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Modeling Quantum Tunneling in Terahertz Scanning Tunneling Microscopes

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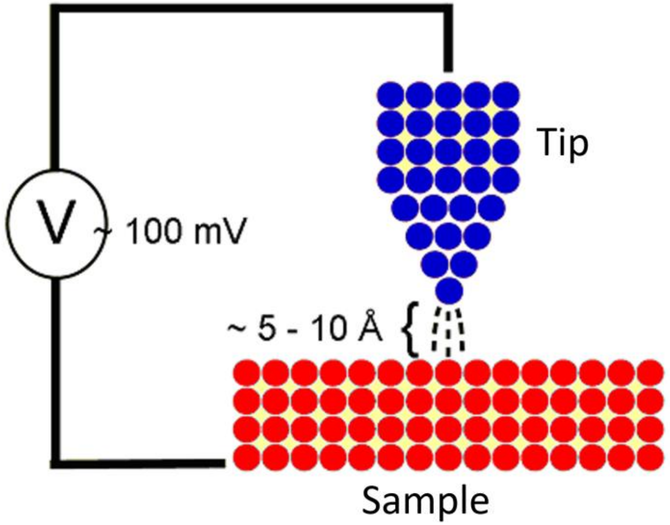
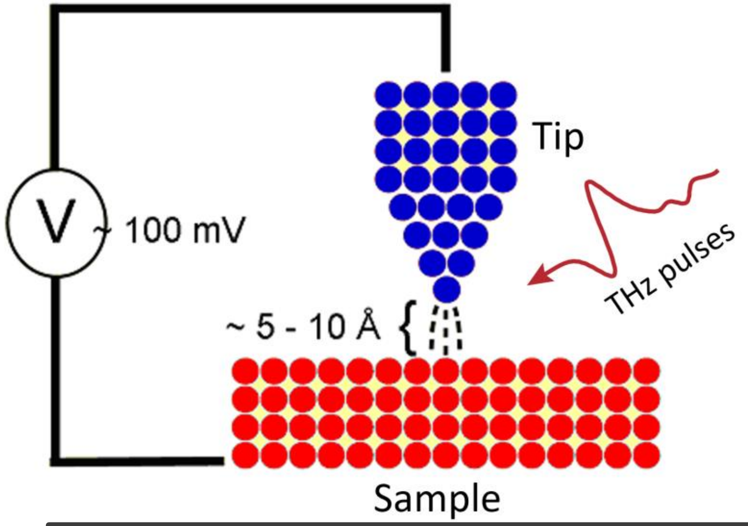
Why Terahertz Scanning Tunneling Microscope (THz-STM)?

A conventional STM



Tunneling current ~ 100 pA

THz-STM



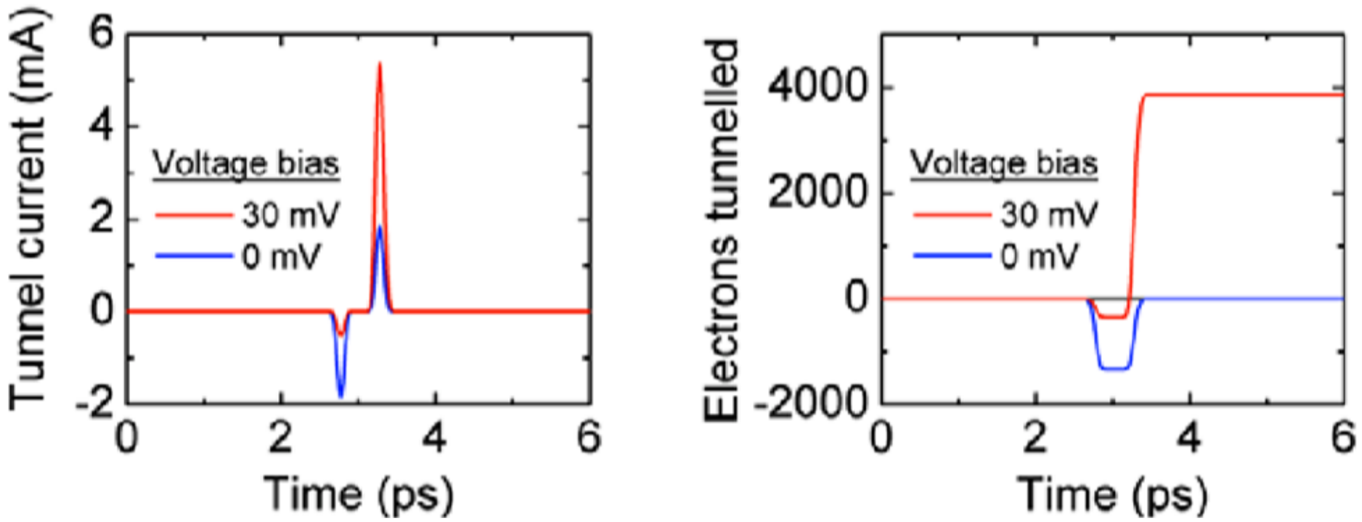
Peak Tunneling current ~ 100 μA [1-4]

