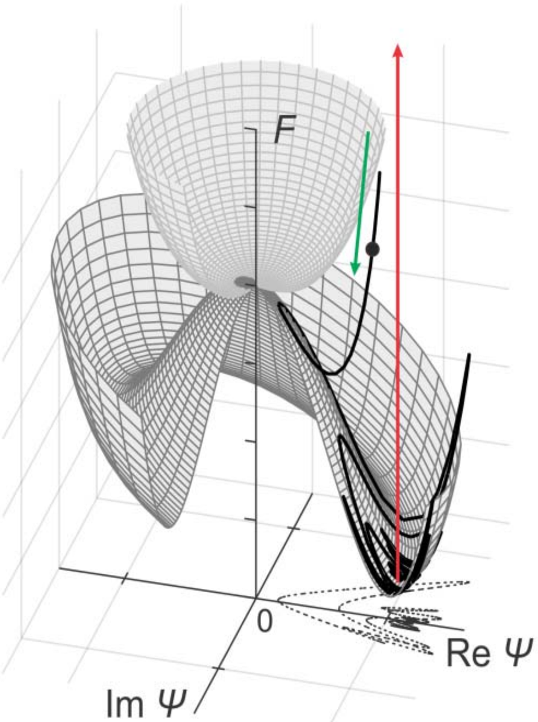
Phase mode

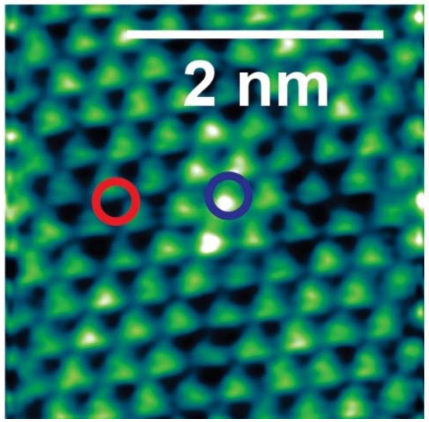




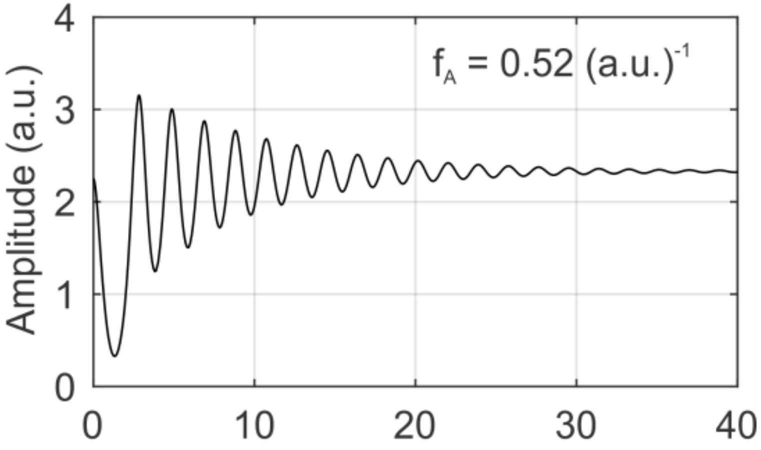
Perfect crystal	Perfect conductor by phase mode
Real crystal	Insulator for E < $E_{pinning}$ Pinning at defects ➔ Gap in phase mode

