



# Scanning tunneling microscopy and its application in studying quantum materials

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Beijing, 2024.05



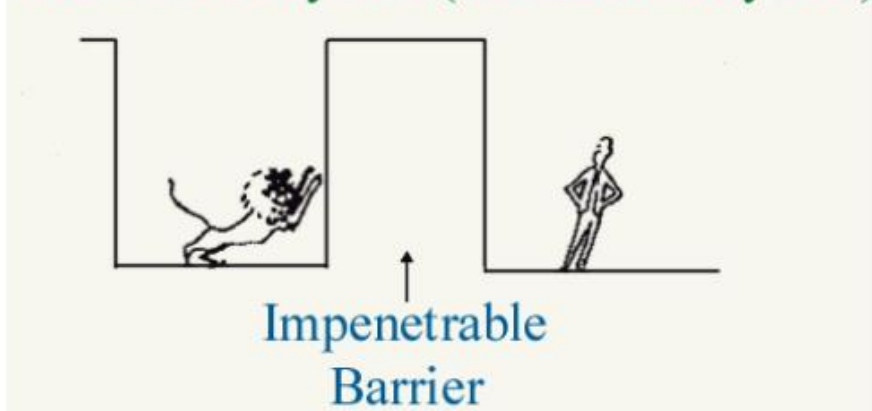
- **Introduction for STM: principles and instrumentation**
- **Application of STM in the study of superconductivity**
- **Inelastic tunneling spectroscopy, spin-resolved STM, time-resolved STM**



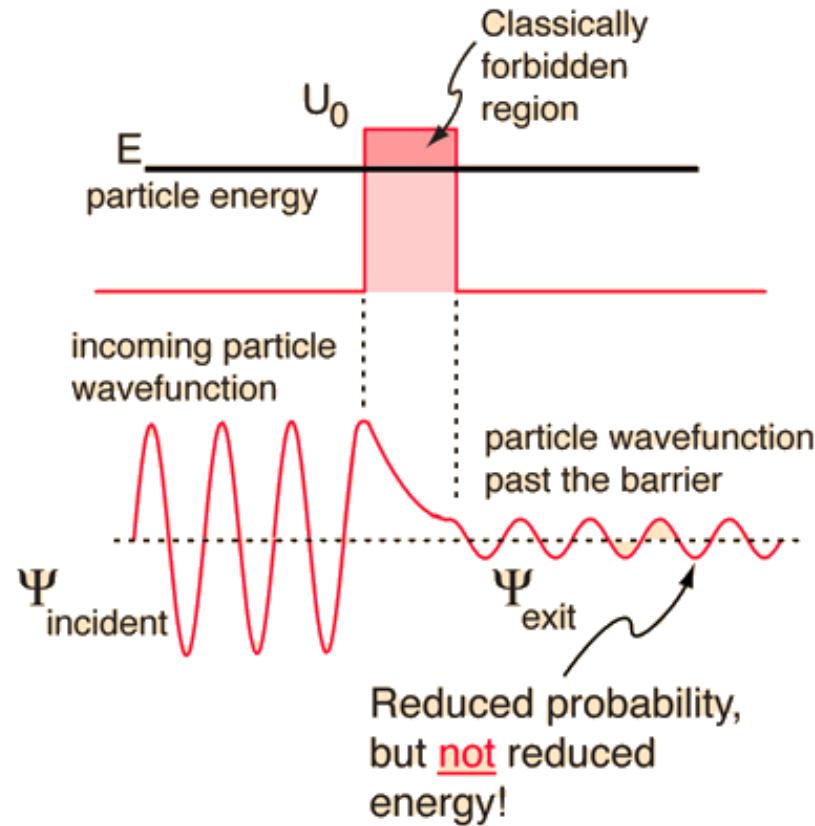
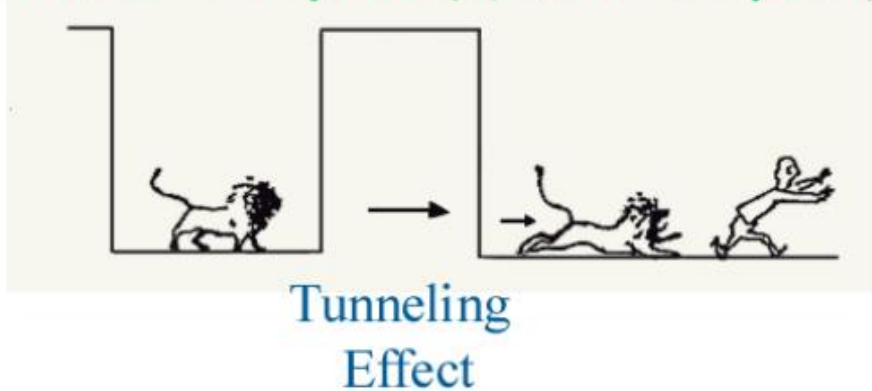
# STM principles: the concept of tunneling



## Pre-1900 Physics (Classical Physics)



## Post-1900 Physics (Quantum Physics)



$$-\frac{\hbar^2}{2m} \frac{\partial^2 \psi(x)}{\partial x^2} = (E - U_0) \psi(x)$$

$$T \approx e^{-2\alpha d}$$

where

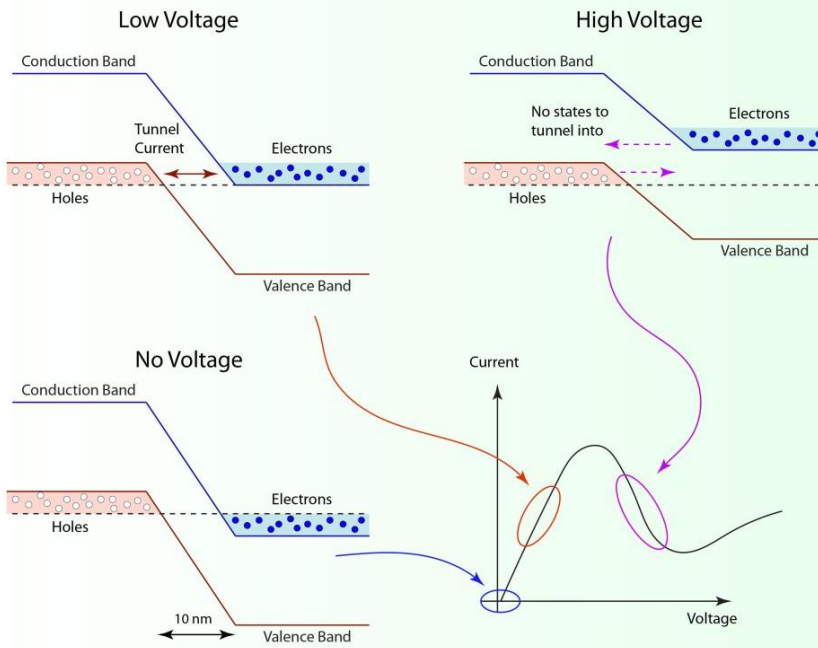
$$\alpha = \sqrt{\frac{2m(U_0 - E)}{\hbar^2}}$$

- Solid-state electron tunneling

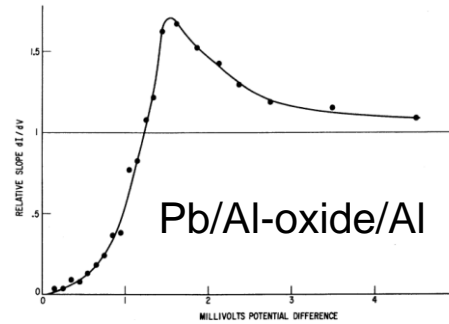


Leo Esaki

Esaki Tunnel Diode



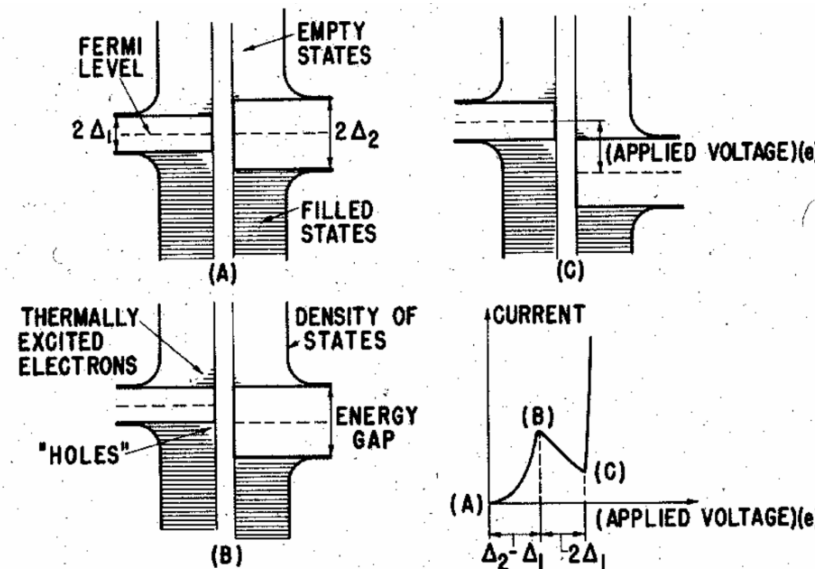
- Superconducting tunneling



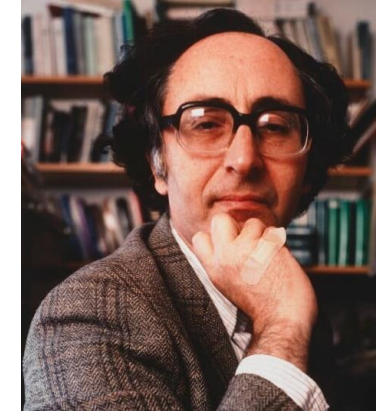
Superconducting gap of lead (at 1.6K)



Ivar Giaever



- Josephson effect



Brian Josephson

**DC effect:**

Supercurrent can flow through insulating layers with no resistance.

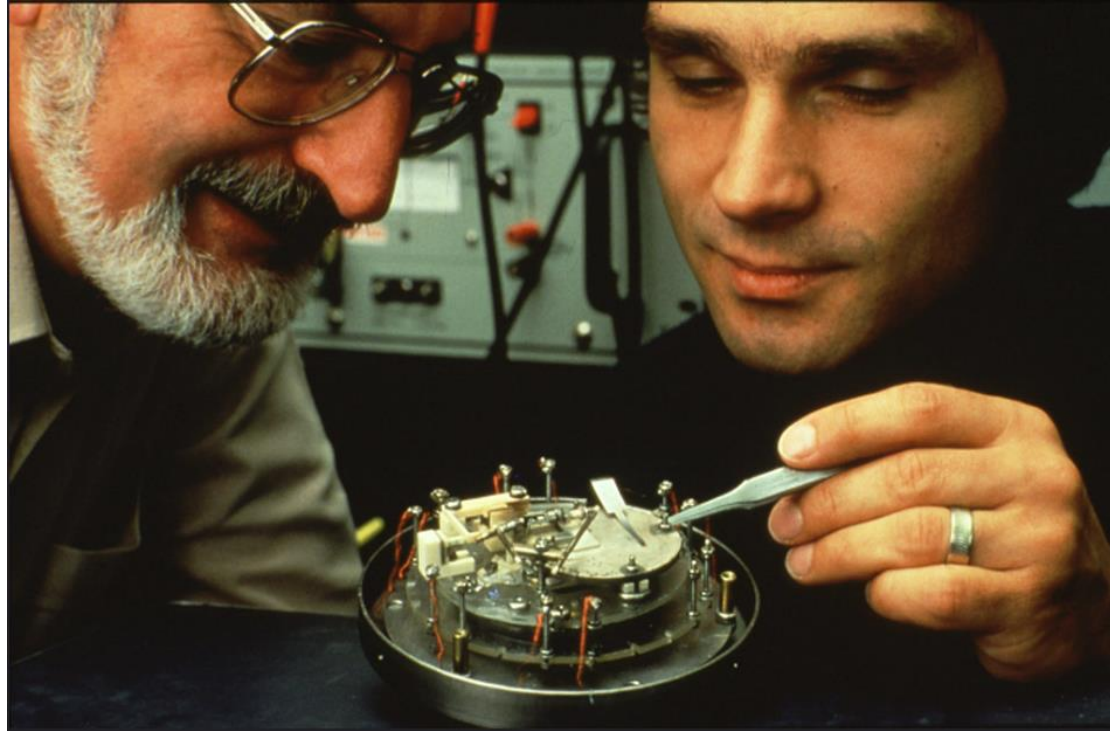
**AC effect:**

With an applied voltage,  $V$ , across the insulating layer, the junction would radiate at a frequency of  $2eV/h$ .

# The birth of STM

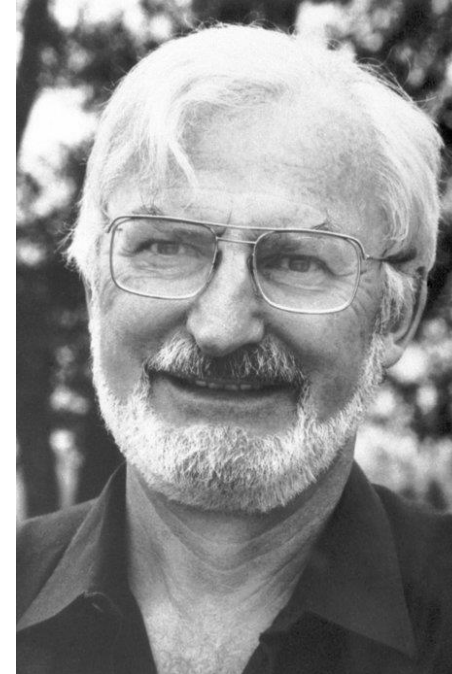


- Invented by Gerd Binnig and Heinrich Rohrer in 1981

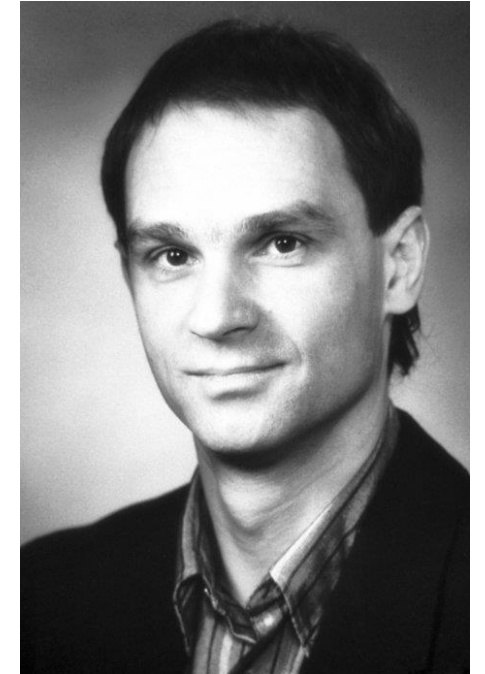


Heinrich Rohrer and Gerd Binnig  
IBM Research – Zurich

## The Nobel Prize in Physics 1986



Heinrich Rohrer  
Prize share: 1/4



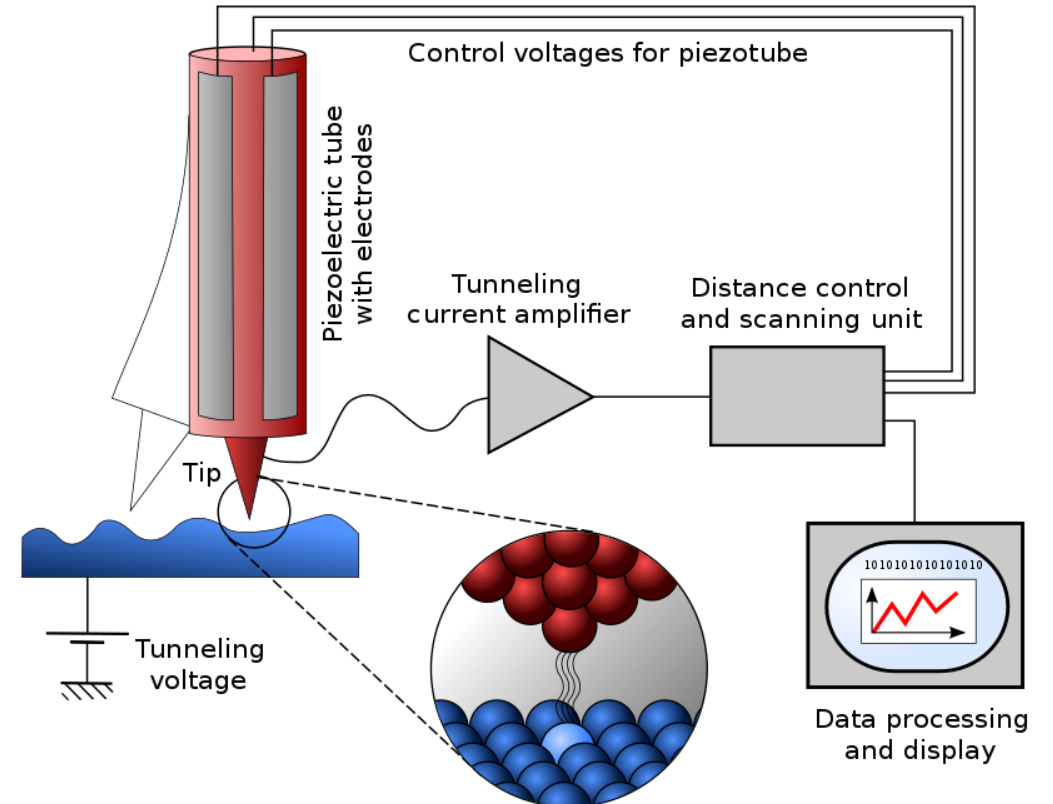
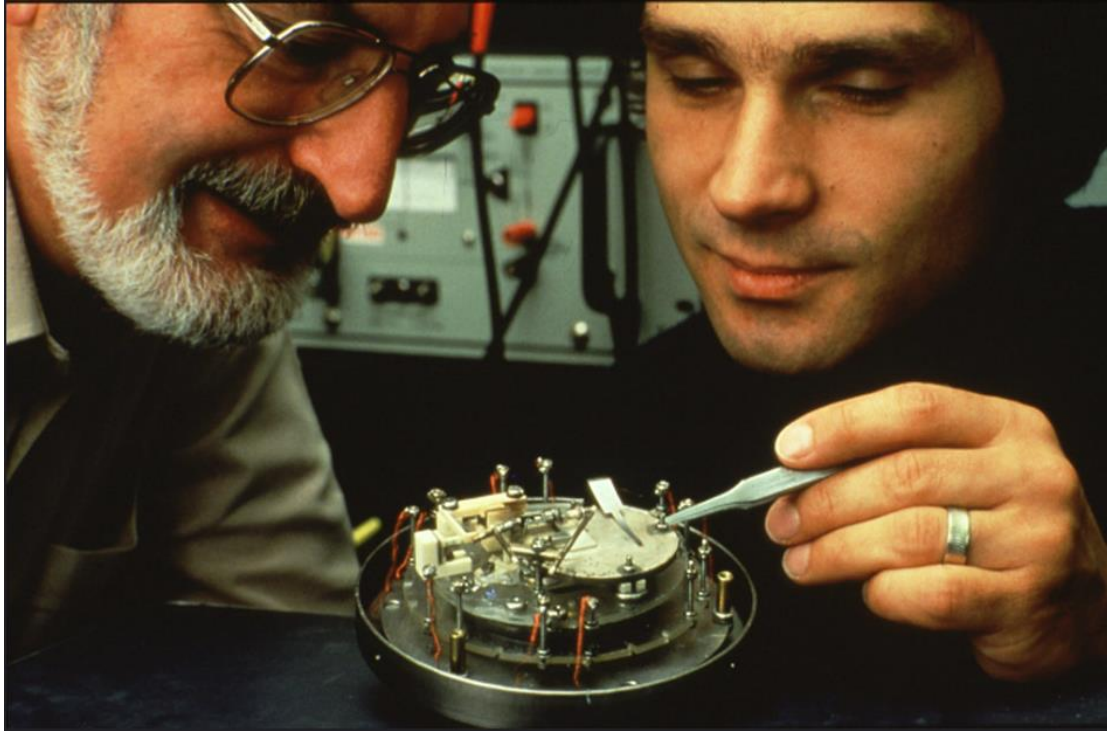
Gerd Binnig  
Prize share: 1/4

"for their design of the scanning tunneling microscope"

# The birth of STM

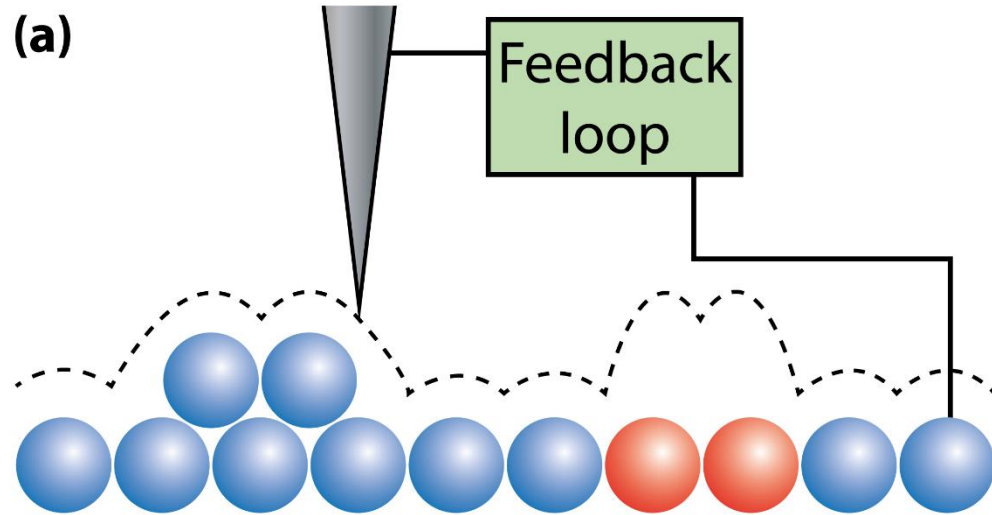


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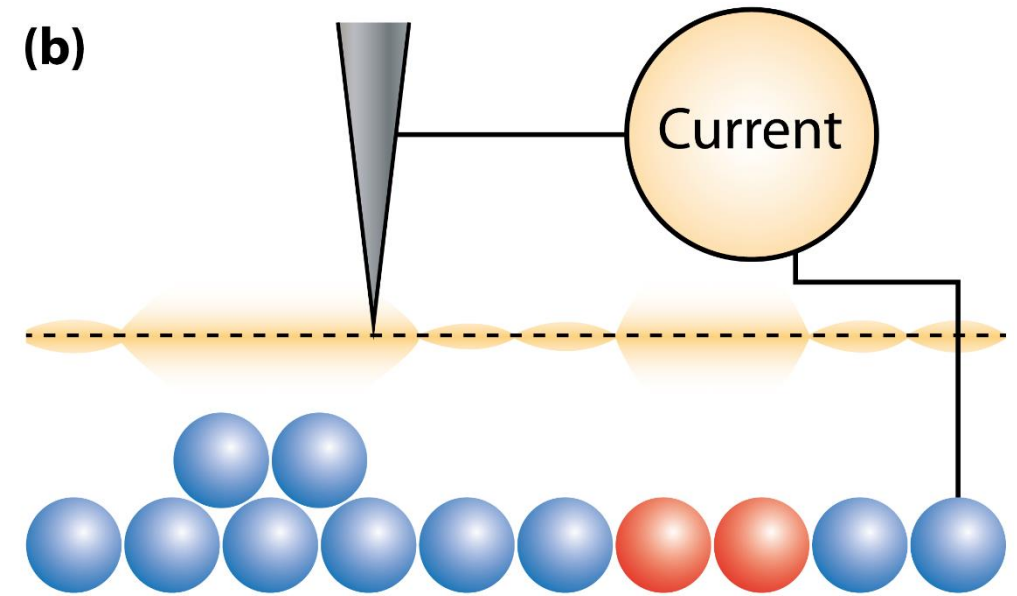
Gerd Binnig and Heinrich Rohrer  
IBM Research – Zurich