- To record the movement of a single molecule on its intrinsic timescale, STMs must have ultrafast temporal resolution and atomic spatial resolution – THz STM
- Peak THz voltage bias transients greater than 3 V across the STM junction can be achieved → field emission of subpicosecond tunnel currents with current densities exceeding $10^9\ \mathrm{A/cm^2}$.

[1] Cocker et al., **Nat. Photonics** 7, 620 (2013)
[2] Jelic, V. et al., **Nat. Phys.** 13, 591–598 (2017)
[3] Luo, Y. et al., **Phys. Rev. B** 102, 205417 (2020)
[4] Tachizaki, T., **APL Mater.** 9, 060903 (2021)

Quantum Tunneling at Tip-Sample Junction

- ➢ Nonlinear current-voltage behavior of the tunnel junction leads to rectified ultrafast current burst
- ➢ It is important to: understand, improve, and control this rectification



[1] Cocker et al., **Nat. Photonics** 7, 620 (2013)

- ➢ High tunneling voltage at tip-sample junction → Field Emission regime → Effects of **Exchange Correlation Potential** and **Space Charge Potential** become crucial
- ➢ Existing theoretical studies[1-3] use Simmons [4] & Bardeen theory of tunneling which ignore both
- ➢ We consider them in our model to study the scaling of tunneling current at THz-STMs for different bias, THz field, tip-height and material properties

Goal: Manipulation of electron transport at the tunnel junction

[2] Jelic, V. *et al*., **Nat. Phys.** 13, 591–598 (2017)

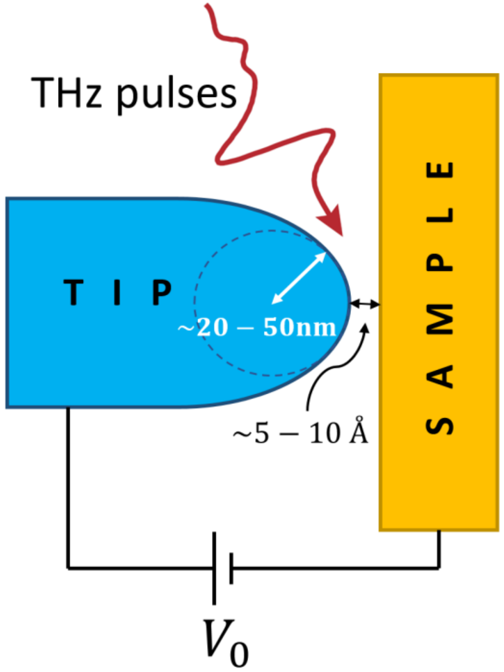
[3] Luo, Y. et al., **Phys. Rev. B** 102, 205417 (2020)

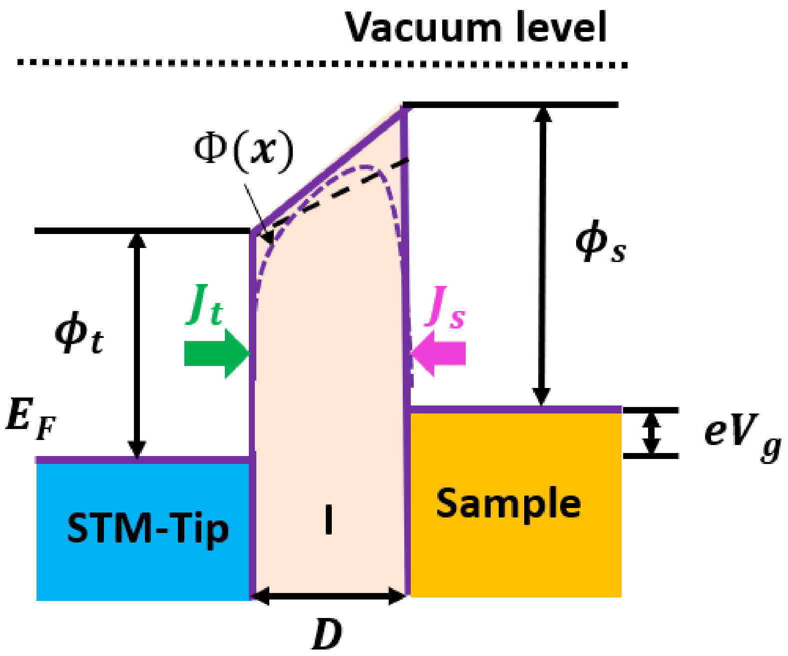
[4] J. G. Simmons, **J. Appl. Phys.** 34, 2581 (1963)