Atomic-scale CDW dynamics at a pinning center

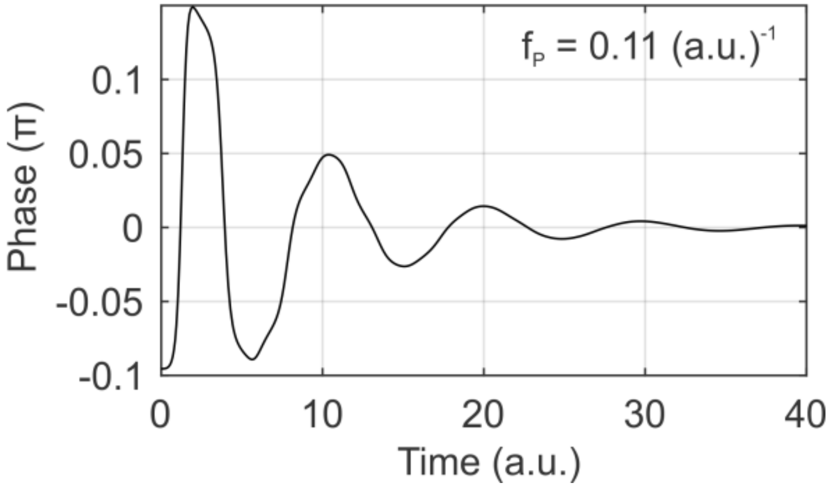




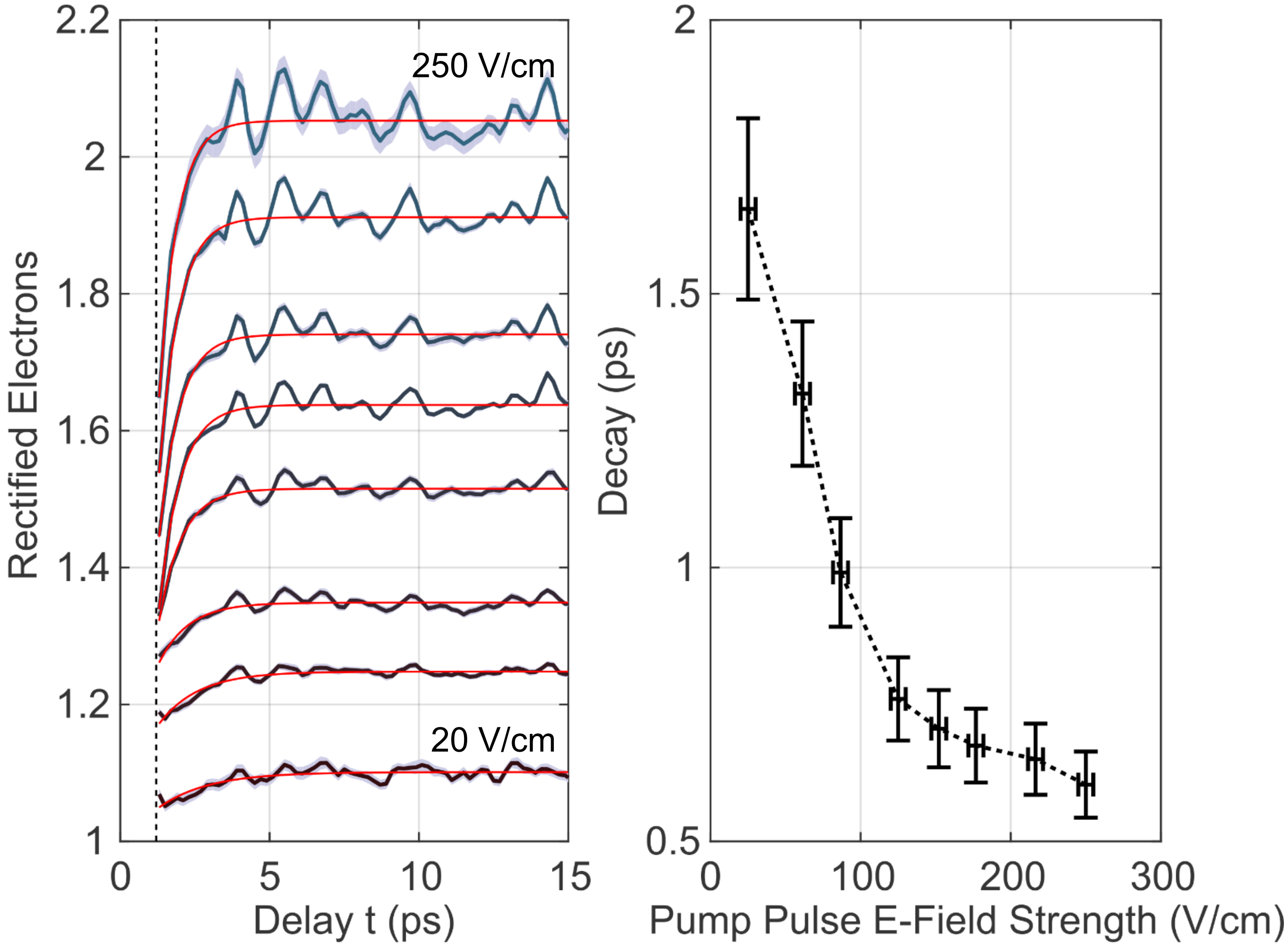
Amplitude mode