- **Constant current mode**

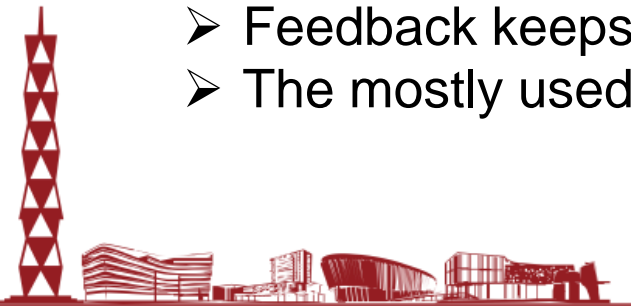


- Feedback keeps current as constant
- The mostly used mode

- **Constant height mode**



- Feedback is disabled
- Only suitable for very flat surface and low thermal drift case



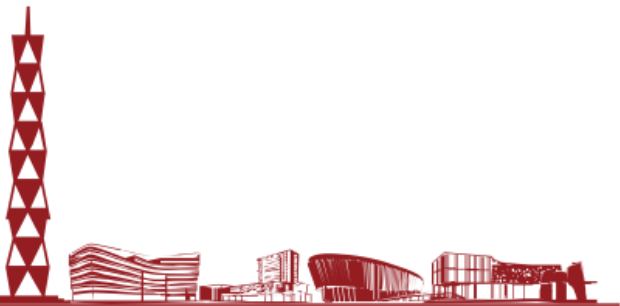
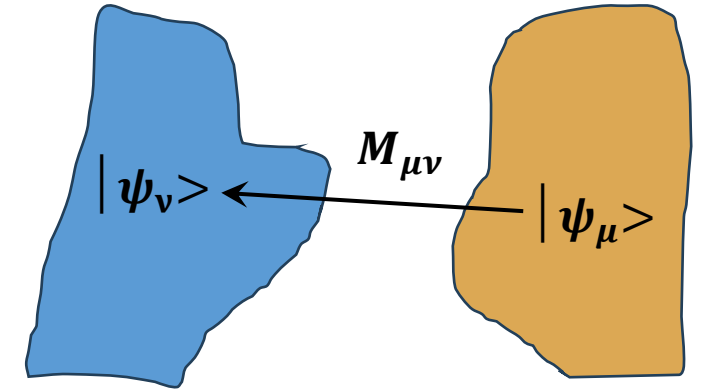
# Bardeen's theory of tunneling



- Tip and sample are considered as two electrodes
- Transmission rate calculated by perturbation
- Neglect tip-sample interaction

- Tunneling matrix: 
$$M_{\mu\nu} = \frac{\hbar^2}{2m} \int d\vec{S} \cdot (\psi_\mu^* \vec{\nabla} \psi_\nu - \psi_\nu \vec{\nabla} \psi_\mu^*)$$

$M_{\mu\nu}$  is the tunneling matrix element between states  $\psi_\mu$  of the tip and  $\psi_\nu$  of the sample surface





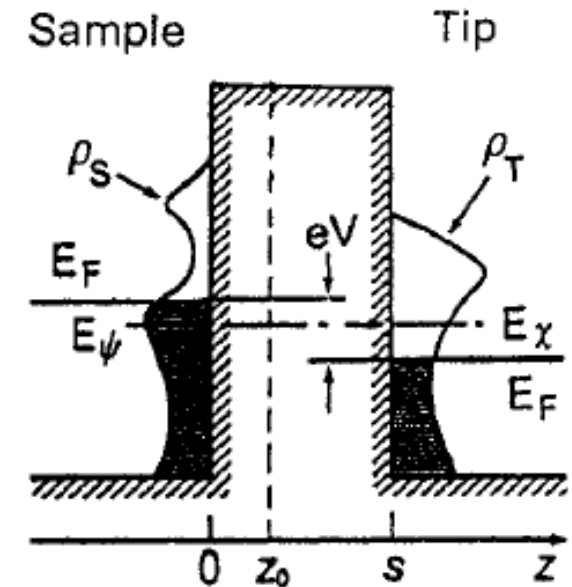
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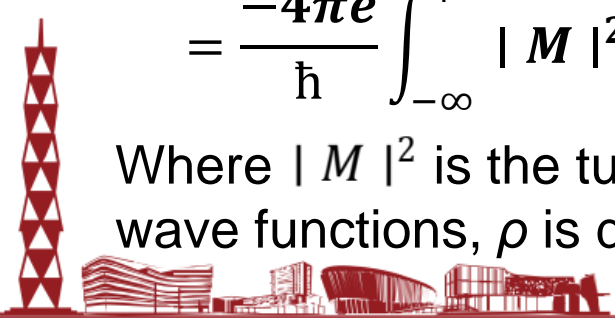


When **Bias Voltage** is applied, the net tunneling current can be described as:

$$I = \frac{-4\pi e}{\hbar} \int_{-\infty}^{+\infty} |M|^2 \rho_s(\epsilon) \rho_t(\epsilon - eV) \{f(\epsilon - eV)[(1 - f(\epsilon))] - f(\epsilon) * [(1 - f(\epsilon - eV))]\} d\epsilon$$

$$= \frac{-4\pi e}{\hbar} \int_{-\infty}^{+\infty} |M|^2 \rho_s(\epsilon) \rho_t(\epsilon - eV) [f(\epsilon - eV) - f(\epsilon)] d\epsilon$$

Where  $|M|^2$  is the tunneling Matrix element, which is proportional to the overlap of tip and sample wave functions,  $\rho$  is density of states (DOS) of sample or tip,  $f(x)$  is the Fermi function.



# Bardeen's theory of tunneling



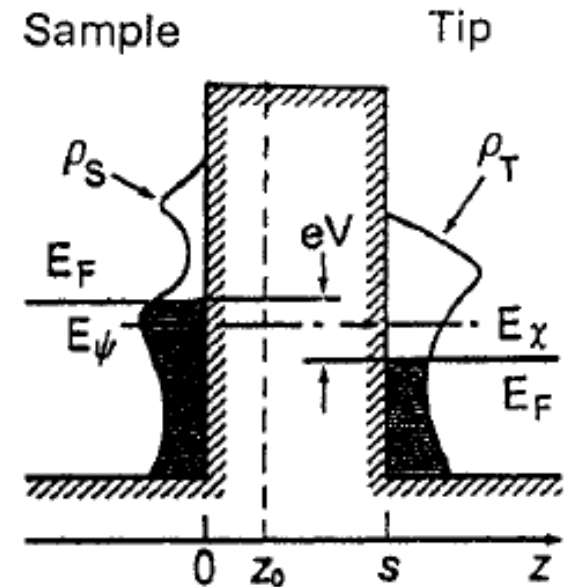
Consider  $T = 0$  K and a simple metal tip with a “flat” DOS:

$$I \approx -\frac{4\pi e}{\hbar} \rho_t(0) \int_0^{+eV} |M|^2 \rho_s(\epsilon) d\epsilon$$

If we further assume the **tunneling matrix is constant**, at  $T = 0$  K:

$$I \propto |M|^2 \int_0^{+eV} \rho_s(\epsilon) d\epsilon \quad \longrightarrow \quad \frac{dI}{dV} \propto \rho_s(\epsilon)$$

When  $T > 0$  K: 
$$\frac{dI}{dV} \propto \int_{-\infty}^{+\infty} \rho_s(\epsilon) f'(\epsilon + eV) d\epsilon$$



- Where  $f'$  is the derivative of the Fermi function.
- **Therefore the tunneling conductance measures the thermally smeared LDOS !**
- Other factors can also broaden  $dI/dV$  spectrum, such as **electrical (RF) noise**.

# Bardeen's theory of tunneling



- Tunneling matrix

$$M_{\mu\nu} = \frac{\hbar^2}{2m} \int d\vec{S} \cdot (\psi_{\mu}^* \vec{\nabla} \psi_{\nu} - \psi_{\nu} \vec{\nabla} \psi_{\mu}^*)$$

$M_{\mu\nu}$  is the tunneling matrix element between states  $\psi_{\mu}$  of the tip and  $\psi_{\nu}$  of the sample surface

- We model the tip as a locally spherical potential well where it approaches nearest to the sample surface

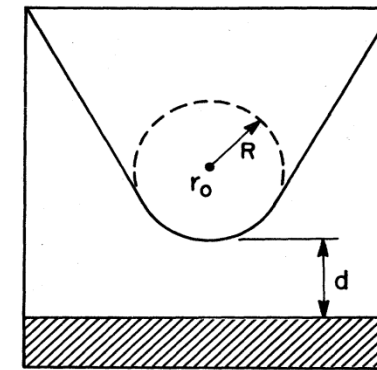
$$M_{\mu\nu} \propto \psi_{\nu}(\vec{r}_0)$$

$$|\psi_{\nu}(\vec{r}_0)|^2 \propto e^{-2\kappa(R+d)}$$

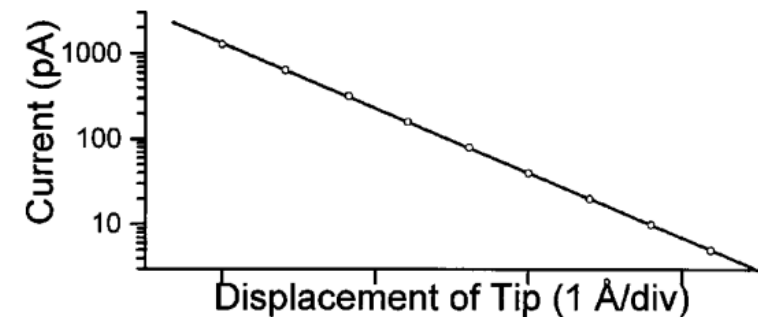
$$I \propto |M|^2 \propto e^{-2\kappa(R+d)} \quad \kappa = \sqrt{\frac{2m\phi}{\hbar^2}}$$

Tunneling current decays exponentially as increasing the tip-sample distance

Tersoff-Hamann Model



Tersoff *et al.*  
PRL 50, 1998  
(1983)



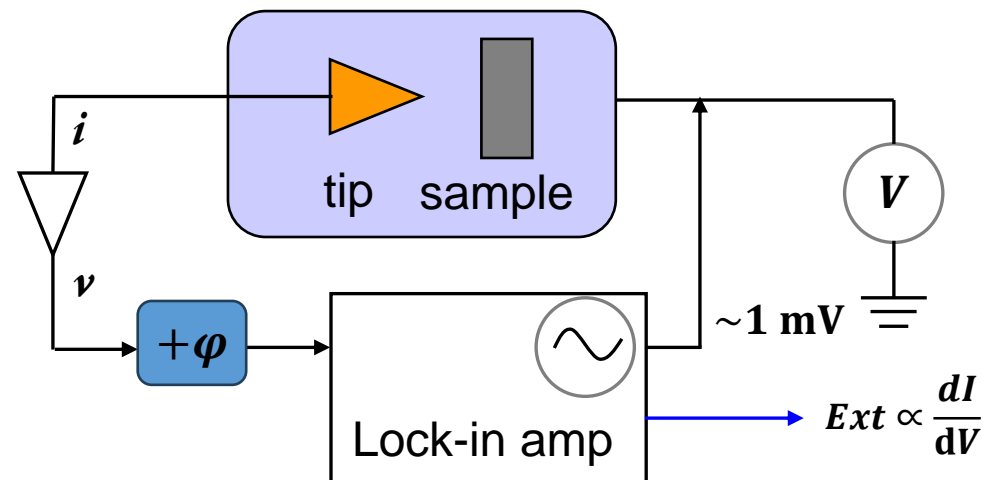
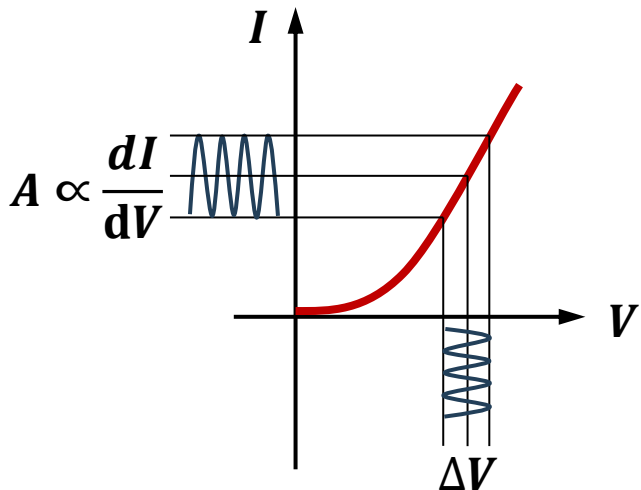
Pan *et al.* APL 73, 2992 (1998)

➤ To obtain high quality  $dI/dV$  spectroscopy, we need:

- A stable tunneling junction (low drift, low vibration)
- A clean and metallic tip (with “flat” DOS)
- Use Lock-in amplifier
- Careful shielding, grounding, and RF filters

➤ Lock-in technique:  $\Delta V = V' \cos(\omega t)$

$$I(V_0 + \Delta V) = I(V_0) + \left. \frac{dI}{dV} \right|_{V_0} V' \cos(\omega t) + \frac{1}{2} \frac{d^2 I}{dV^2} [V' \cos(\omega t)]^2 + \dots$$



- Local density of states spectrum at a single location  $\vec{r}$  is related to the  $\vec{k}$ -space eigenstates  $\psi_k(\vec{r})$  by:

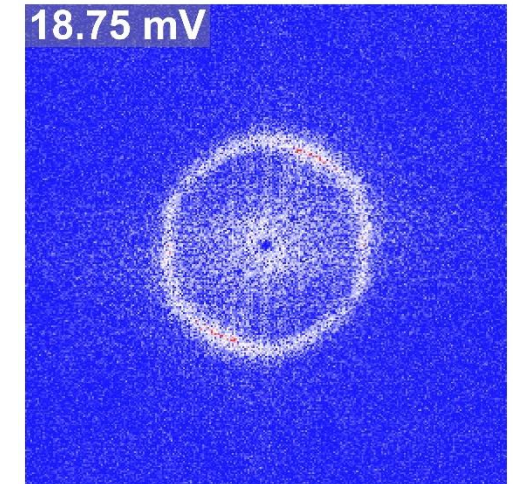
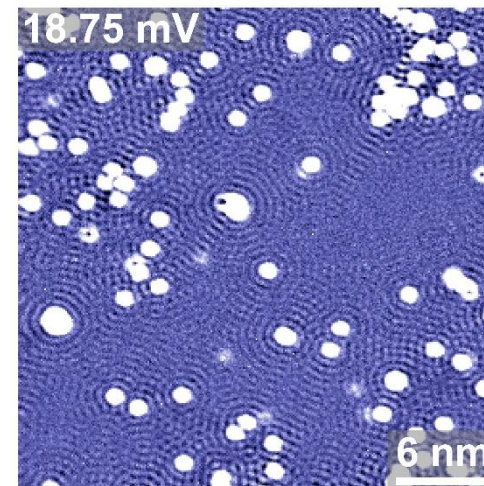
$$\rho_s(E, \vec{r}) \propto \sum_k |\psi_k(\vec{r})|^2 \delta(E - \varepsilon(\vec{k}))$$

- Sources of disorder such as impurities or crystal defects cause **elastic scattering** which mixes eigenstates of different  $\vec{k}$  but the same  $\varepsilon(\vec{k})$ .
- When scattering mixes states  $\vec{k}_1$  and  $\vec{k}_2$ , the result is a **standing wave** in the quasiparticle wavefunction  $\psi_k$  of wavevector  $\vec{q}_{wfn} = (\vec{k}_1 - \vec{k}_2)/2$ .
- LDOS will contain an interference pattern with wavevector  $\vec{q} = (\vec{k}_1 - \vec{k}_2)/2$ , or wavelength  $\lambda = 2\pi/q$ . It is called “**quasi-particle interference**” (QPI).

- In a simple metal, the amplitude of scattering obeys Fermi's golden rule:

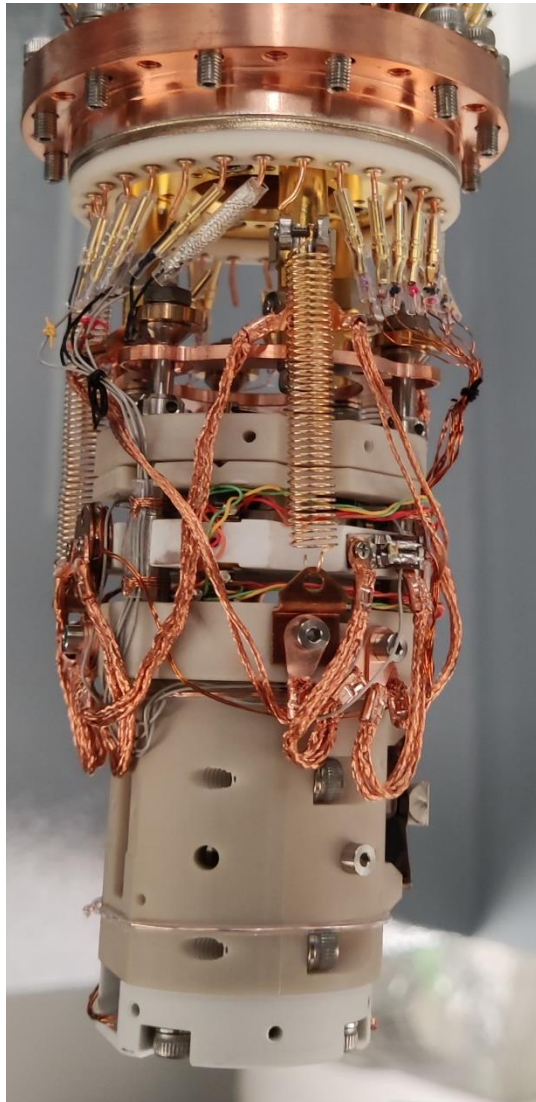
$$\omega(i \rightarrow f) \propto \frac{2\pi}{\hbar} |V(\vec{q})|^2 \rho_i(E_i, \vec{k}_i) \rho_f(E_f, \vec{k}_f)$$

- The scattering vectors can be obtained from the FFT of the QPI, which provide the information from  $\vec{k}$ -space.

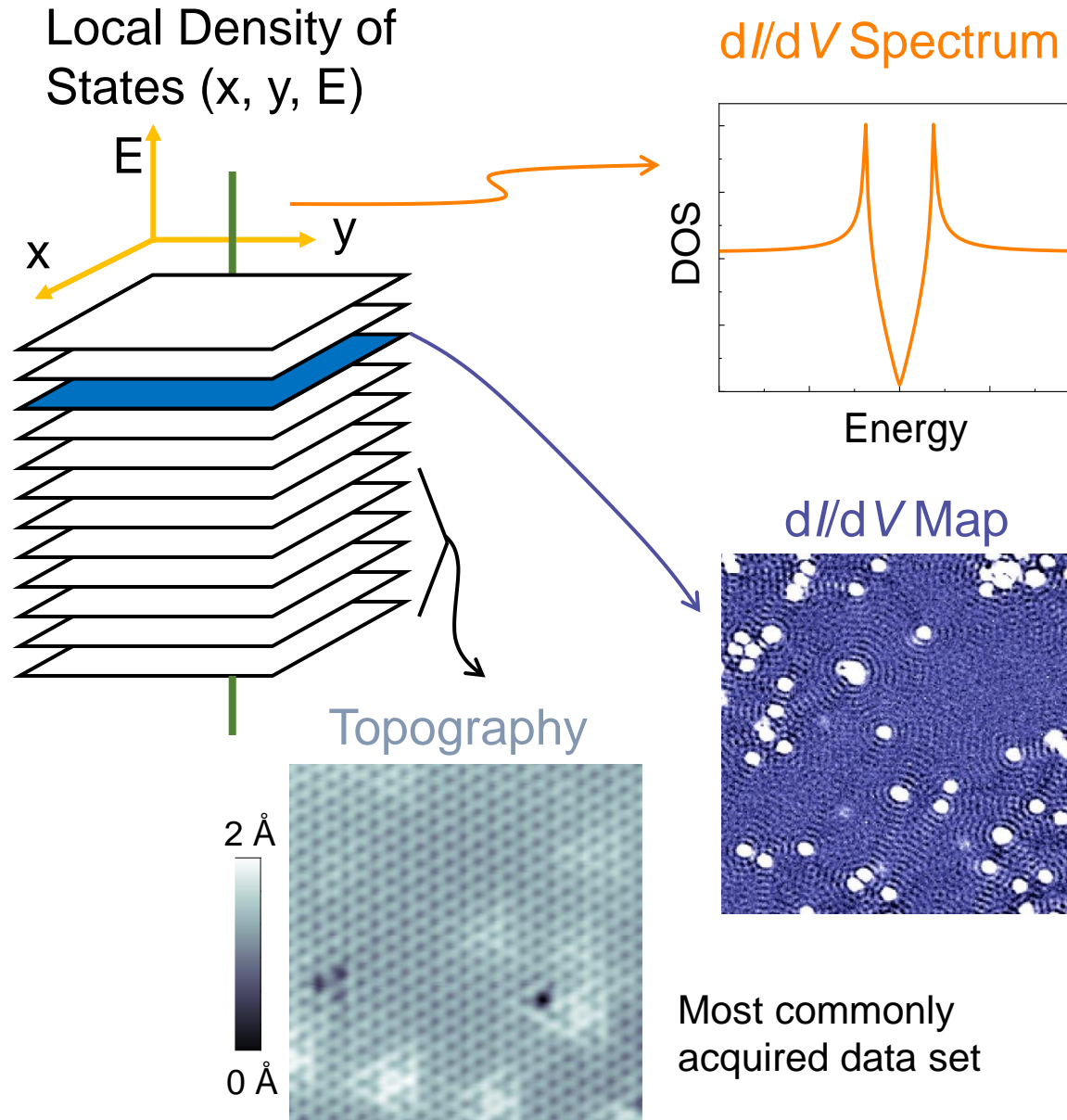


Kong et al. *Nano Lett.* (2022)