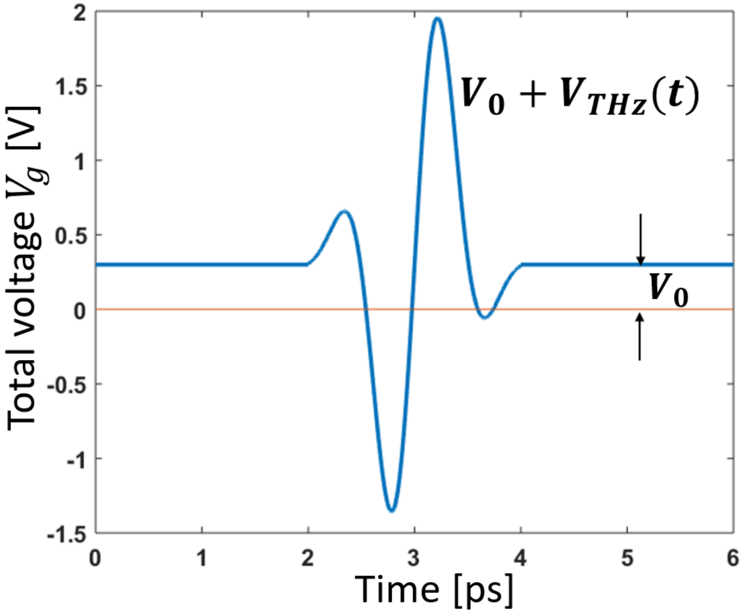
Outline

- Introduction
  - Potential barrier between tip and sample
  - Forward and reverse bias
- Self-consistent model formulation for a metal-insulator-metal junction
- Current-voltage characteristics
- Rectification of the tip-sample tunnel junction
  - Validation with experiment
  - For different time-dependent bias voltages
  - For different metal tips
  - For different tip heights
- Summary
- Future work

Terahertz Scanning Tunneling Microscope (THz-STM)







- THz pulse is coupled into the STM junction
- Transient tunnel currents between the tip and the sample

- The potential profile at tip-sample junction
  - $\phi_{t/s} = W_{t/s} - X$, $W_{t/s}$ = work function of tip/sample, X = electron affinity of the insulator
  - $J_{t/s}$ = electron current densities emitted from the tip/sample

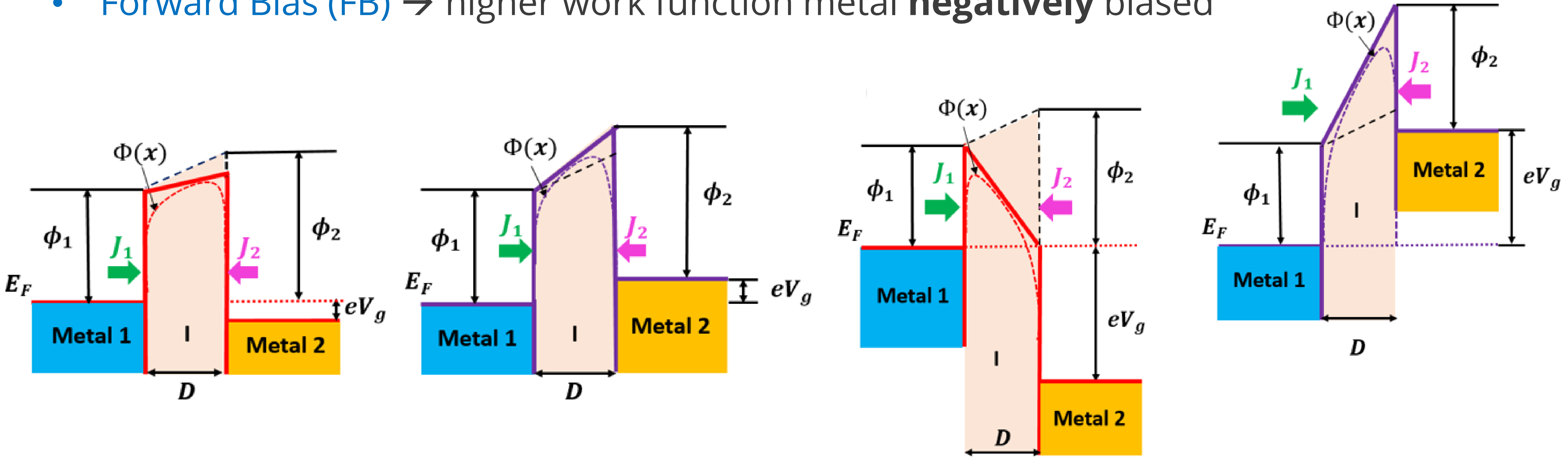
Total voltage applied to the tip-sample junction