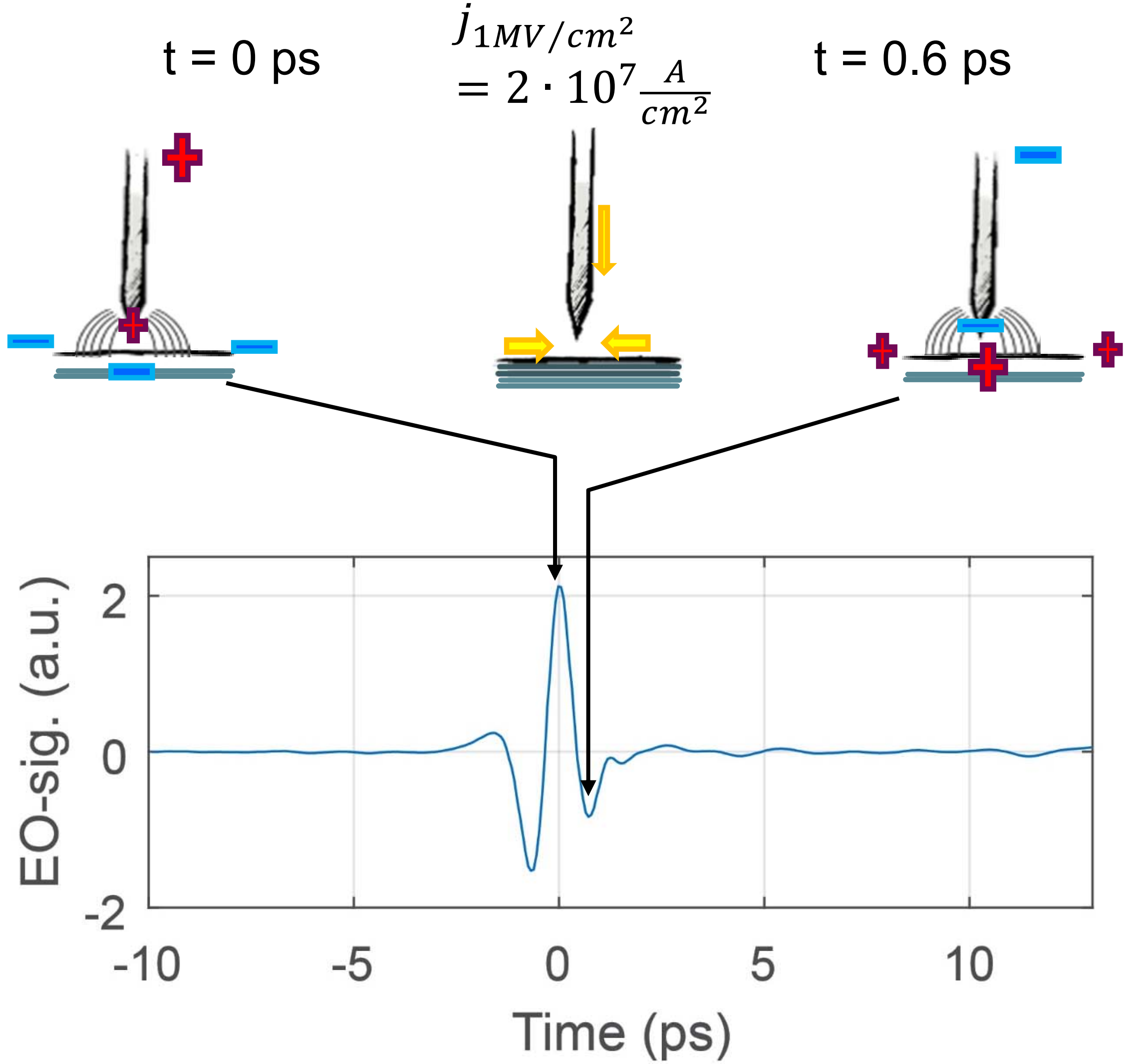
Pinned phase mode



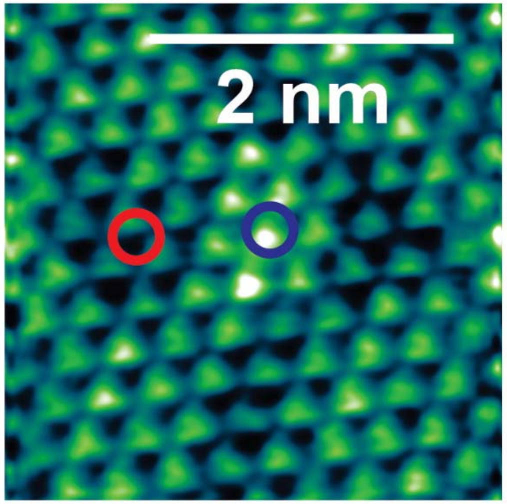
Ultrafast CDW recovery after electronic excitation