# Scanning tunneling microscopy/spectroscopy



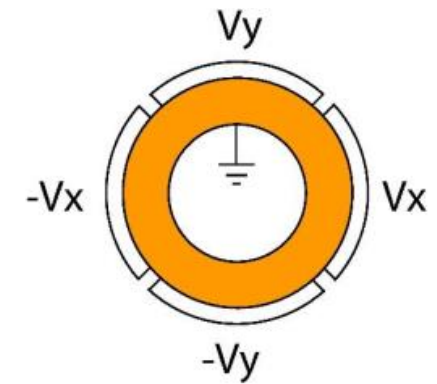
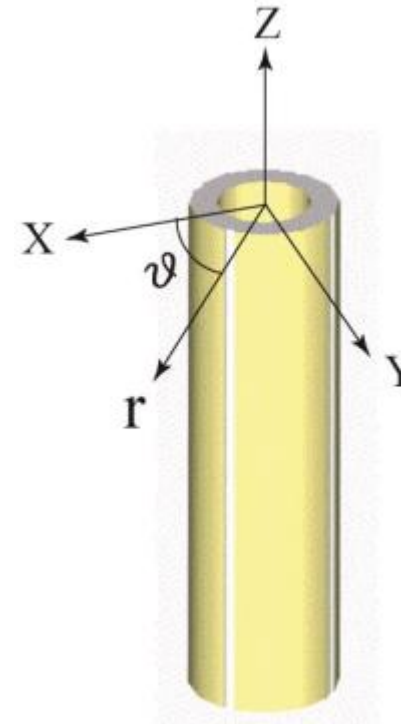
Low temperature STM scanner



# Instrumentation-STM scanner

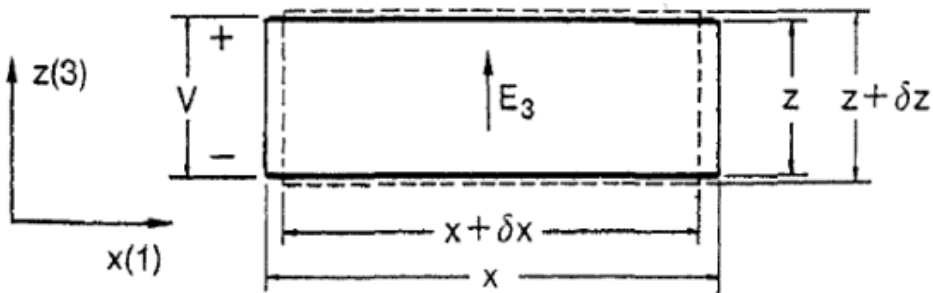


- Piezoelectric ceramics
- Lead zirconate titanate (PZT)

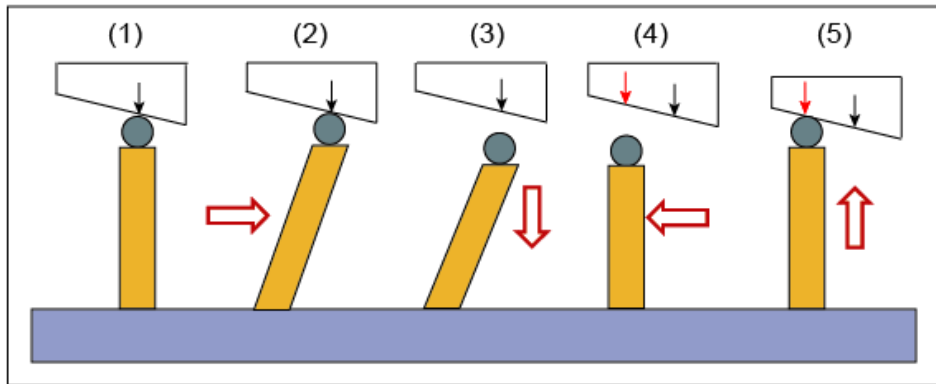
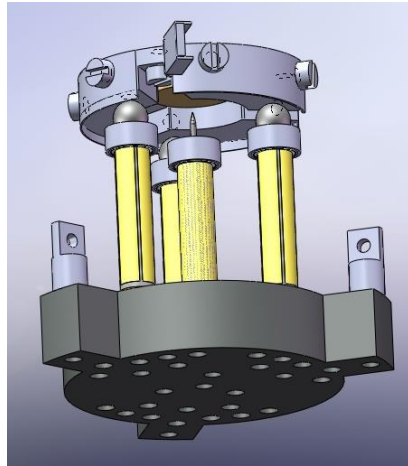


$$\Delta x = 0.9 d_{31} \cdot V \cdot L^2 / D$$

1V~10A

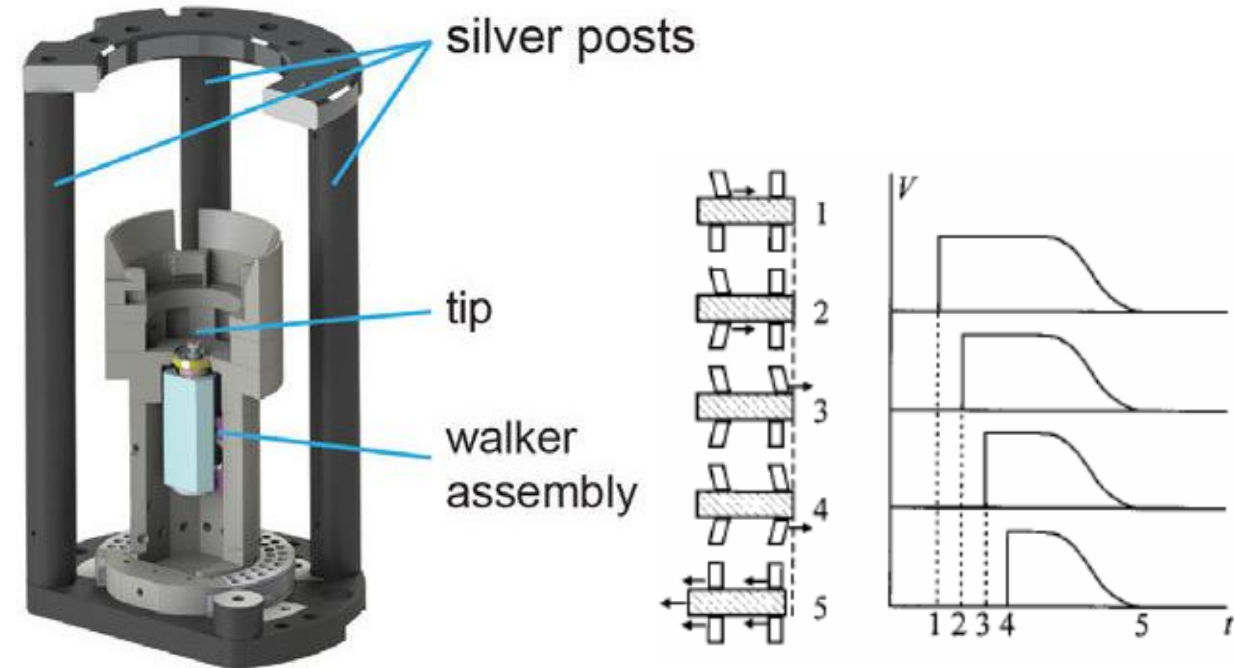


- Besocke design (beetle type)



- **Advantage:** Very low thermal drift, good vibration isolation
- **Disadvantage:** Short travel length of coarse motion

- Pan design (the walker)



- **Advantage:** Compact design, Low thermal drift, easy to integrate with magnets



- Vibration isolation is critical for achieving atomic resolution and high quality spectroscopy  
< 1pm vibration is desired for STM

## Methods:

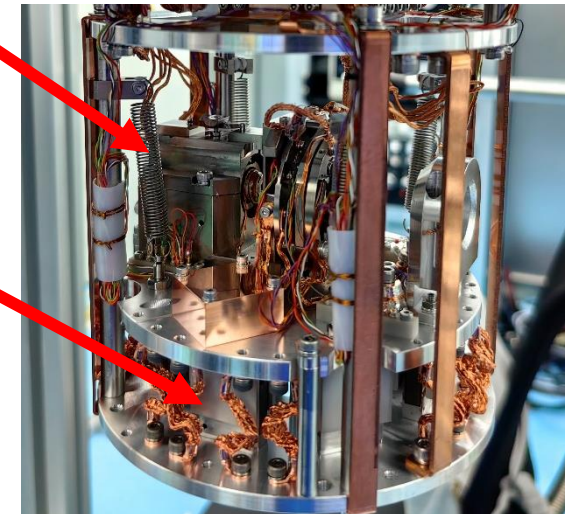
- Basement with solid foundation
- Isolation table
- Spring with damping
- Rigid design
- Turn off Vibration source (pumps)
- Acoustic-isolation room

Isolation table



Spring

Eddy current damper



- Investigate phenomena only occur at low temperature (**Superconductivity**)
- High energy resolution
- Low thermal drift (critical for spectroscopy, mapping)
- Slow down dynamics (diffusion, desorption,...)

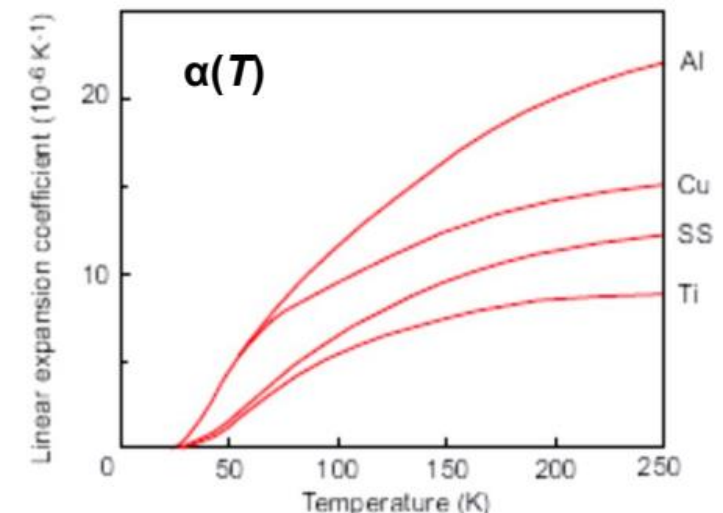
- Energy resolution:  $\frac{dI}{dV} \propto \int_{-\infty}^{+\infty} \rho_s(\varepsilon) f'(\varepsilon + eV) d\varepsilon$

- Thermal broaden:  $\Delta E \approx 3.2 k_B T$

	RT	77 K	4.2 K
$\Delta E$	~83 meV	~21 meV	~1.2 meV

	400 mK	40 mK
$\Delta E$	~0.1 meV	~10 $\mu$ eV

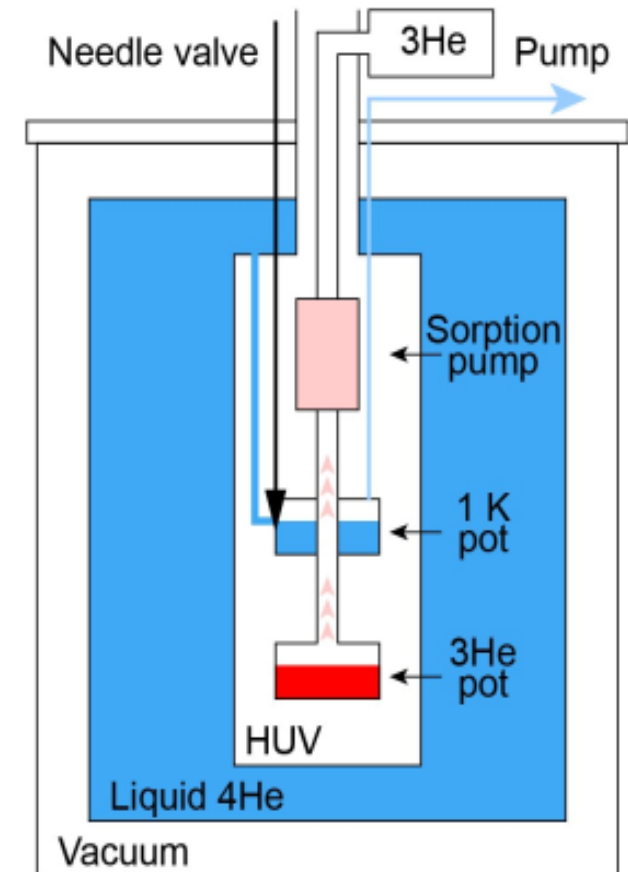
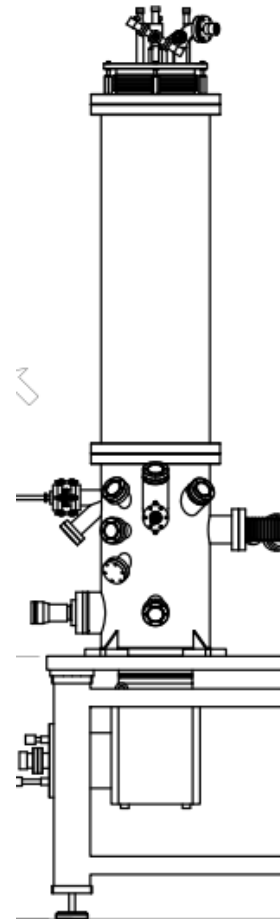
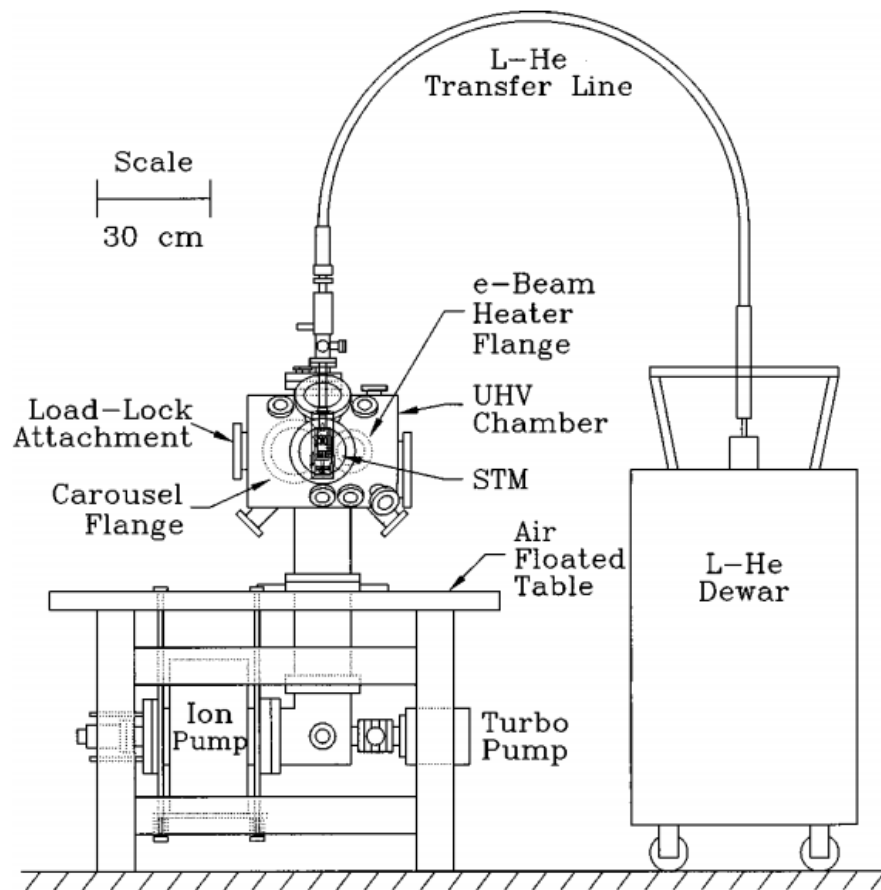
- Thermal drift:  
 $\Delta L = \alpha(T) L \Delta T$



- Continuous flow

- LHe cryostat

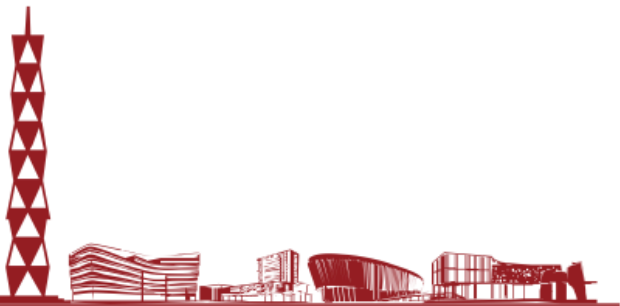
- LHe cryostat with He-3 insert



- Introduction to STM: principles and instrumentation
- Application of STM in the study of superconductivity
- Inelastic tunneling spectroscopy, spin-resolved STM, time-resolved STM



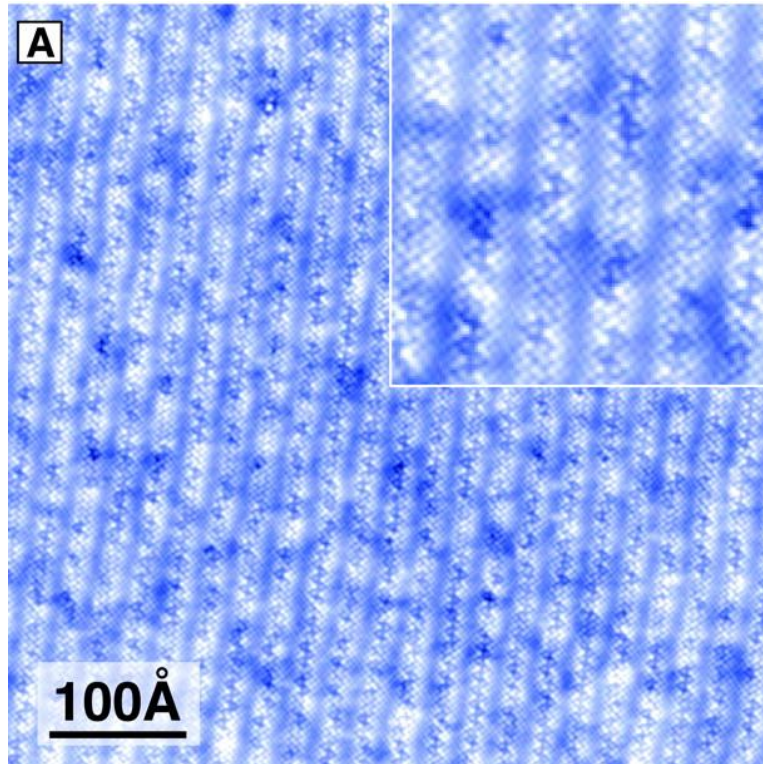
- Surface imaging: atomic structure, reconstruction, domains, defects, *etc...*
- Tunneling Spectroscopy (STS):
  - Measurement of superconducting gap
  - Local states (impurity states, vortex states...)
- Spectroscopy mapping:
  - The imaging of quasi-particle interfere (QPI)
  - Other static orders (CDW, SDW, nematic order...)



# STM in the study of superconductivity

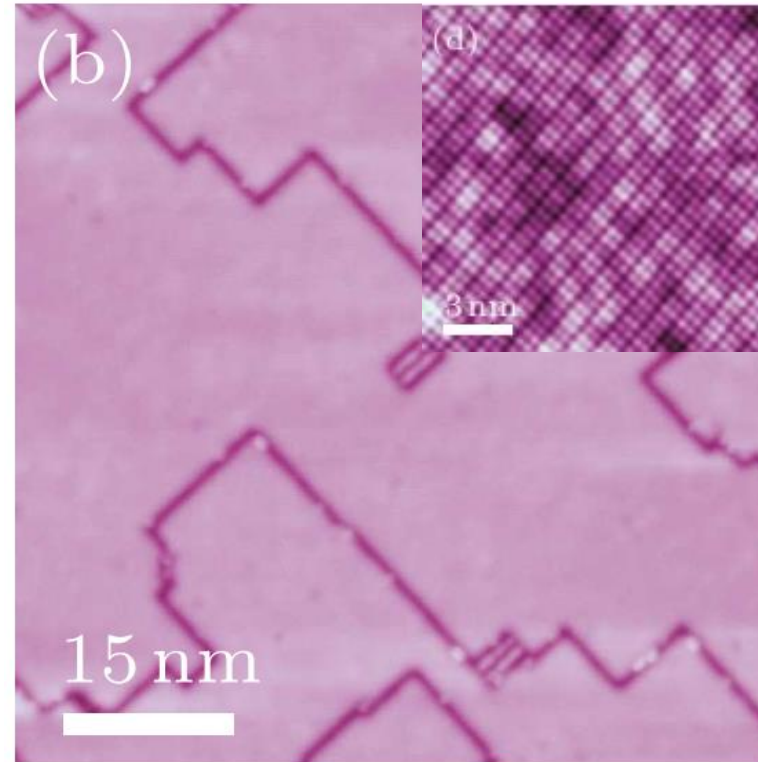


## ➤ Surface imaging



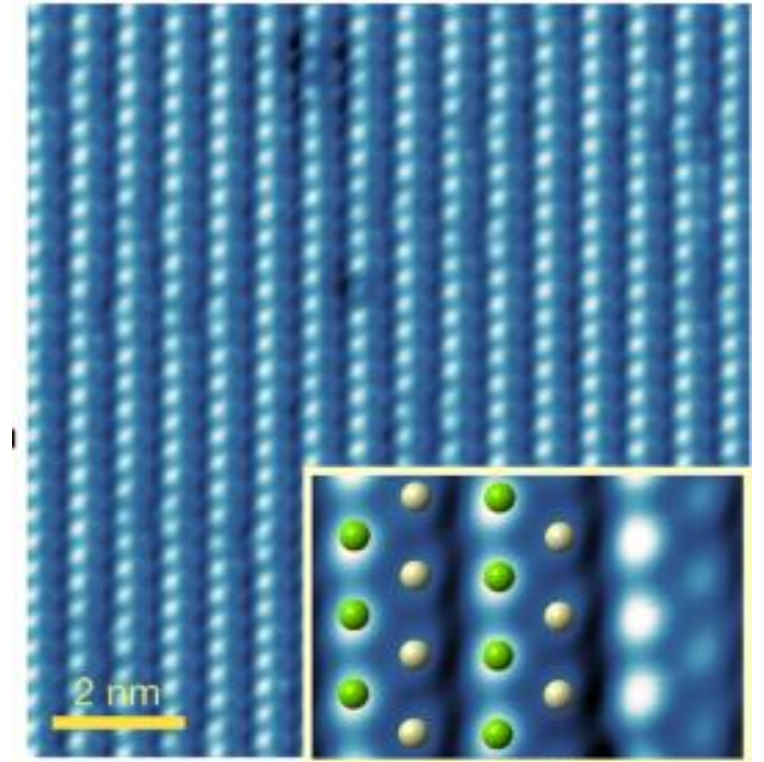
BSCCO (2212)

Hoffman *et al.* *Science* 295, 466 (2002)



FeSe/STO

Wang *et al.* *CPL* 29, 037402 (2012)



UTe<sub>2</sub>

Lin *et al.* *Nature* 579, 523 (2020)

## ➤ Tunneling Spectroscopy: measuring superconducting gap

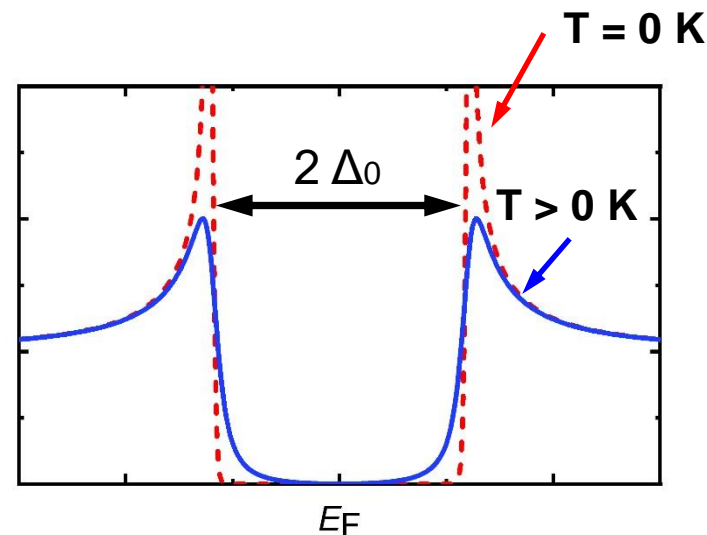
- The gap opening at  $E_F$  in DOS is the important characteristic of superconductivity
- Due to the gap opening, the quasi-particle energy changed to:

$$E_k = \sqrt{\varepsilon_k^2 + \Delta(k)^2}$$

where  $\Delta(k)$  is the SC gap or order parameter

- DOS is given by:  $\rho_\varepsilon = \text{Re} \left( \frac{|E|}{\sqrt{(E^2 - \Delta(k)^2)}} \right)$  where  $\text{Re}$  indicates the real part of the expression that follows

- For a isotropic s-wave superconductor,  $\Delta(k)$  is independent of  $k$ :  $\Delta(k) = \Delta_0$ . Which means the Fermi surface is **fully gapped**.



The DOS of a isotropic s-wave superconductor

## ➤ Tunneling Spectroscopy: measuring superconducting gap

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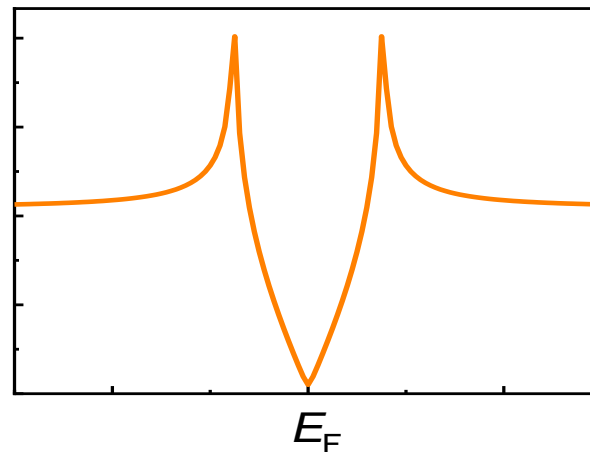
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- For a d-wave superconductor, the order parameter is:

$$\Delta(k) = \Delta_0 \cos(2\theta)$$

$$\left( \tan \theta = \frac{k_x}{k_y} \right)$$



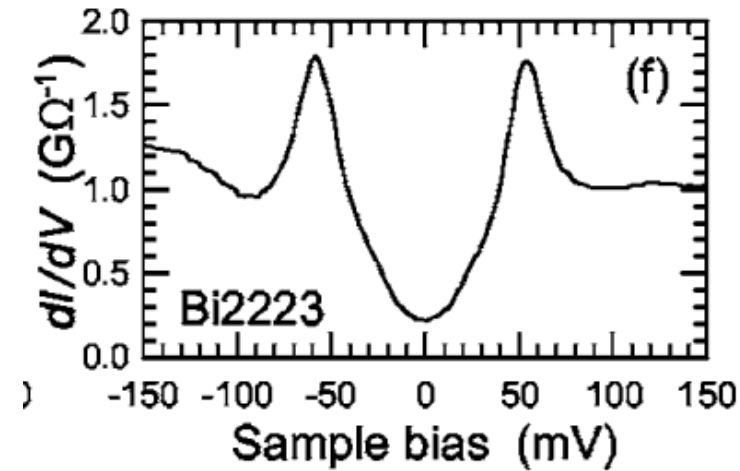
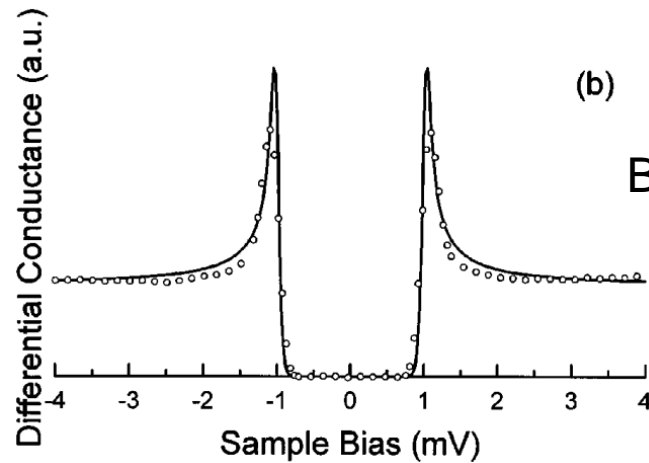
The DOS of a d-wave superconductor  
It is not fully gapped, and the V-shaped  $dI/dV$  implies the existence of nodes in the SC gap.



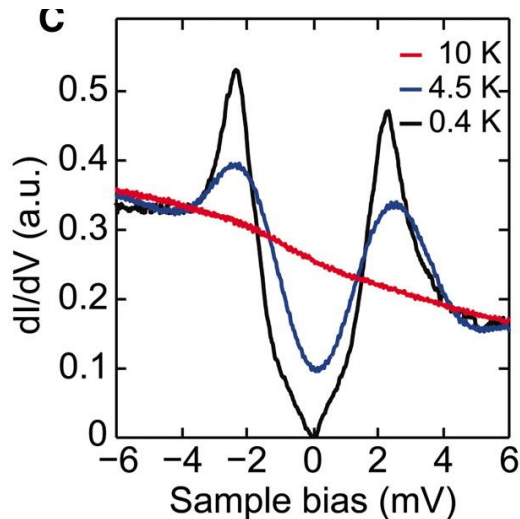
# STM in the study of superconductivity



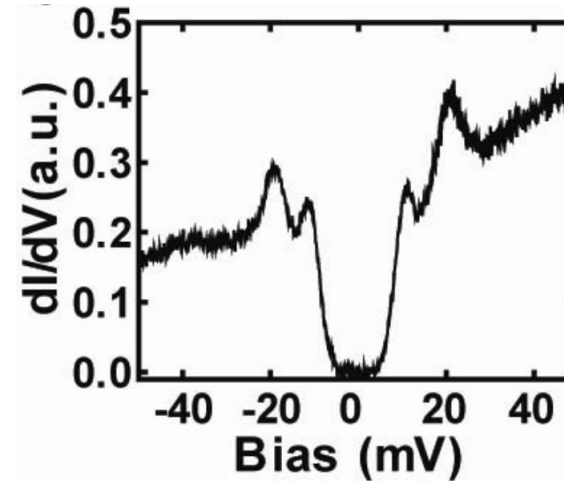
## ➤ Tunneling Spectroscopy: measuring superconducting gap



Cuprate (d-wave)



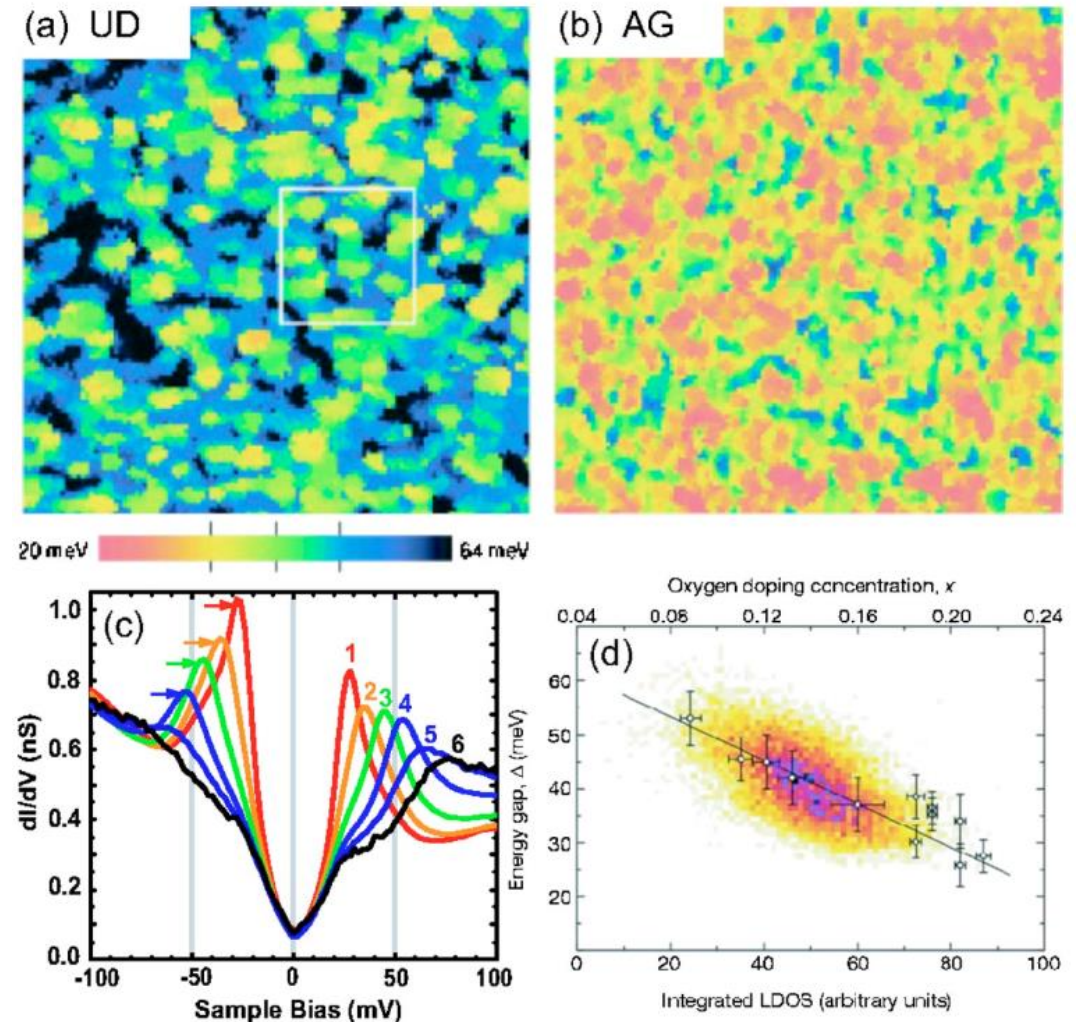
FeSe films (nodal)



Single layer FeSe/STO  
(nodeless)

## ➤ Spatial variation of SC gap

- As a real space probe, STM is capable of detecting the spatial dependence of the gap.
- The spatial inhomogeneity of the SC gap may be attribute to:
  - ✓ Doping inhomogeneity
  - ✓ Low sample quality
  - ✓ Intrinsic factors

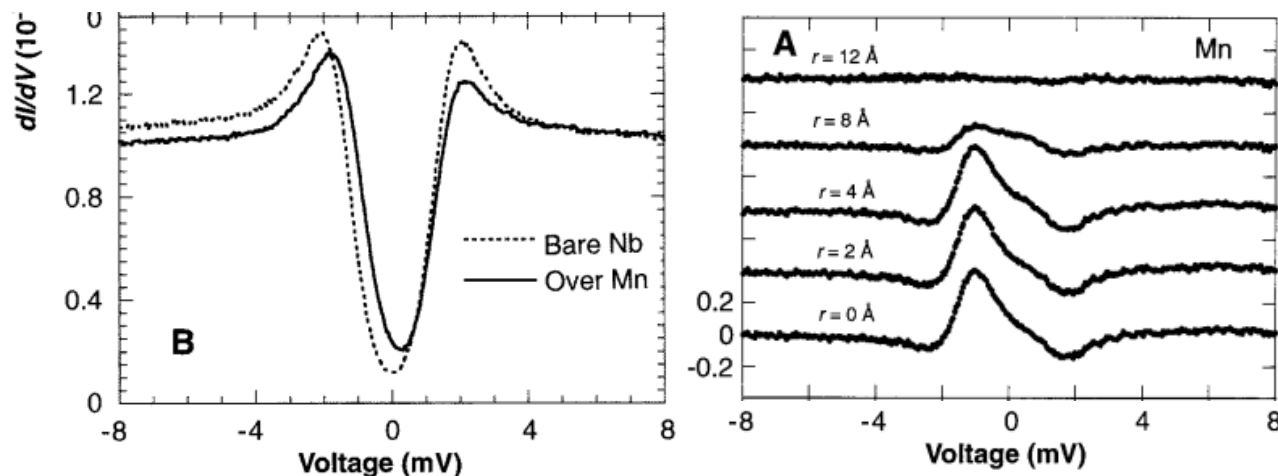


## ➤ In-gap states

### Anderson's theorem:

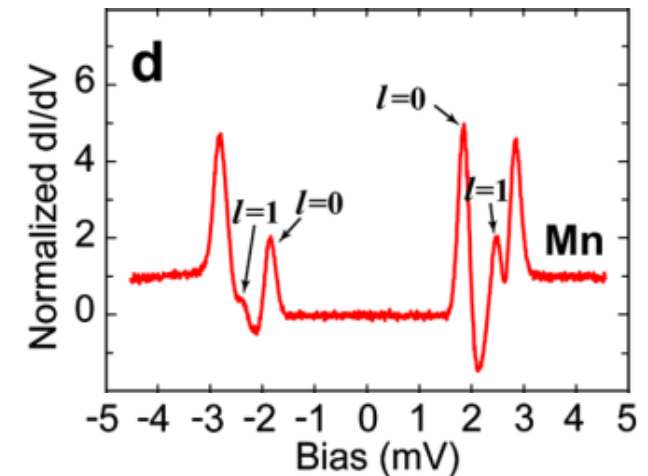
- For a s-wave superconductor, the bound states that generated by **potential scattering** (Non-magnetic scattering) are on the gap edge. Therefore non-magnetic impurities do not suppress the s-wave superconductivity.
- For **magnetic impurities**, which can flip the spin of paired electrons, will suppress the superconductivity and introduce bound states inside of the superconducting gap.

Mn impurity in Nb



Yazdani *et al.* *Science* 275, 1767 (1997)

Mn impurity in Pb

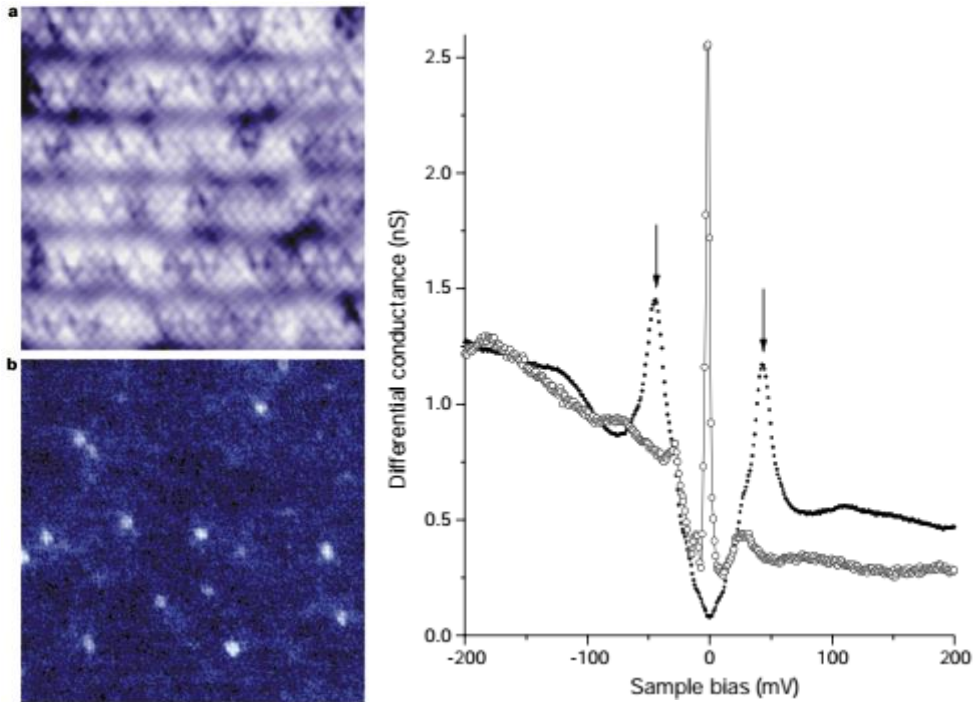


Ji *et al.* *PRL* 100, 226801 (2008)

## ➤ In-gap states

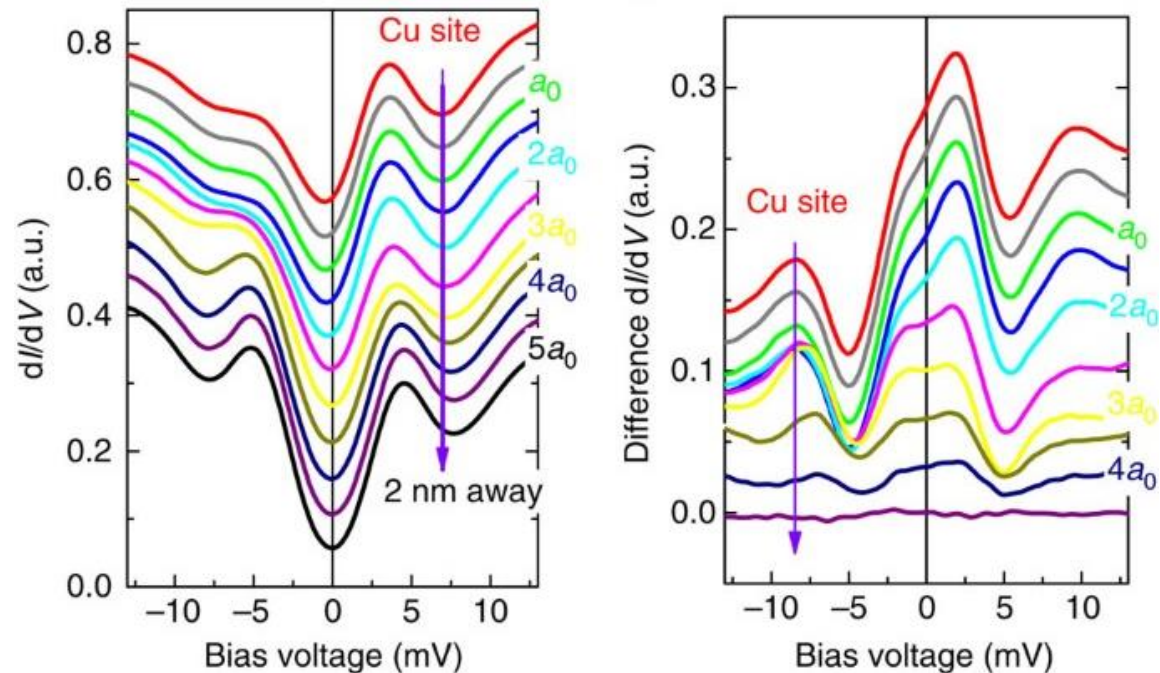
- For phase change pairing (such as d-wave, s<sup>±</sup>), nonmagnetic impurities can also suppress superconductivity.

Zn in BSCCO



Pan *et al.*, *Nature* 403, 746 (2003)

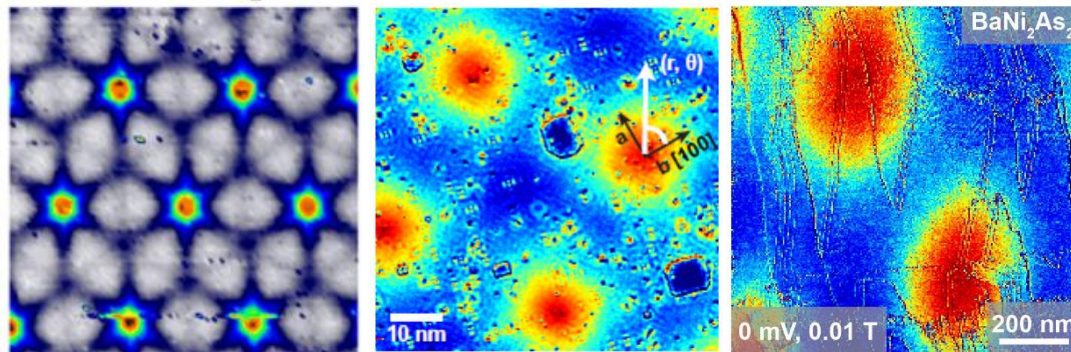
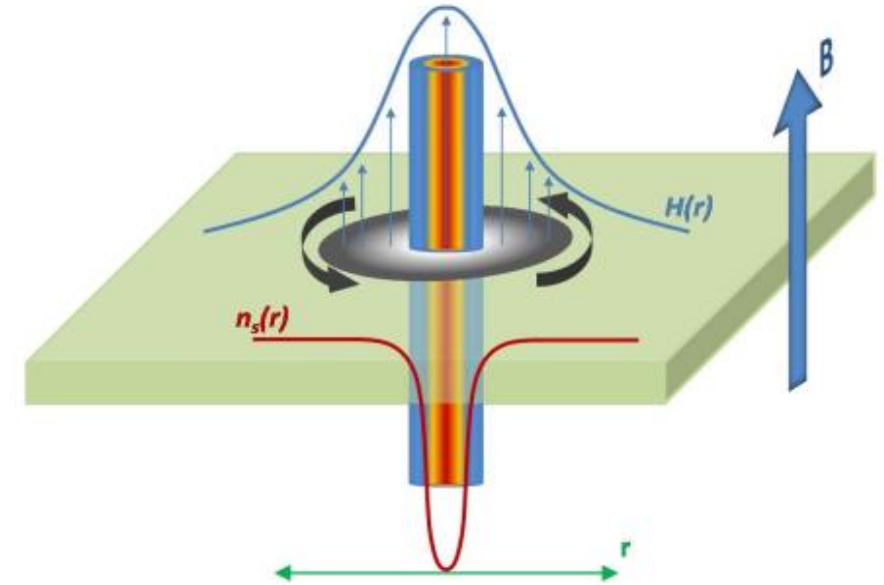
Cu in NaFeAs



Yang, H. *et al.*, *Nat. Comm.* 4, 2749 (2013)

## ➤ Vortex imaging

- For type II superconductor, the  $\mathbf{B}$  field can penetrate into the bulk in the form of Vortices.
- Supercurrent circulates around the normal core of the vortex.
- The core has a size the superconducting coherence length  $\xi$  (parameter of a Ginzburg–Landau theory).
- The supercurrents decay on the distance about  $\lambda$  (London penetration depth) from the core.
- For type-II superconductor,  $\lambda > \xi/\sqrt{2}$
- Vortex with quantized flux of  $\Phi_0 = h/2e = 2.07 \times 10^{-15}$  Wb



## ➤ Mapping vortex can provide useful information:

- Coherence length ( $\xi$ )
- Vortex pinning and dynamics
- The shape of the vortex core reflects the anisotropy of  $\Delta(\mathbf{k})$

## ➤ Vortex core states

Volume 9, number 4

PHYSICS LETTERS

1 May 1964

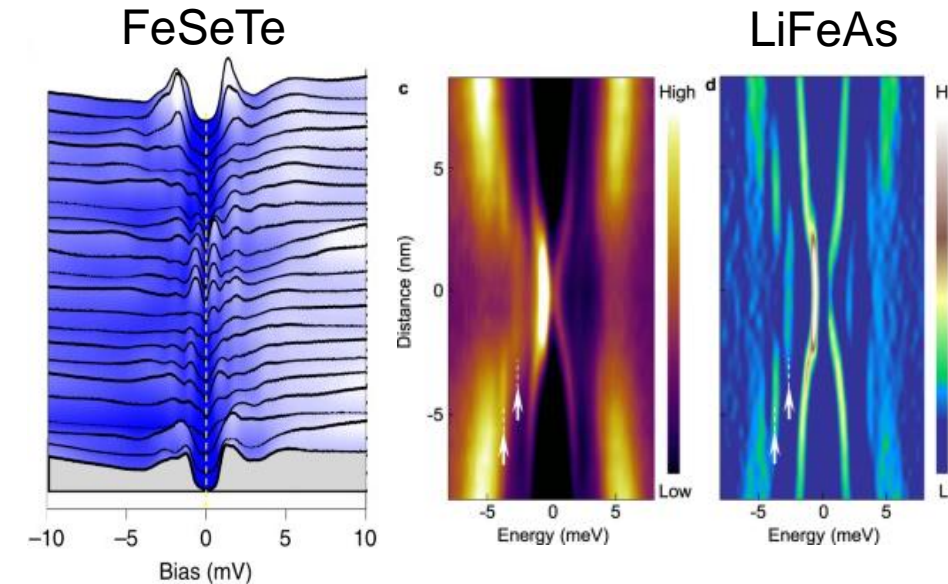
### BOUND FERMION STATES ON A VORTEX LINE IN A TYPE II SUPERCONDUCTOR

C. CAROLI, P. G. DE GENNES, J. MATRICON  
*Service de Physique des Solides, Faculté des Sciences, Orsay (S & O)*

Received 31 March 1964

This note discusses the excitations of low energy  $\epsilon \ll \Delta_\infty$  (where  $\Delta_\infty$  is the gap in zero field) which exist near an Abrikosov vortex line <sup>1)</sup> in a pure superconductor of type II. **The energy gap  $\epsilon_0$  for these excitations is very small  $\epsilon_0 \sim \Delta_\infty^2/E_F$  where  $E_F$  is the Fermi energy.** Above  $\epsilon_0$  the density of states is finite and comparable to that of a cylinder of normal metal of radius  $\xi$  (the coherence length). These low lying states will play a major role in the discussion of transport and relaxation phenomena in type II superconductors at low temperatures.