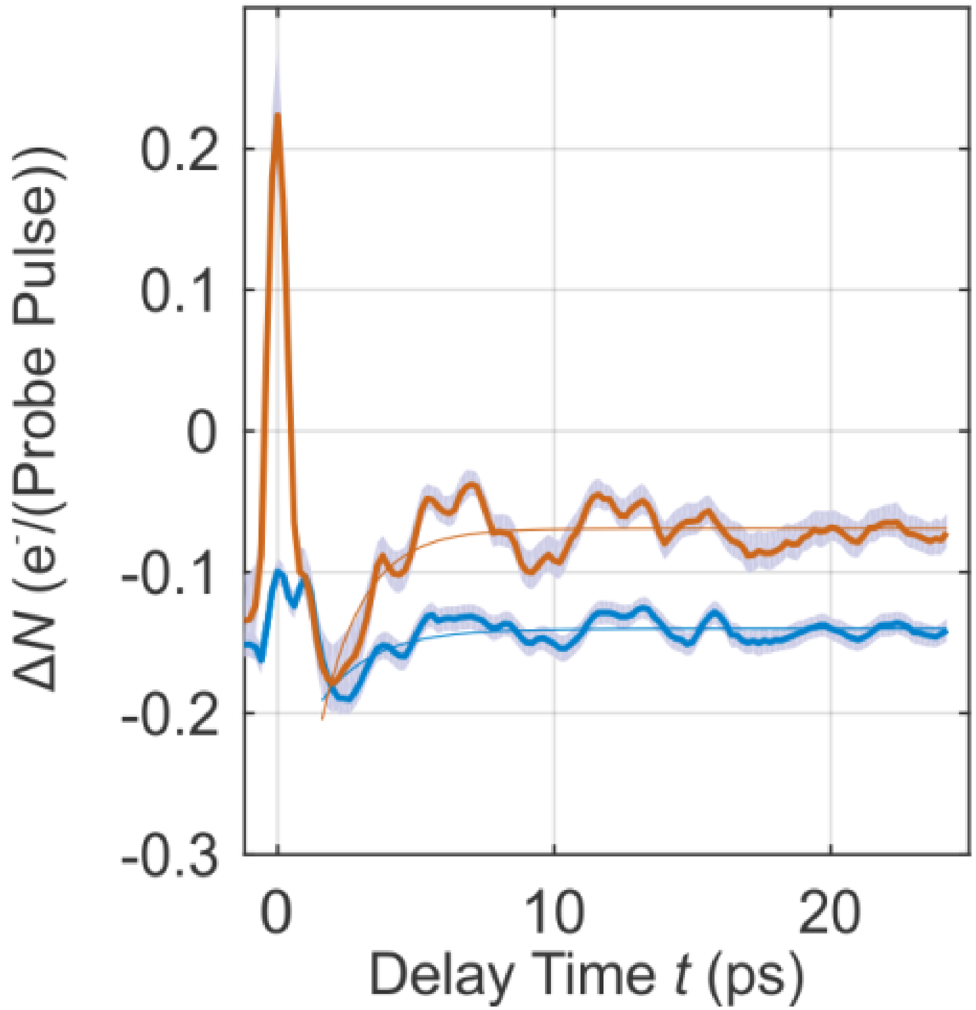


Electric-field-driven CDW excitation

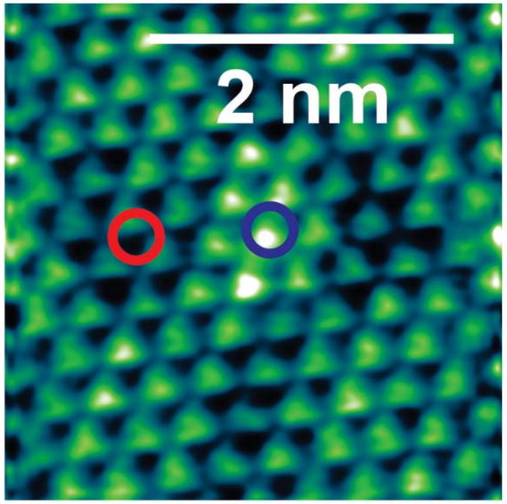