- Due to the confinement to the quasiparticles by the vortex core, Caroli-de Gennes-Matricon (CdGM) predicted that there are confined low-energy bound states with the energy levels at about  $E_\mu = \pm \mu\Delta^2/E_F$  ( $\mu = 1/2, 3/2, 5/2, \dots$ ) with  $\Delta$  the superconducting energy gap and  $E_F$  the Fermi energy.



Chen *et al.*, Nature Commun. (2018)

Kong *et al.*, Nature Commun. (2021)

- The energy of core states relies on the pairing symmetry.
  - For fully gapped s-wave, the core states are discrete
  - For d-wave, the core states are expected to be continuous
  - For p-wave, the core states are at Zero energy

## ➤ QPI in SC: Bogoliubov quasiparticles

$$\omega(i \rightarrow f) \propto \frac{2\pi}{\hbar} |\mathbf{u}_{ki}\mathbf{u}_{kf}^* \pm \mathbf{v}_{ki}\mathbf{v}_{kf}^*|^2 |V(\vec{q})|^2 \rho_i(E_i, \vec{k}_i) \rho_f(E_f, \vec{k}_f)$$

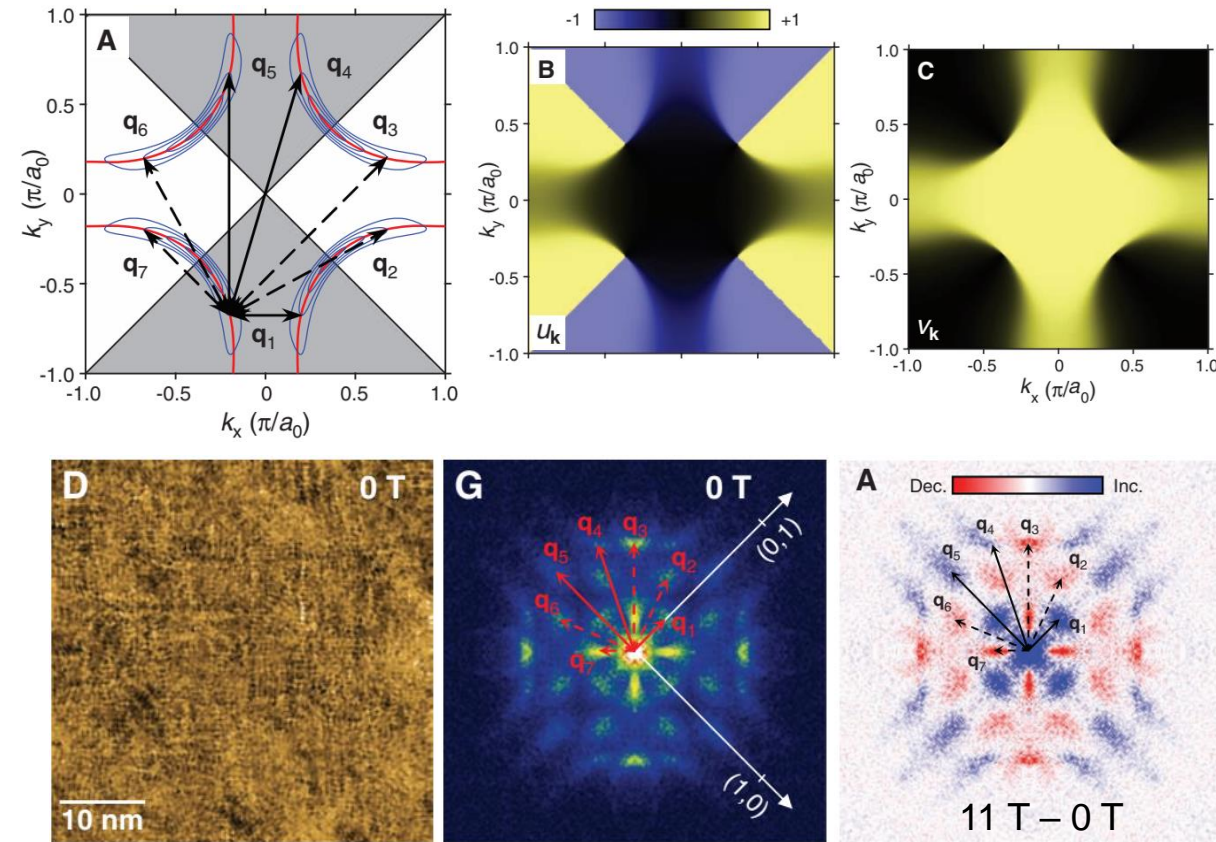
$\mathbf{u}_k$  and  $\mathbf{v}_k$  are Bogoliubov coefficients:

$$\mathbf{u}_k = \text{sign}[\Delta(k)] \sqrt{\frac{1}{2} \left[ 1 + \frac{\varepsilon(k)}{E(k)} \right]} \quad \mathbf{v}_k = \sqrt{1 - |\mathbf{u}_k|^2}$$

$|\mathbf{u}_{ki}\mathbf{u}_{kf}^* \pm \mathbf{v}_{ki}\mathbf{v}_{kf}^*|^2$  is called **coherence factor**, the plus sign is for magnetic scatterers, and the minus sign is for non-magnetic scatters.

- For non-magnetic scatterers, the coherence factor is suppressed for  $\vec{q}$  that preserve the sign of  $\Delta(k)$ , but enhanced for  $\vec{q}$  that change the sign of  $\Delta(k)$ .
- For magnetic scatterers (such as vortices), the coherence factor is suppressed for  $\vec{q}$  that change the sign of  $\Delta(k)$ , but enhanced for  $\vec{q}$  that preserve the sign of  $\Delta(k)$ .

**QPI is phase sensitive!**

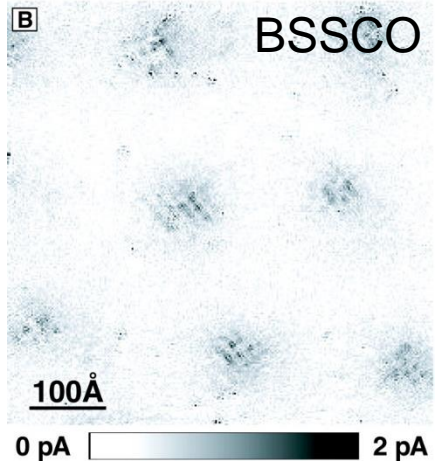


Hanaguri *et al.* *Science* 323, 923 (2009)

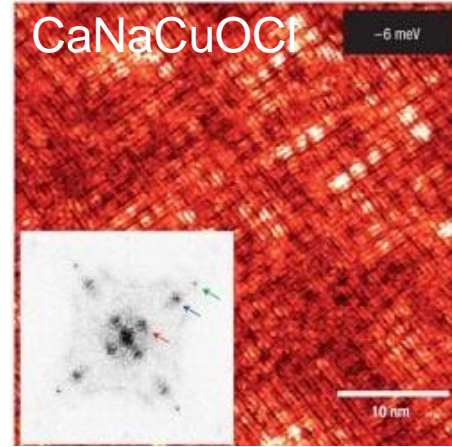
# STM in the study of superconductivity



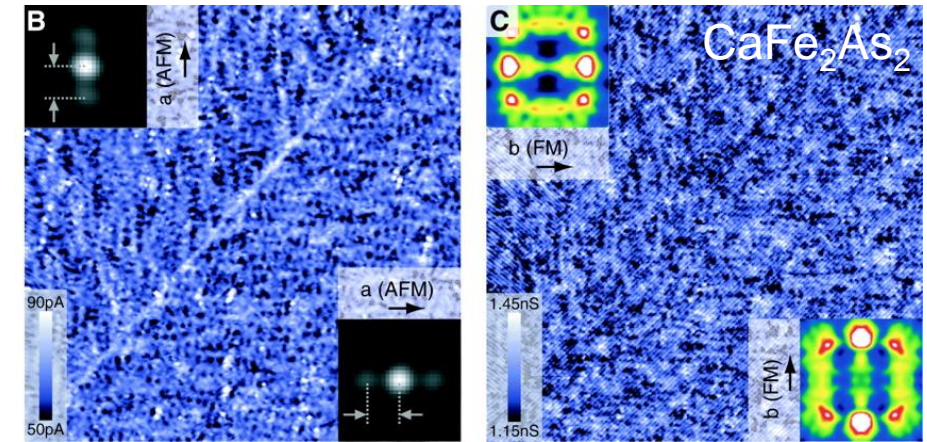
➤ Besides QPI, STM discovered other static spatial modulations in superconductors:



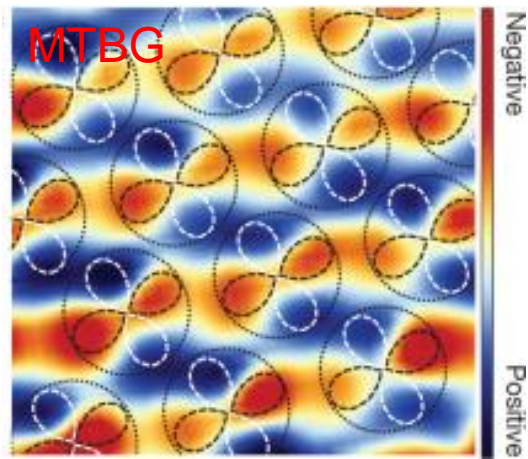
Hoffman, *et al.*, *Science* (2002)



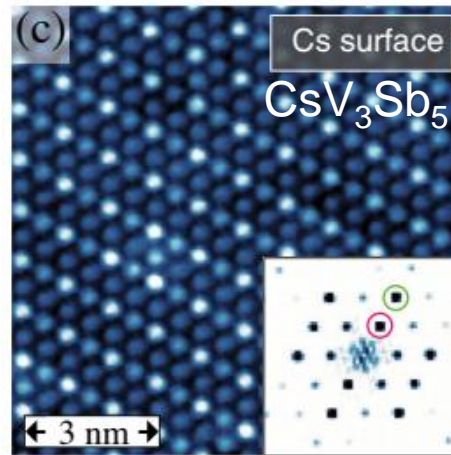
Hanaguri, *et al.*, *Nature Phys.* (2007)



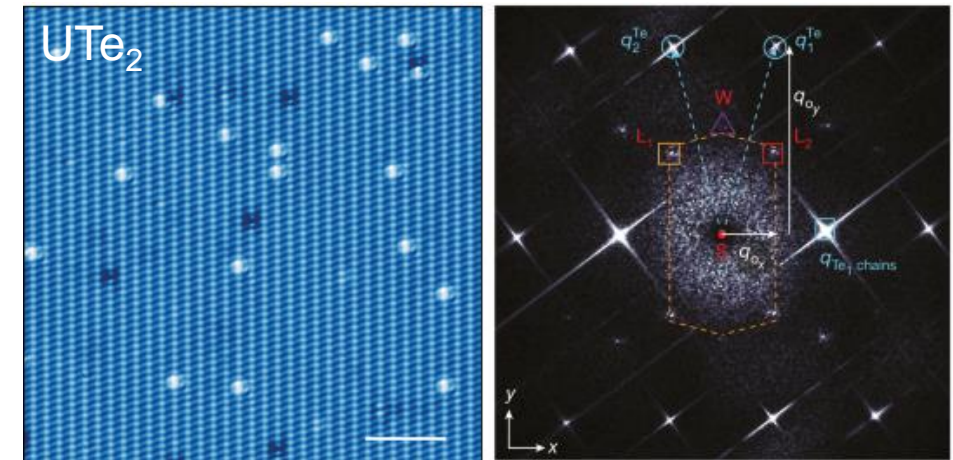
Chuang, *et al.*, *Science* (2010)



Jiang, *et al.*, *Nature* (2019)



Liang, *et al.*, *PRX* (2021)



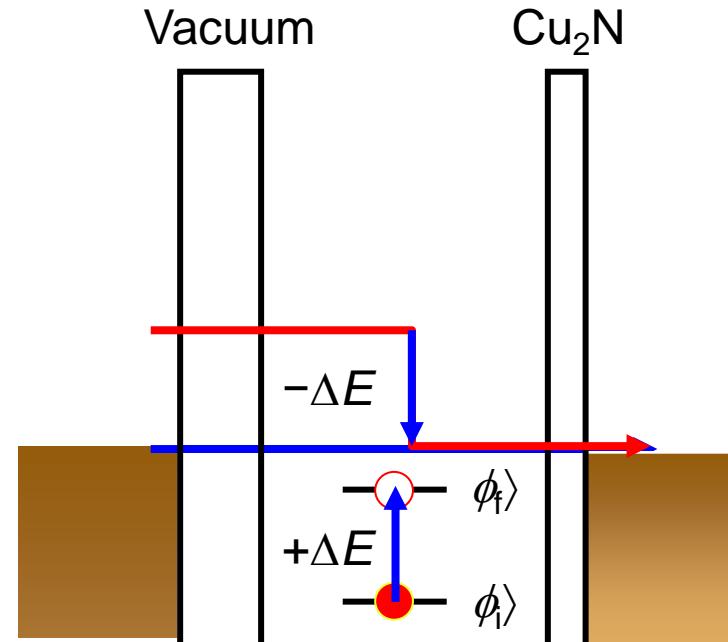
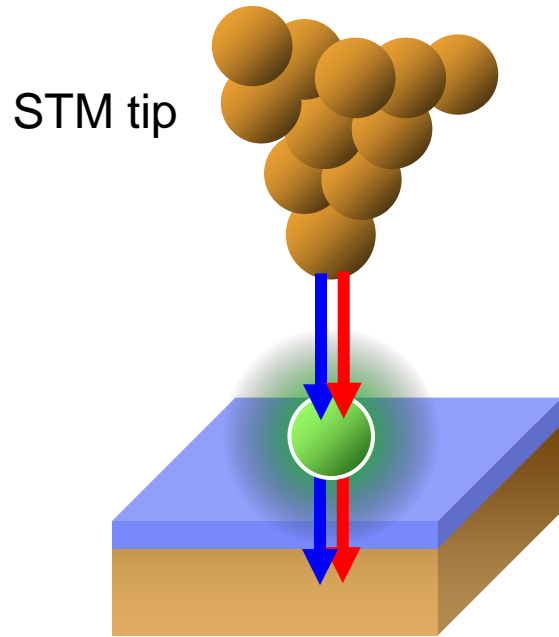
Aishwarya, *et al.*, *Nature* (2023)



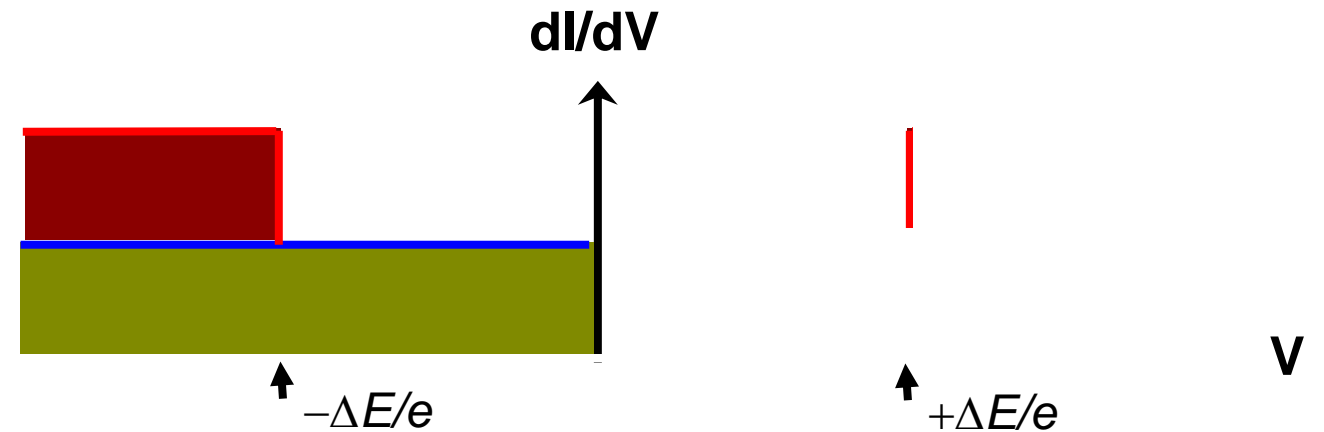
- **Introduction to STM: principles and instrumentation**
- **Application of STM in the study of superconductivity**
- **Inelastic tunneling spectroscopy, spin-resolved STM, time-resolved STM**



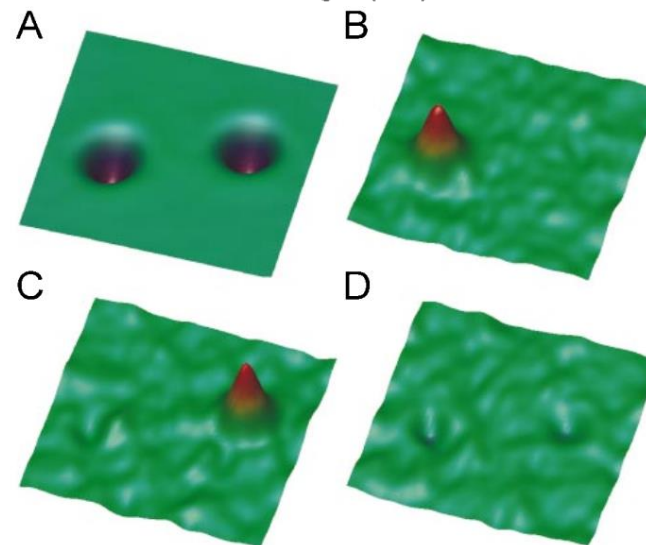
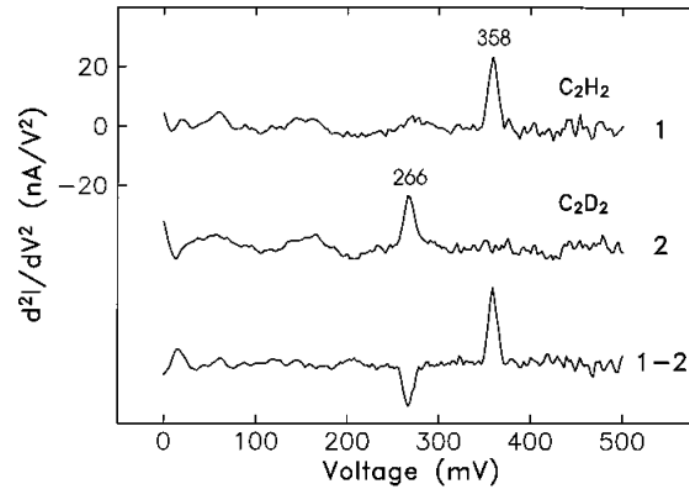
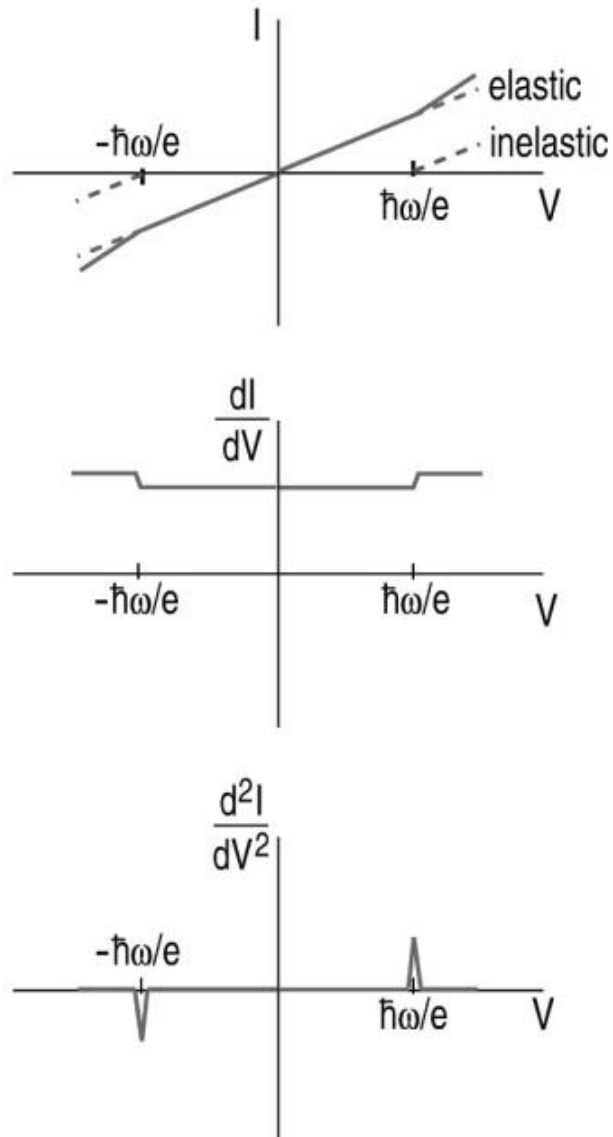
# Inelastic tunneling spectroscopy



Inelastic co-tunneling  
Interaction Hamiltonian:  $\vec{S} \cdot \vec{S} + u$   
e.g. Schrieffer, Wolff *Phys. Rev.* **149** 491 (1966)

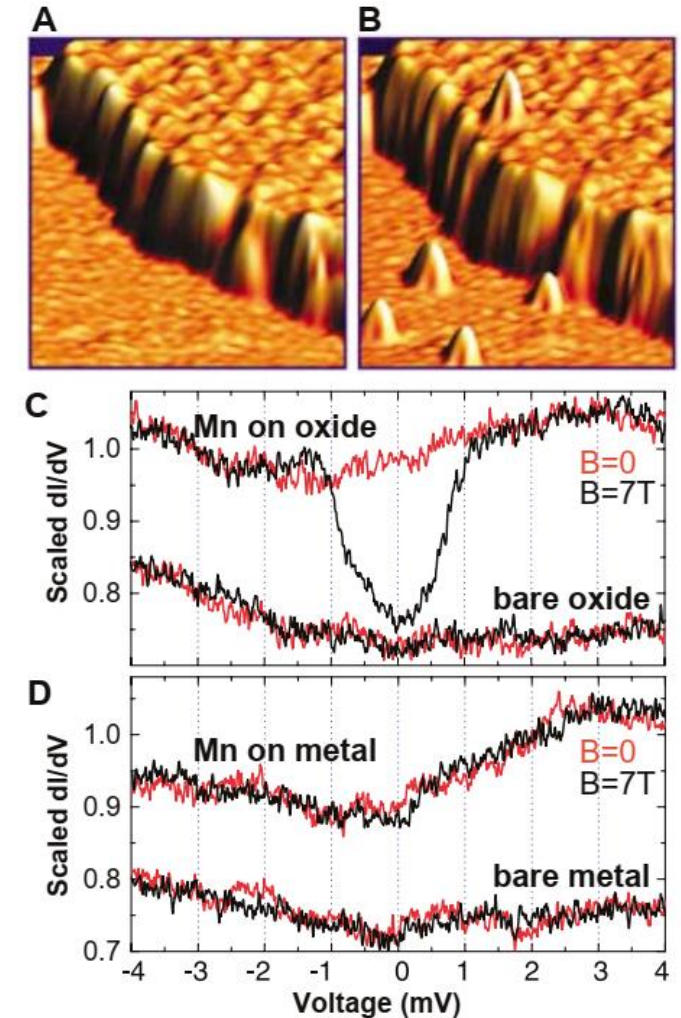


➤ Single molecule vibration

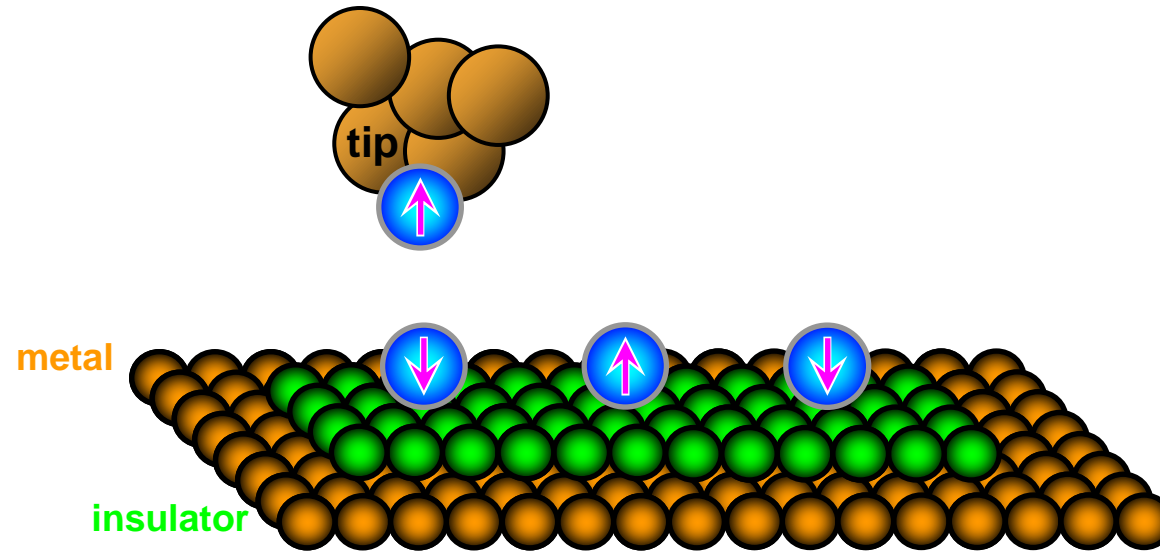


Stipe *et al.* Science (1998)

➤ Single atom spin excitation



Heinrich *et al.* Science (2004)



High current TMR: atom appears sabini STM image

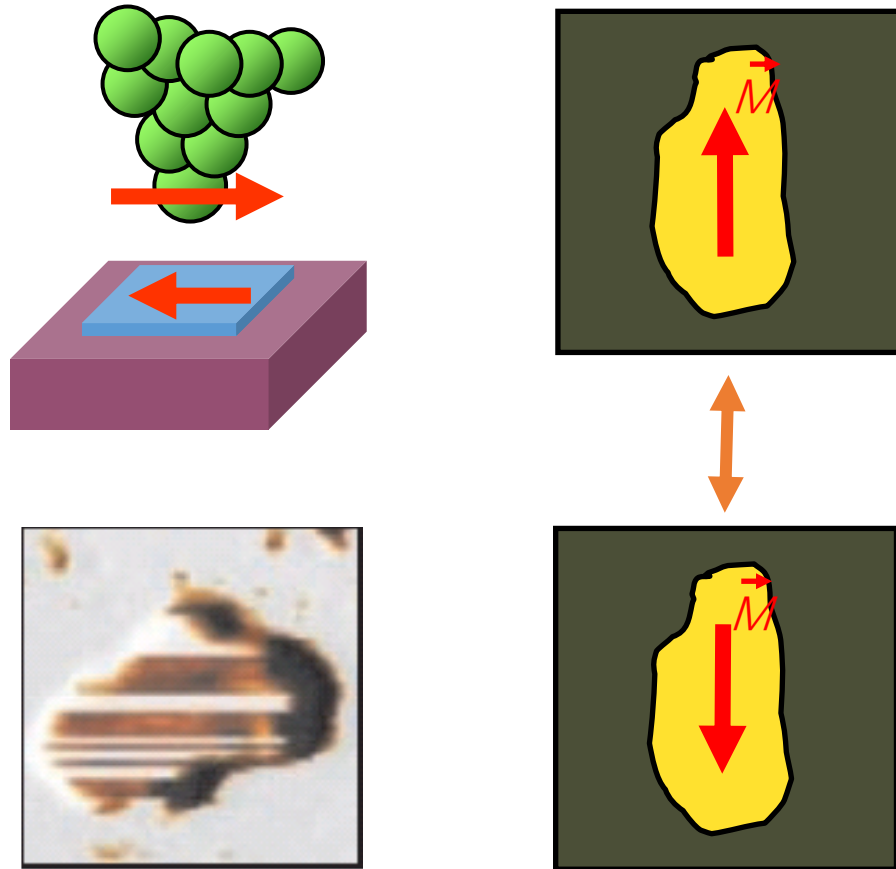
- STM is an atomic-scale tunneling magneto-resistance junction.

# Spin-polarized tunneling



## ➤ Classical magnetism

Spin-polarized tip

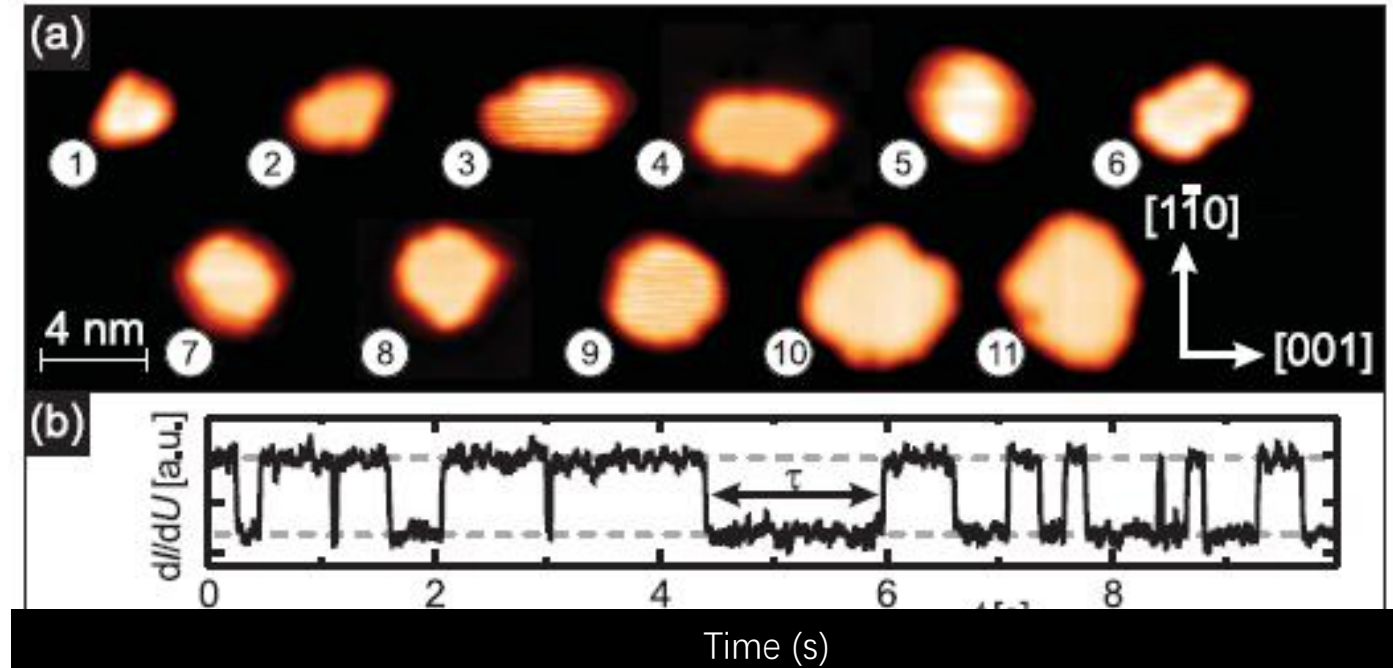


Fe-island on W(110)

$T = 56 \text{ K}$

Krause et al. *PRL* (2009)

Krause et al. *Science* 317 1537 (2007)



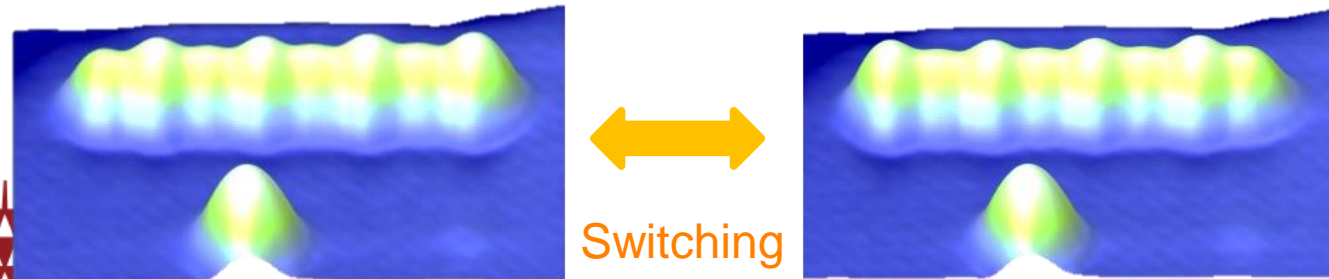
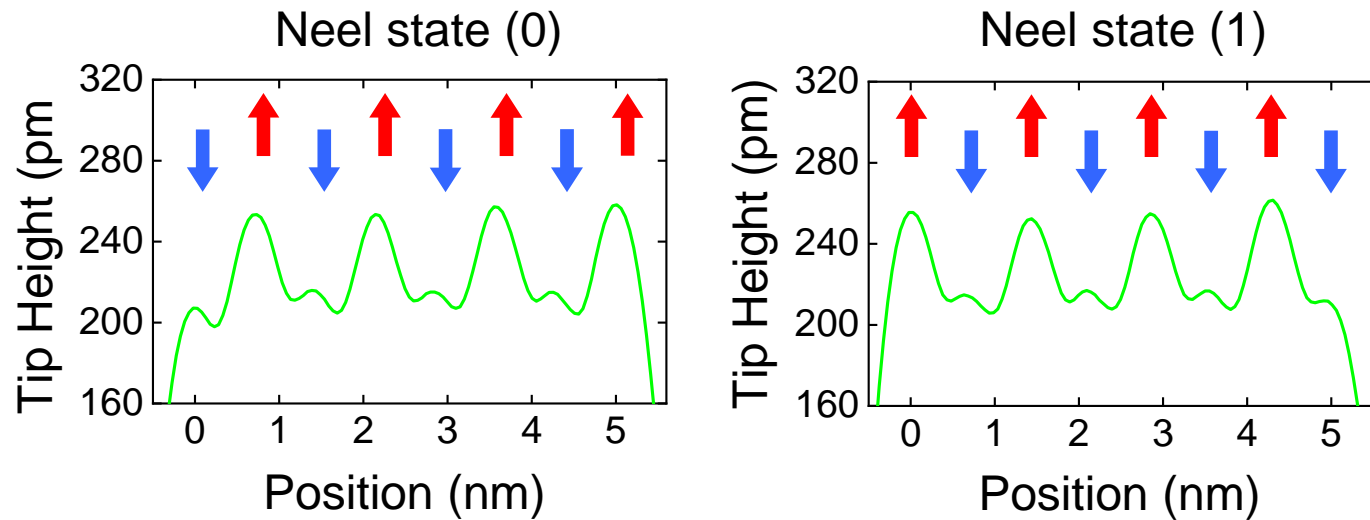
- Detect magnetic orientation by tunnel magnetoresistance
- Thermally activated switching
- Real-time recording of magnetic switching
- Time resolution: ms ( $-\mu\text{s}$ ), noise limited at low tunnel currents

# Spin-polarized tunneling



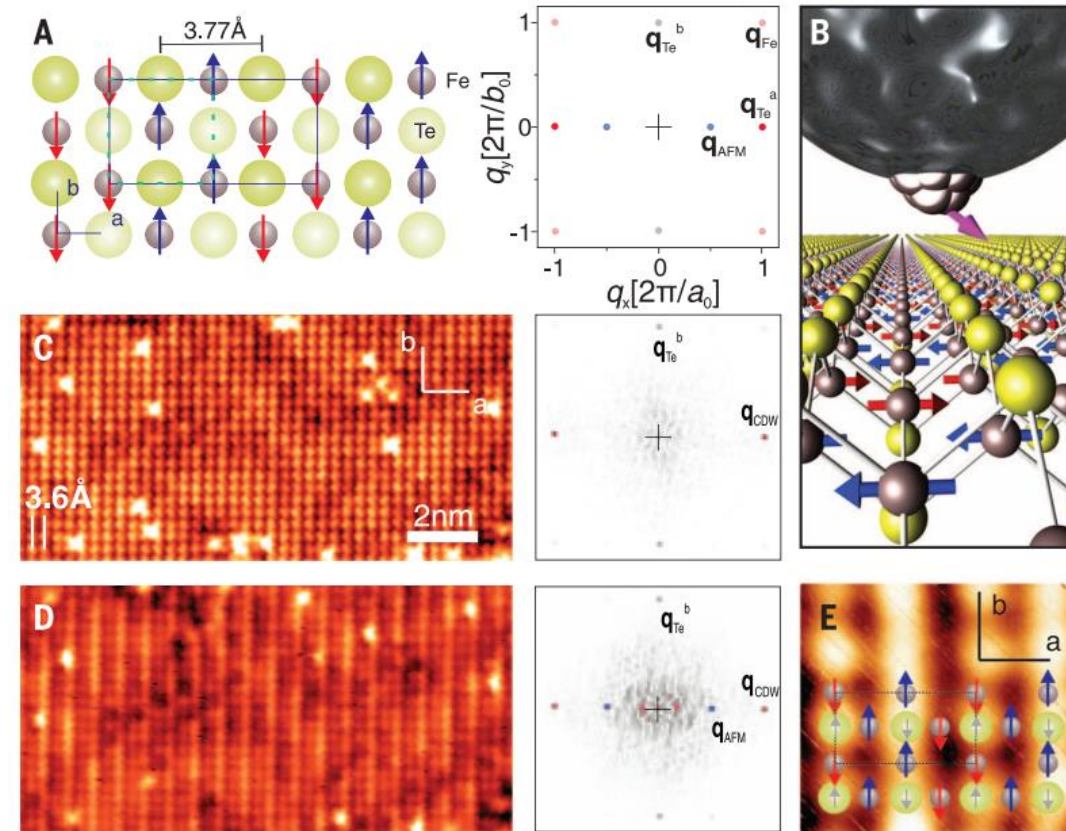
## ➤ Classical magnetism

Fe chain created by STM atom manipulation



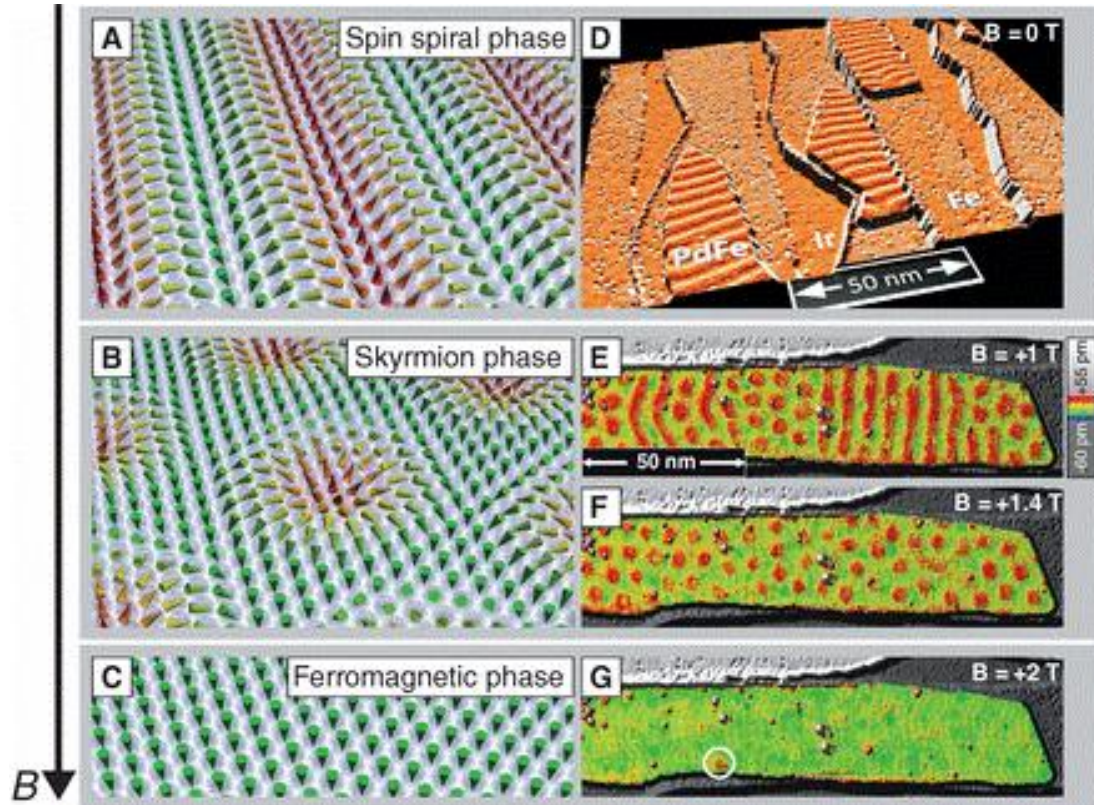
Loth, *et al. Science* (2012)

Surface of  $\text{Fe}_{1+y}\text{Te}$  single crystal



Enayat, *et al. Science* (2014)

➤ Magnetic skyrmion

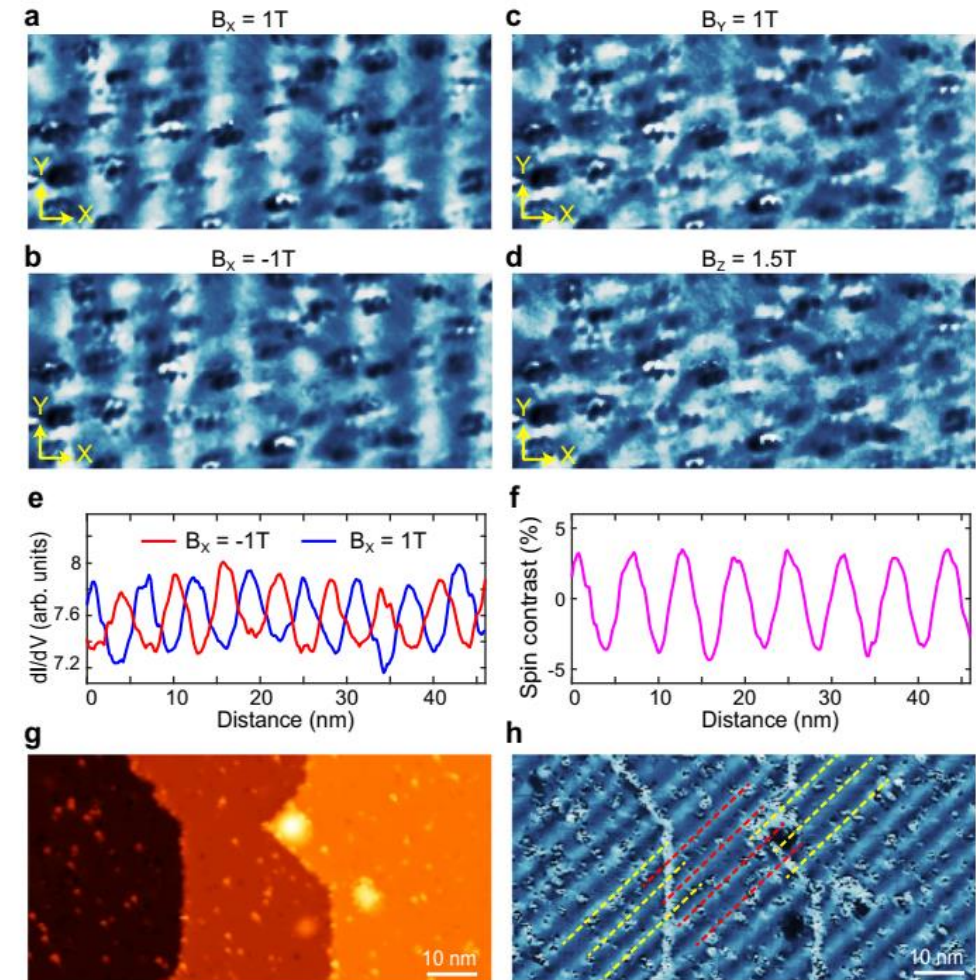


- Magnetic skyrmion: PdFe bilayer on Ir(111) surface

Romming, *et al. Science* (2013)