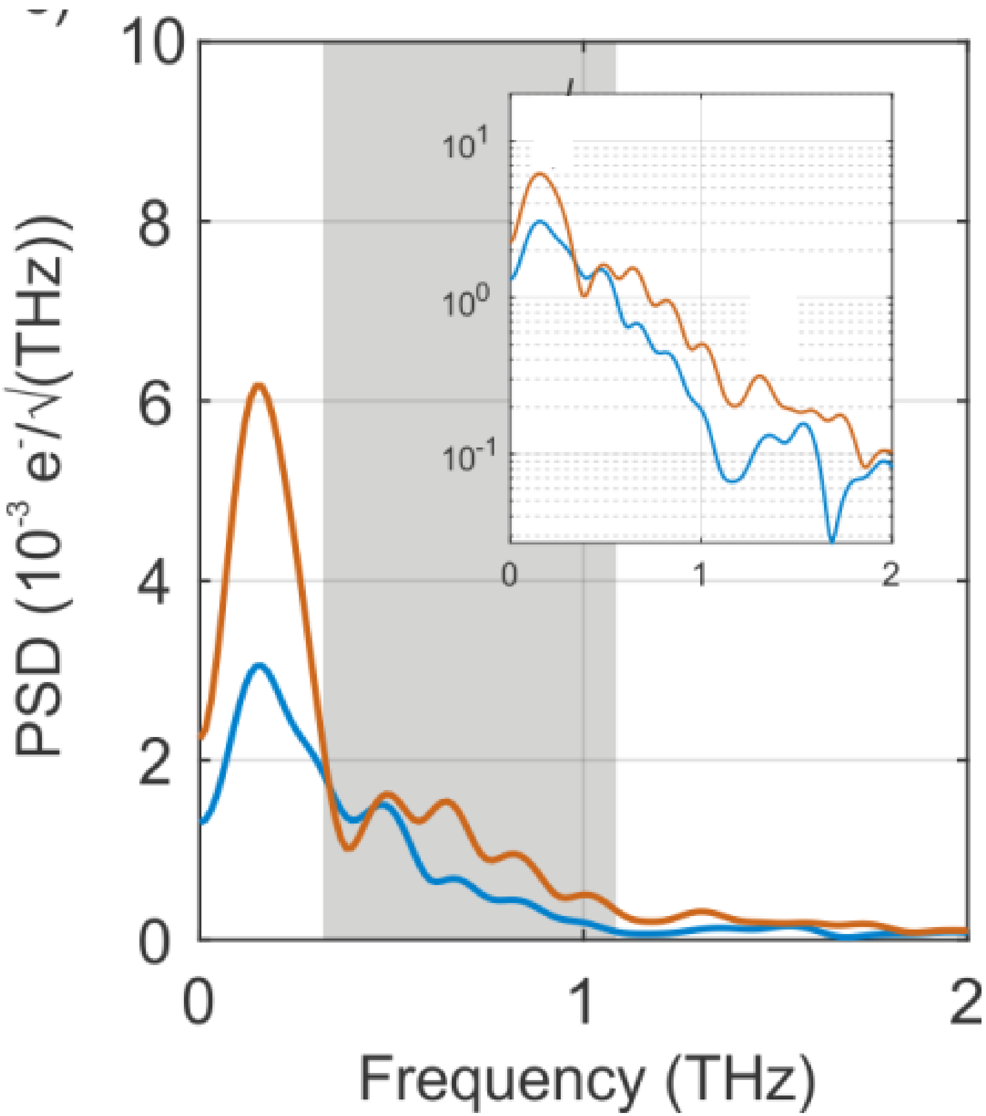
CDW dynamics at atomic pinning site in $NbSe_2$