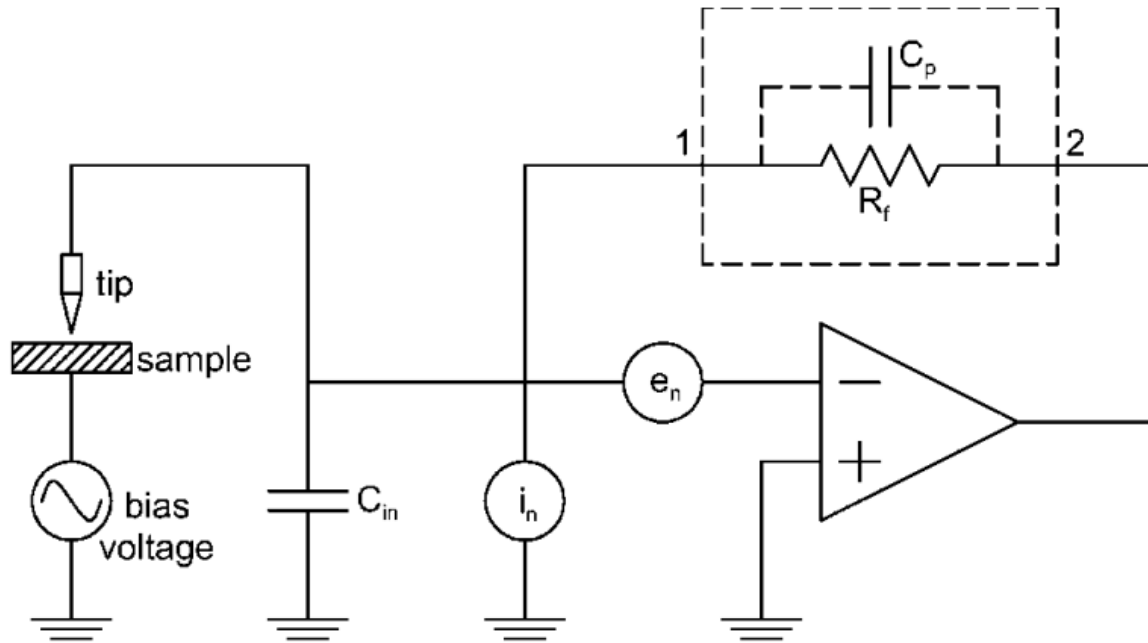
➤ Spin density wave

Cr(001) surface



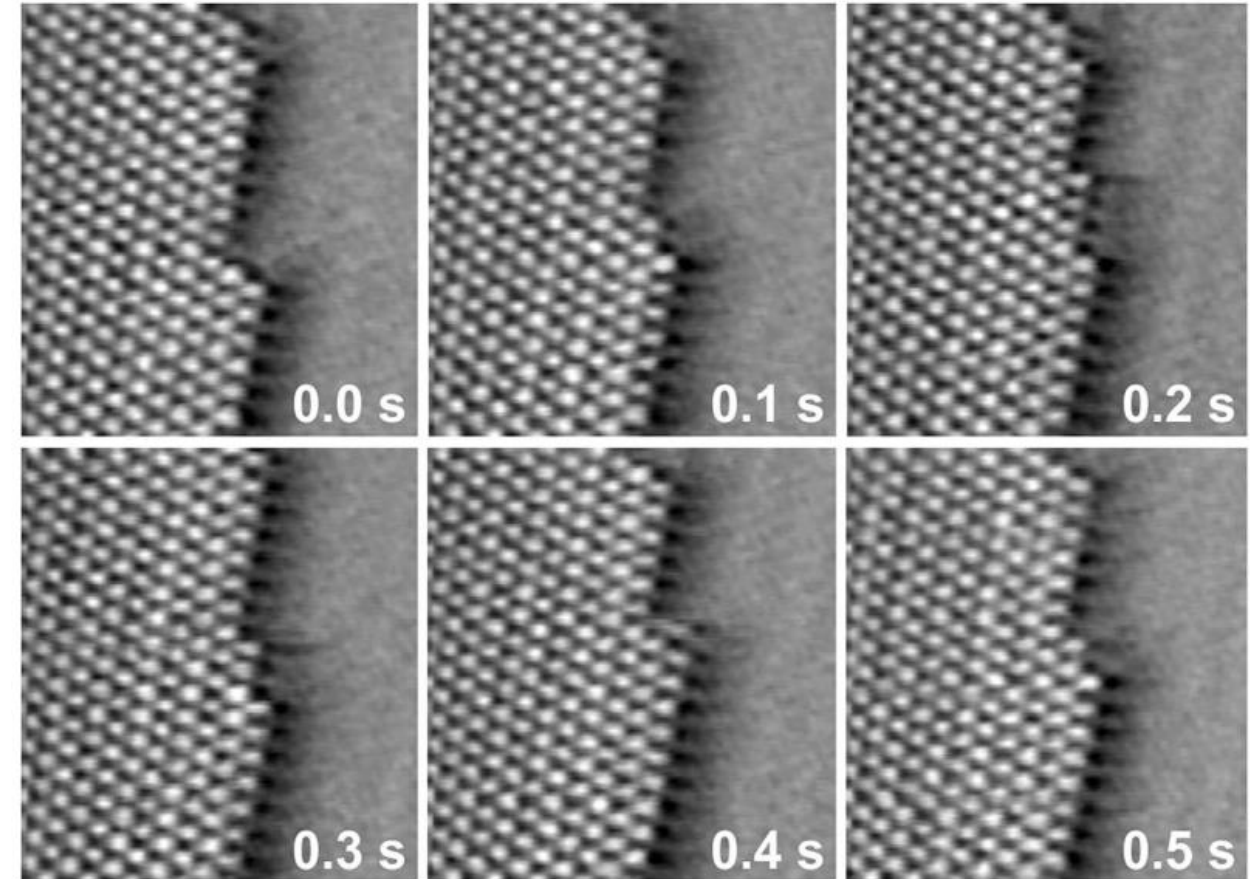
Hu, *et al. Nature Commun.* (2022)

## ➤ STM preamplifier



- Bandwidth: **10 KHz to 1MHz**
- Time resolution: 1-100  $\mu$ s

## ➤ Video-rate STM

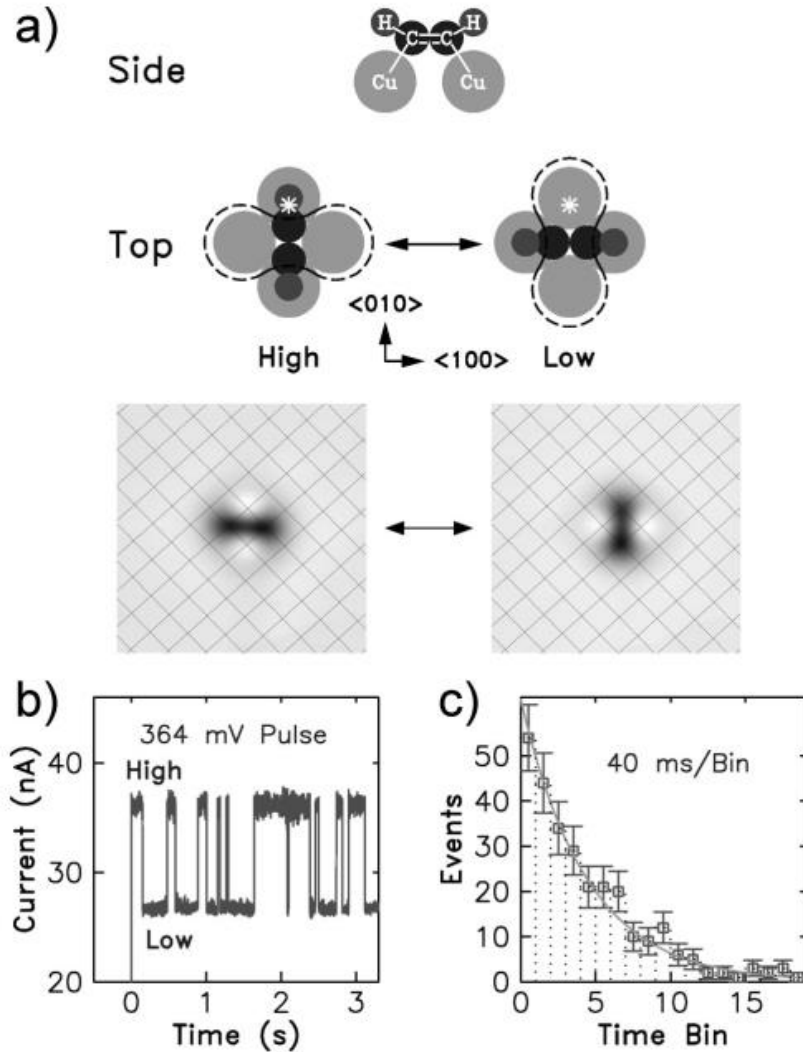


Sequences of STM images of Bi deposits on Au(100)

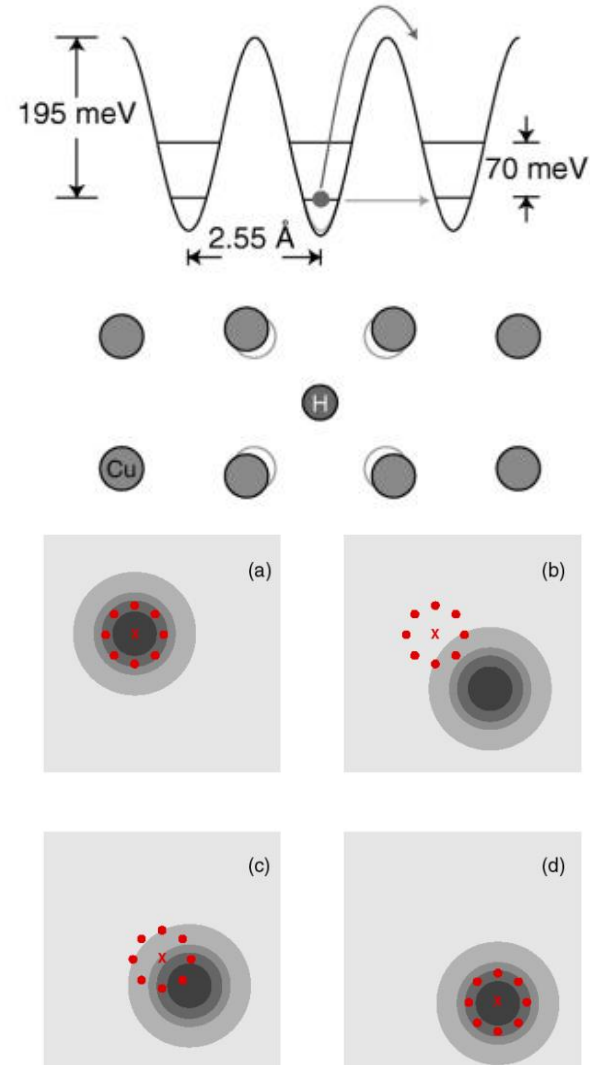
Matsushima, *et al. Faraday Discuss.* (2016)



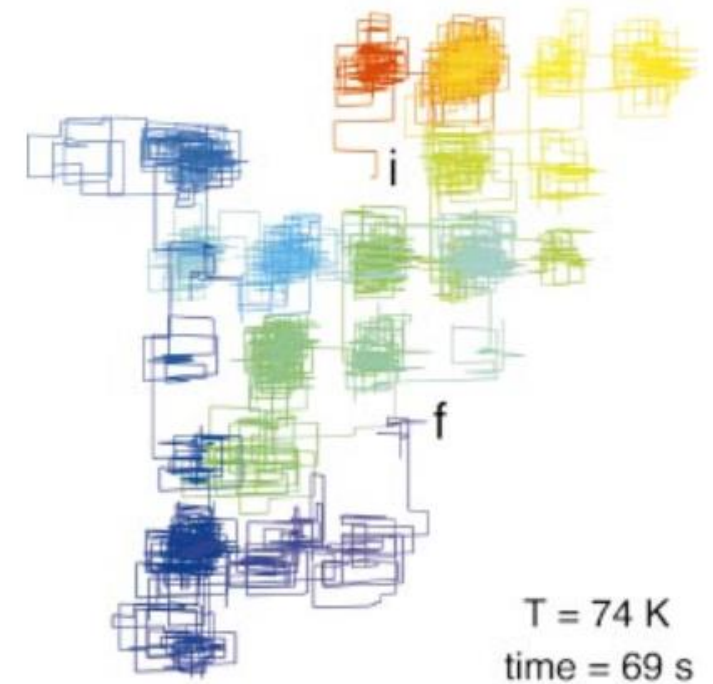
## ➤ Recording $I(t)$ curve



## ➤ Single atom tracking

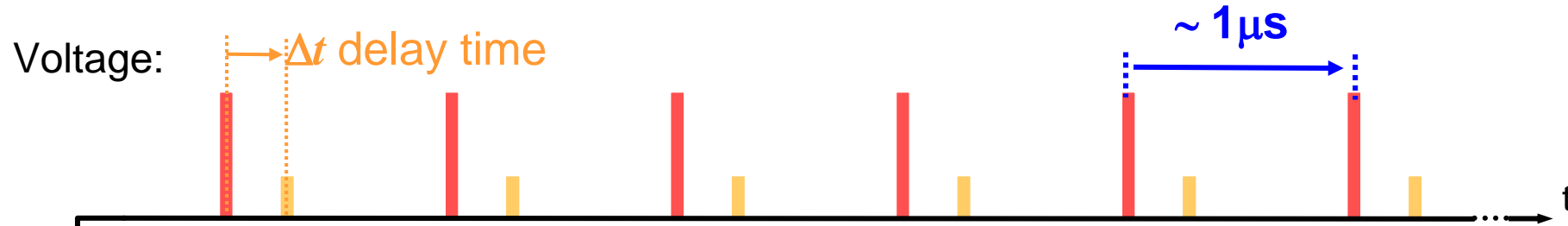


## Single Atom Tracking: H Thermal Diffusion

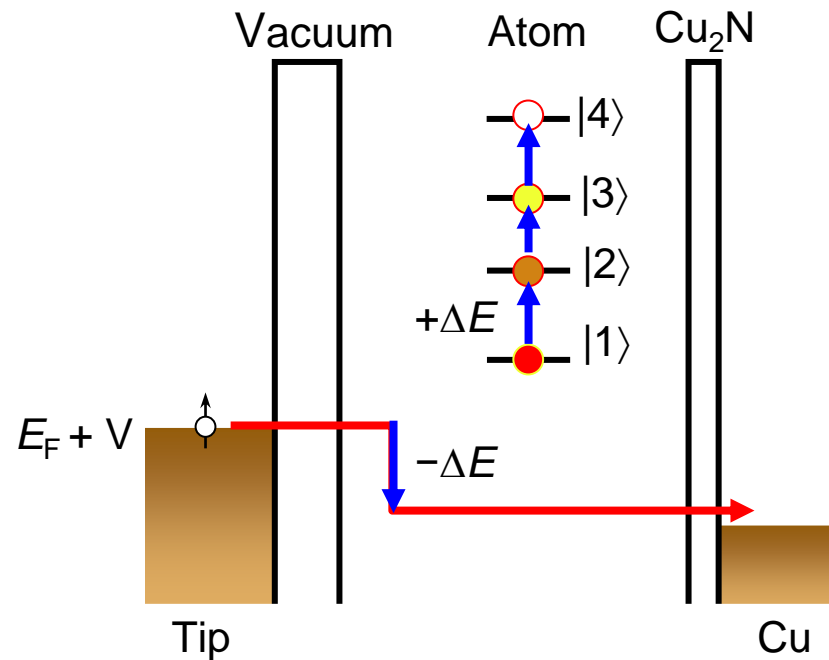
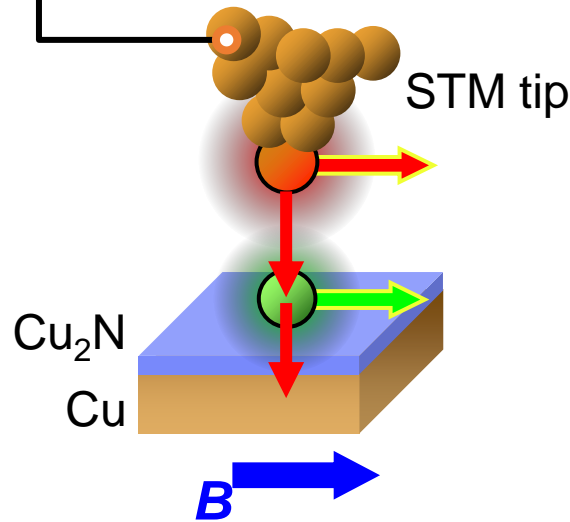


W. Ho, *et al.* JCP (2002)

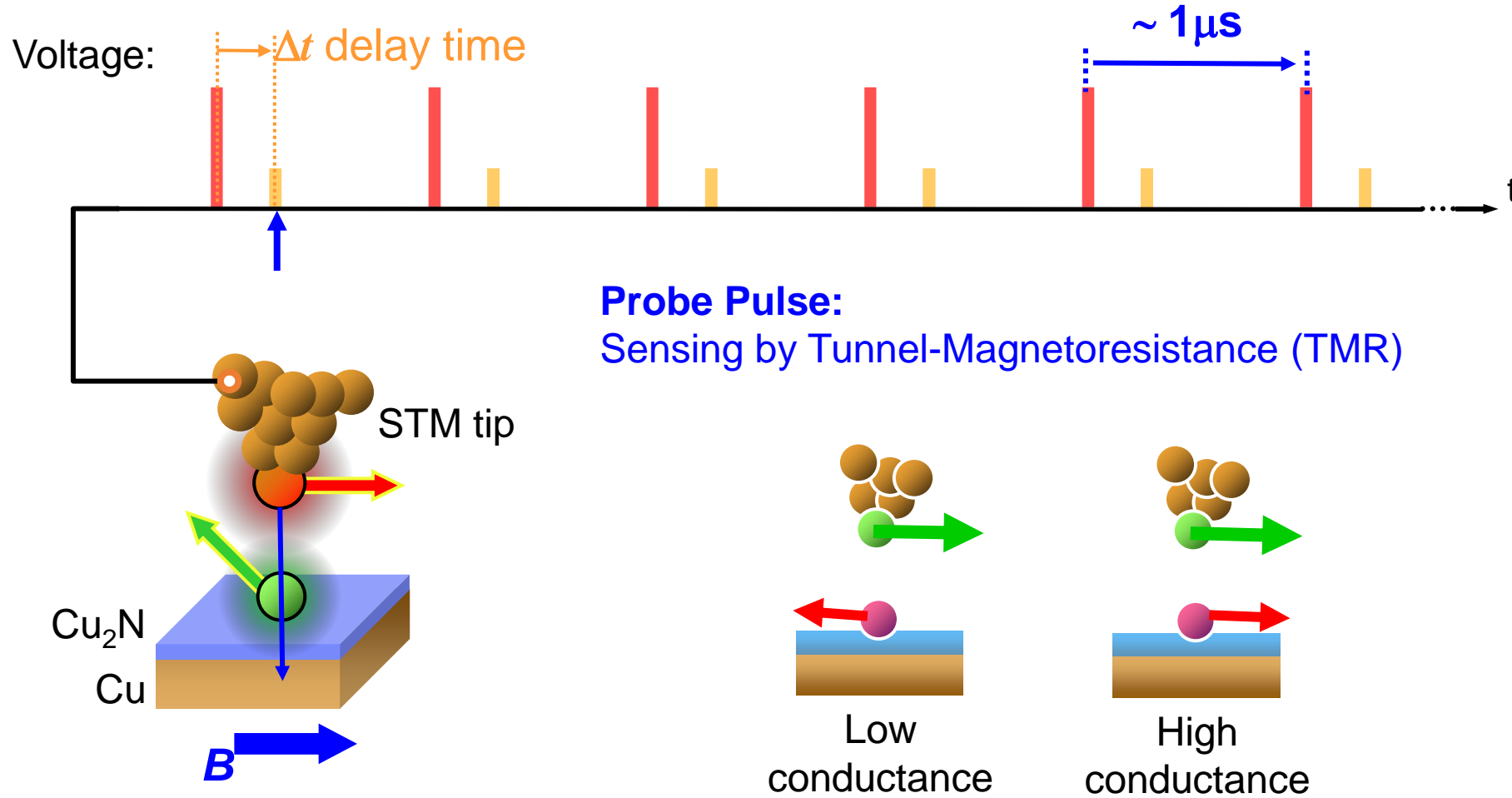
➤ Electronic pump-probe technique



Pump Pulse: Inelastic Tunneling excites spin

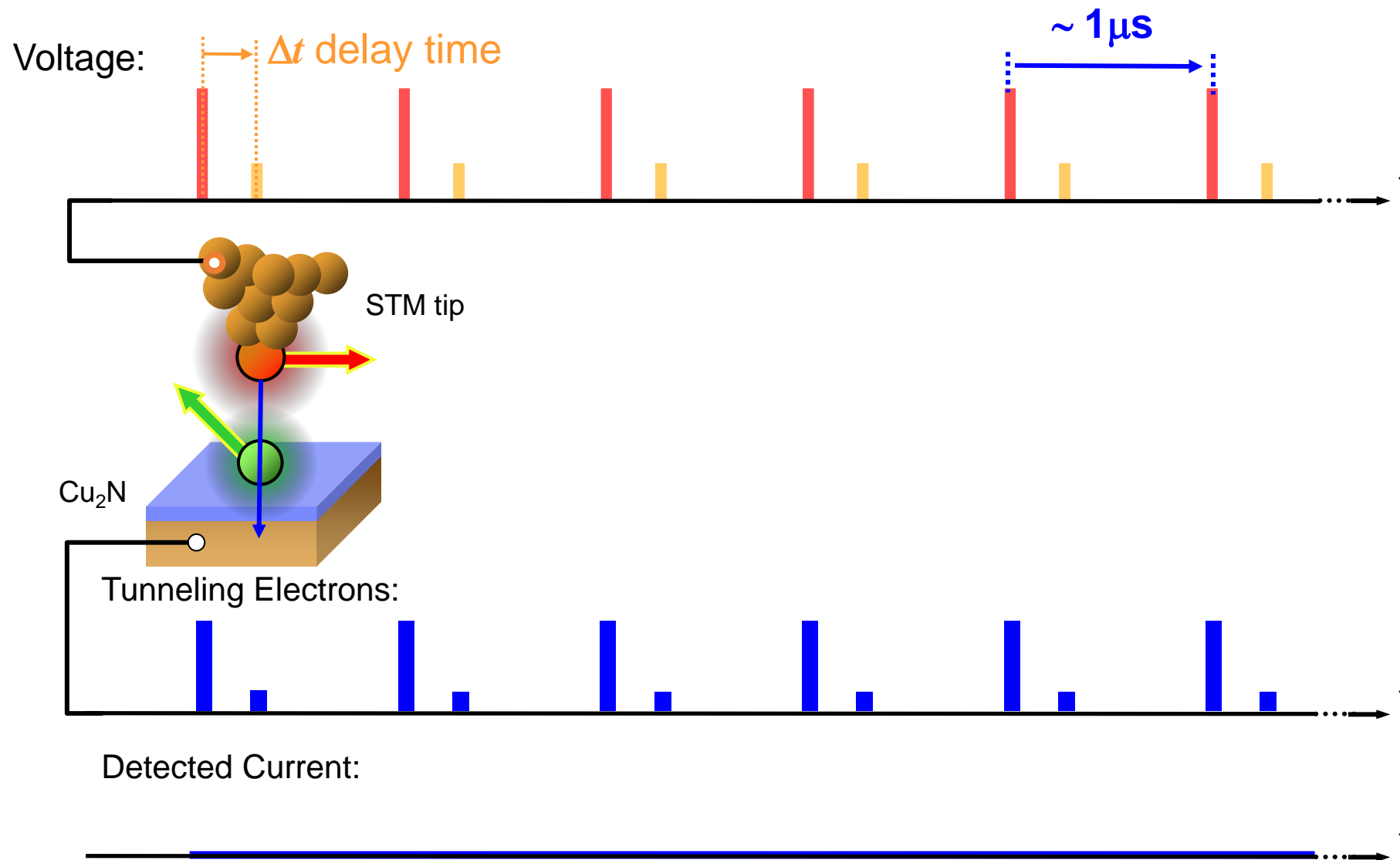


➤ Electronic pump-probe technique

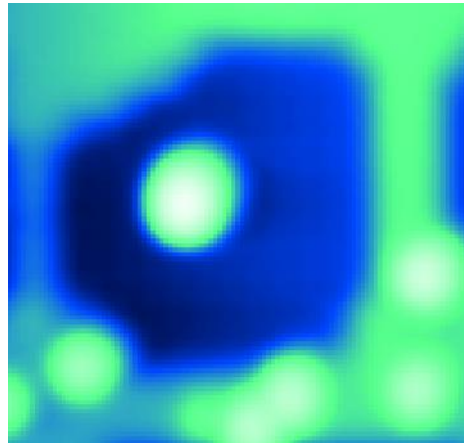


Loth *et. al.*, *Science* (2010)