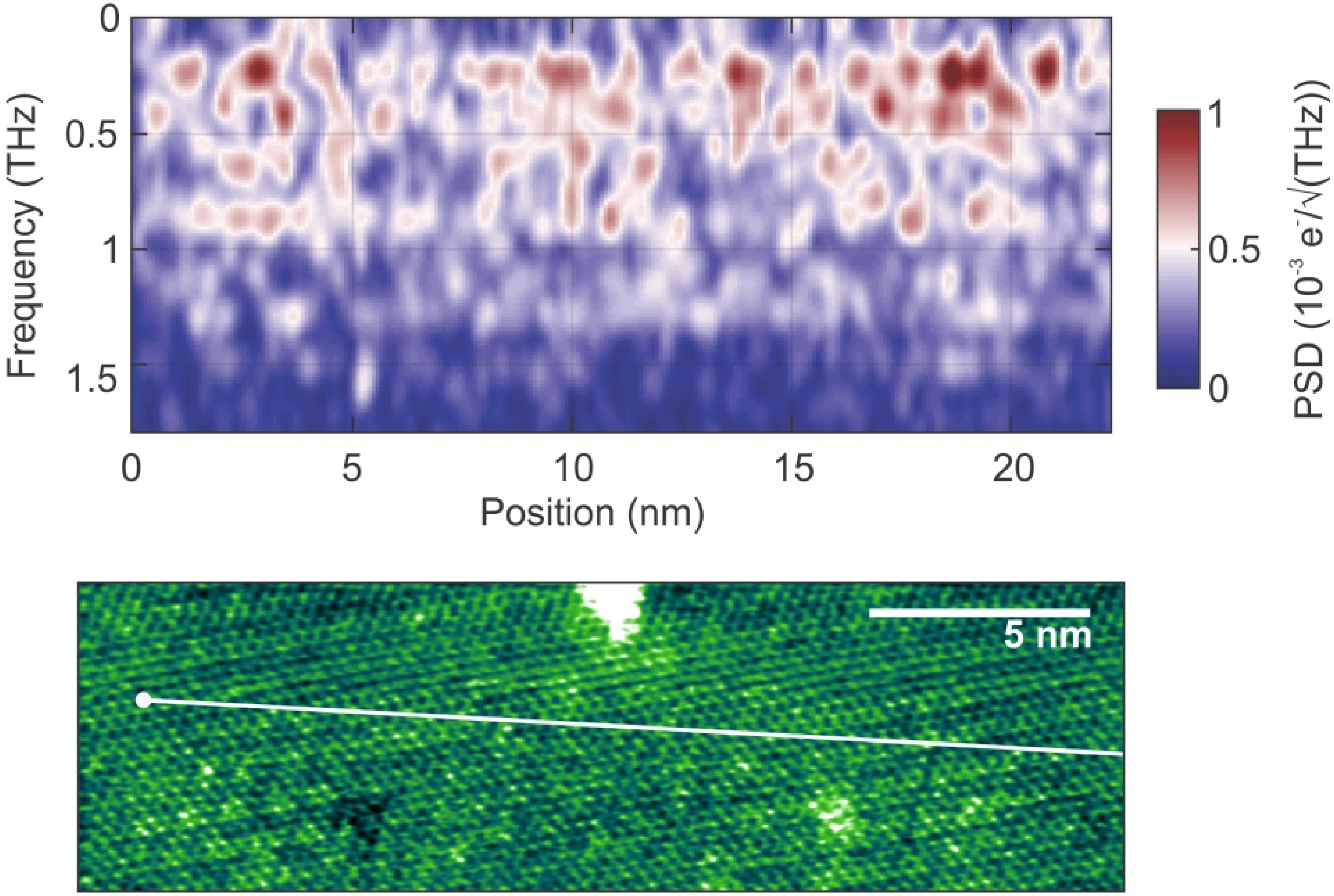


CDW dynamics at atomic pinning site in $NbSe_2$