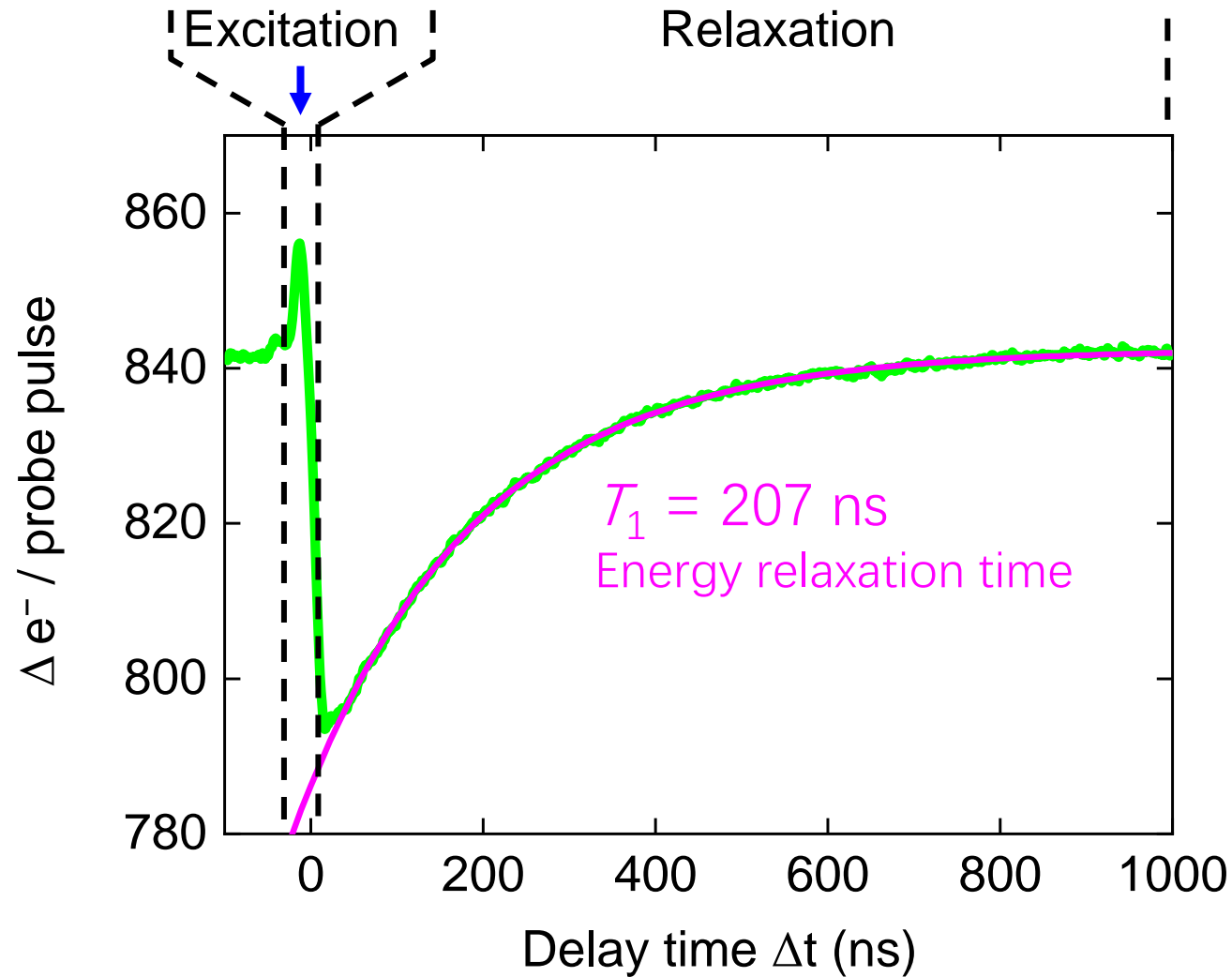
➤ Electronic pump-probe technique



➤ Electronic pump-probe technique

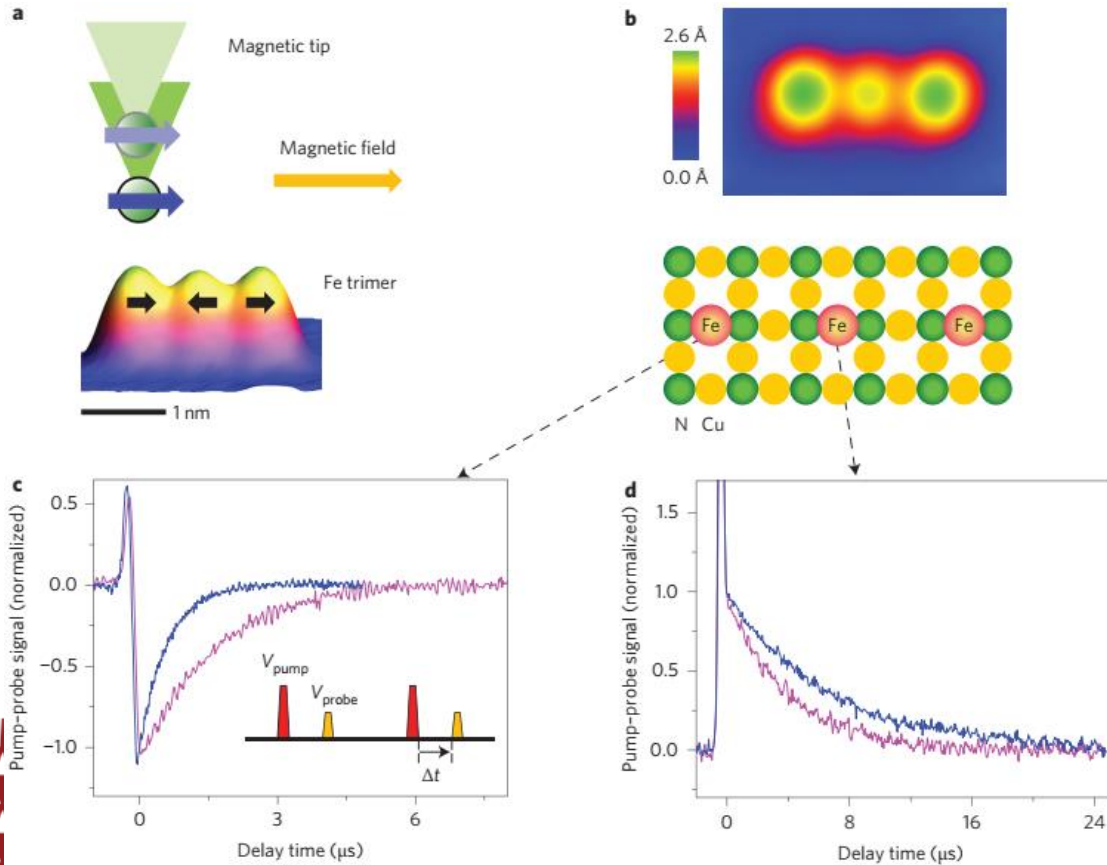


Close spaced  
Fe-Cu dimer  
 $B = 6.5 \text{ T}$



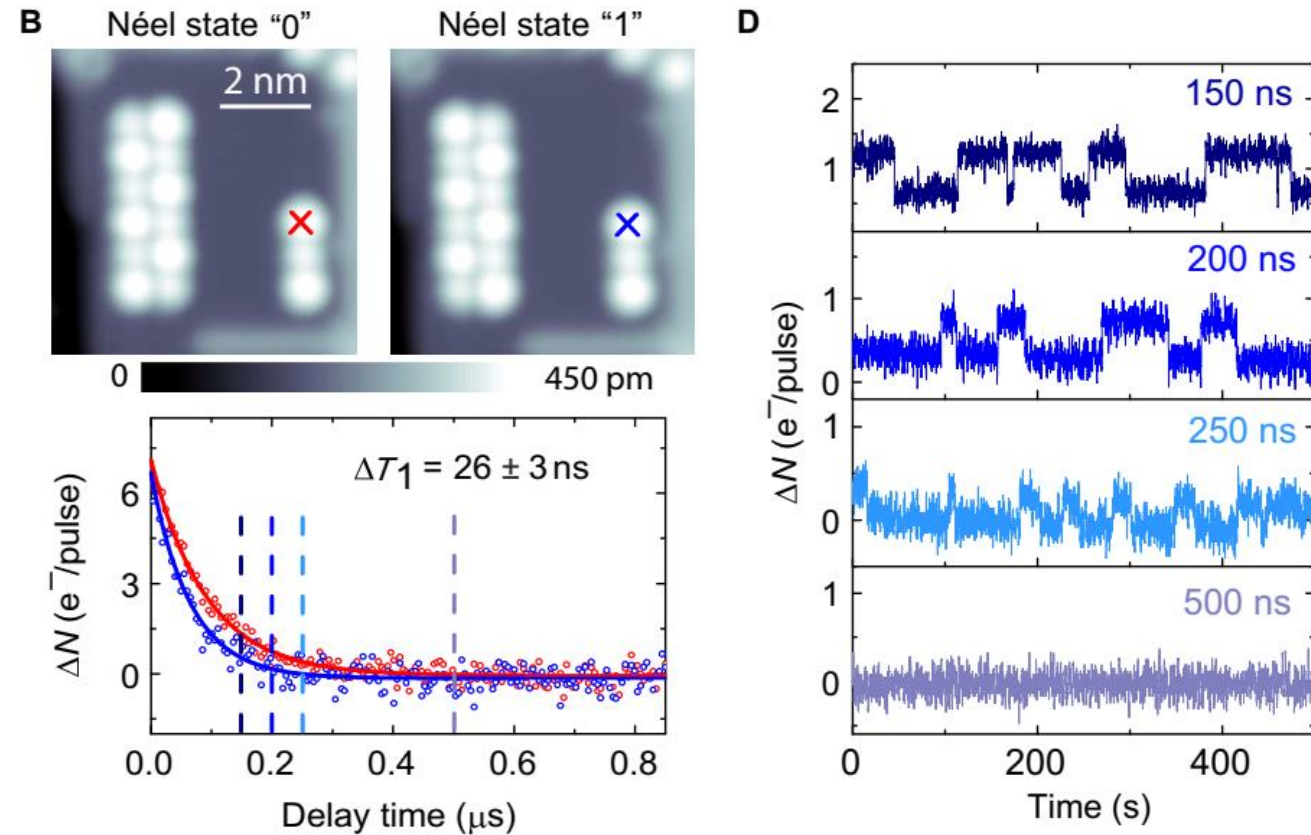
➤ Electronic pump-probe technique

- Control spin dynamics with SP-tip



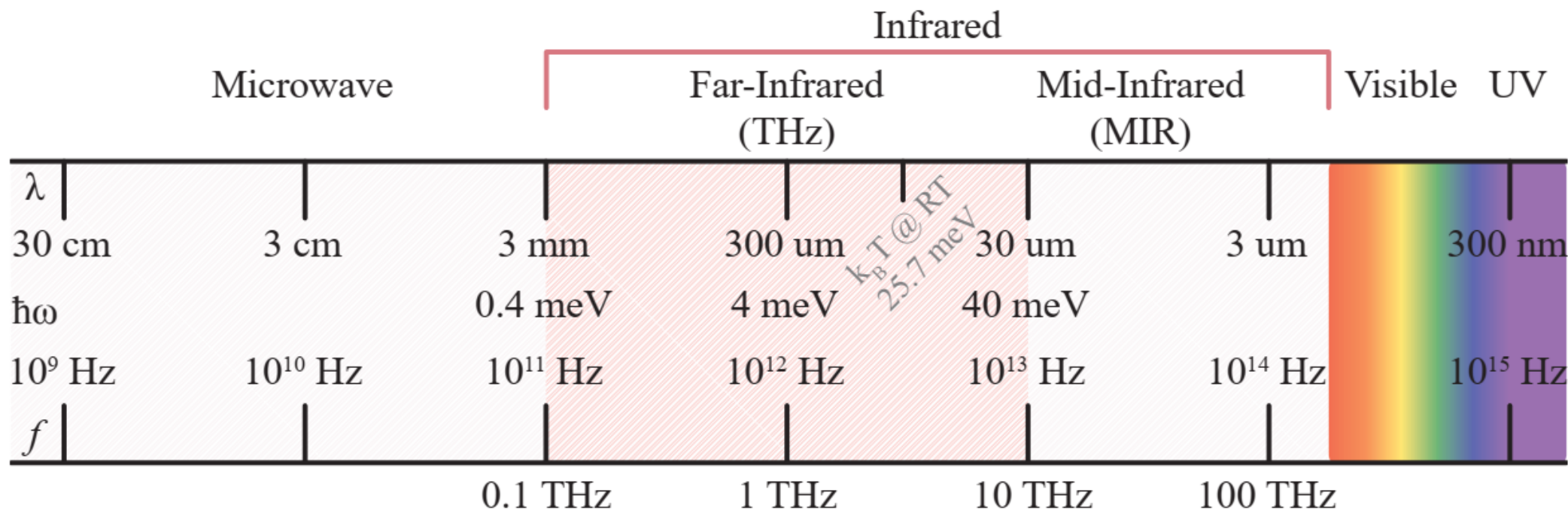
S. Yan *et al.*, *Nature Nanotech.* (2015)

- Sensing local magnetic environment

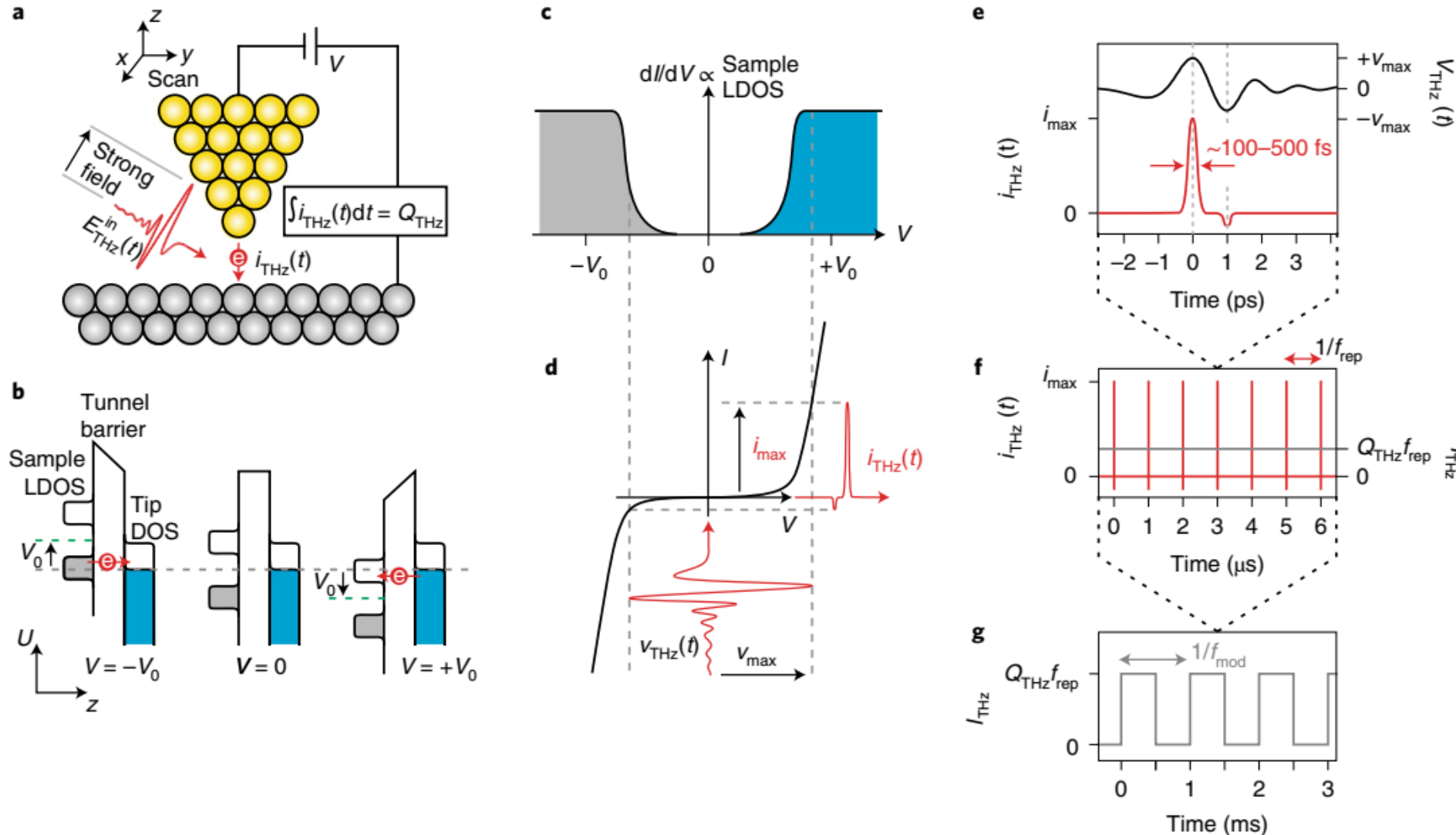


S. Yan *et al.*, *Science Adv.* (2017)

## ➤ THz-STM technique



## ➤ THz-STM technique

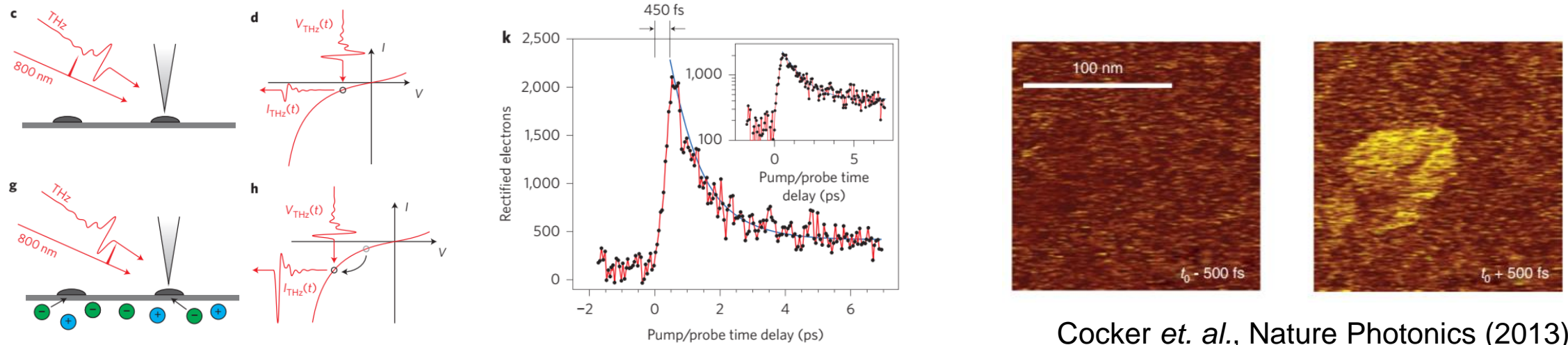


- In THz-STM, the static bias voltage ( $V_{\text{DC}}$ ) is replaced by a time-dependent bias modulation ( $V_{\text{THz}}(t)$ ) created by the electric field of a phase-stable single-cycle THz pulse coupled to the tip.

Cocker *et. al.*, *Nature Photonics* (2013)  
Cocker *et. al.*, *Nature Photonics* (2021)

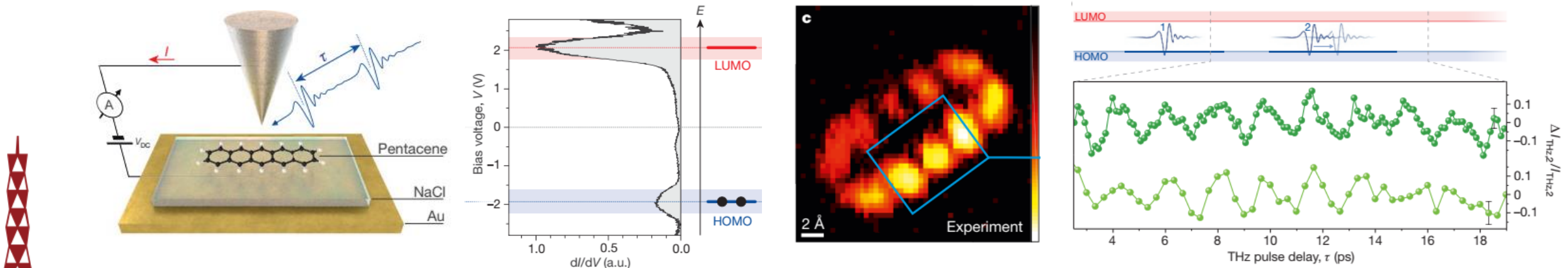


- Transient charging of an InAs nanodot measured with THz-STM



Cocker *et. al.*, Nature Photonics (2013)

- Tracking the ultrafast motion of a single molecule with THz-STM



Cocker *et. al.*, Nature (2016)

- **Introduction for STM: principles and instrumentation**
- **Application of STM in the study of superconductivity**
- **Inelastic tunneling spectroscopy, spin-resolved STM, time-resolved STM**