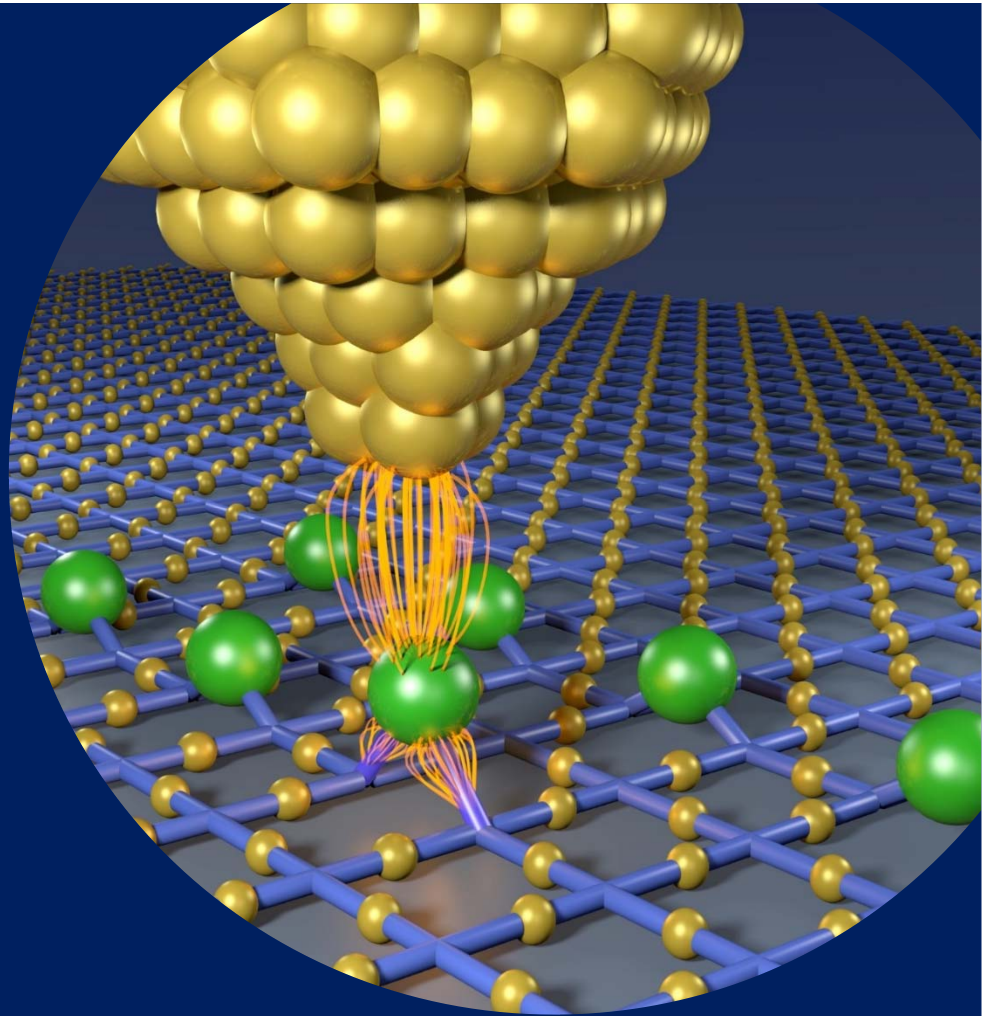
Spatially-resolved CDW dynamics in $NbSe_2$



Ultrafast STM
by microwave & THz excitation

Collective charge density wave dynamics at defects

Atomic-scale magnetic quantum sensing



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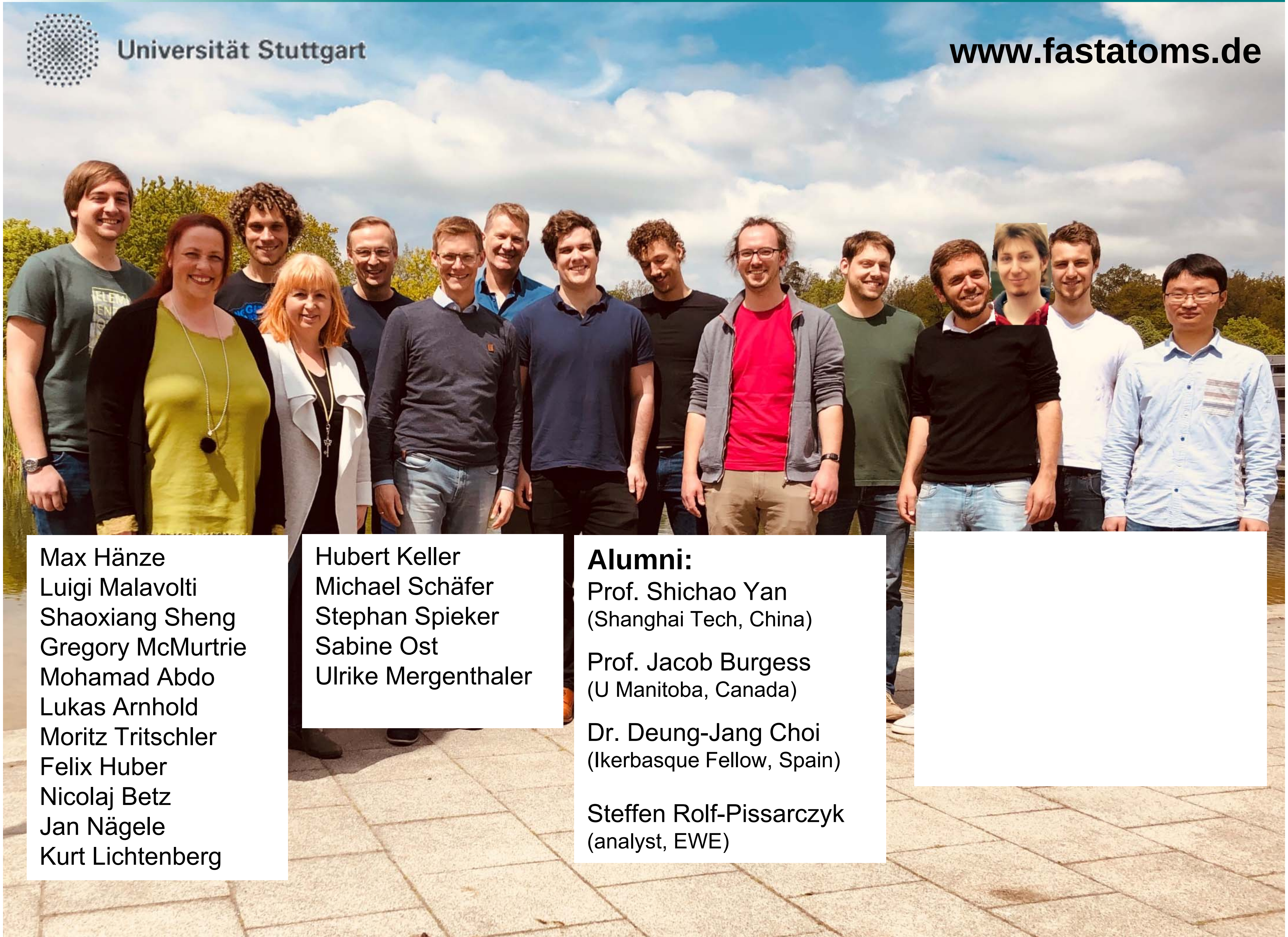

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