S. Banerjee, and P. Zhang, **Phys. Rev. Applied** 18, 024011 (2022)

Forward and Reverse Bias in dissimilar metal-insulator-metal

Due to the inherent work-function difference between the metal electrodes, current transport is polarity dependent in dissimilar metal-insulator-metal (MIM)

- Reverse Bias (RB) → higher work function metal **positively** biased
- Forward Bias (FB) → higher work function metal **negatively** biased



RB ($eV_g < \phi_1$)

FB ($eV_g < \phi_1$)

RB ($eV_g > \phi_1$)

FB ($eV_g > \phi_1$)

Banerjee and Zhang, **AIP Adv**. 9, 085302 (2019)

Potential Barrier at Tip-Sample Junction

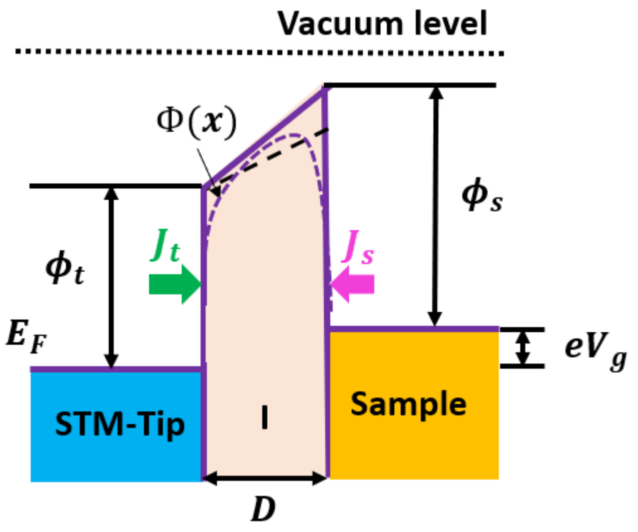


Image charges build up in the metal electrode as carriers approach the metal-insulator interface. The potential associated with these charges reduces the effective barrier height.

Due to electron interaction in a quantum system

Exchange energy is released when two or more electrons with same spin exchange their position.

Correlation energy is a measure of how much the movement of an electron is influenced by the other electrons.

Fermi level

Image charge potential

$$\Phi(x) = E_F + \Phi_w(x) + \Phi_{image}(x) + eV(x) + \Phi_{xc}(x)$$

Exchange correlation potential

$$\Phi_w(x) = \phi_t + (\phi_s - \phi_t)x/D$$

J. G. Simmons, *J. Appl. Phys.*, 1963.

$$eV(x) = eV_g\, x/D + eV_{sc}(x)$$

Bias voltage

Space charge potential

Due to the cloud of free electrons in the insulator. It slows the carriers down.

P. Zhang, **Sci. Rep.** 5, 9826 (2015)

S. Banerjee and P. Zhang, **AIP Adv**. 9, 085302 (2019)

Generalized Self-Consistent Tunneling Model

Symbols have their usual meaning , or, as defined earlier

- The tunneling current density $J_{net} = |J_1 - J_2|$

$$J_1 = \int_{-\infty}^{\infty} N_1(E_x) D(E_x) dE_x \ , \qquad J_2 = \int_{-\infty}^{\infty} N_2(E_x) D(E_x) dE_x$$

- Transmission probability ,

$$D(E_x) = \exp\left[-\frac{2}{\hbar}\int_{x_1}^{x_2}\sqrt{2m[\Phi(x) - E_x]}\, dx\right] \quad \text{(WBK approximation)}$$

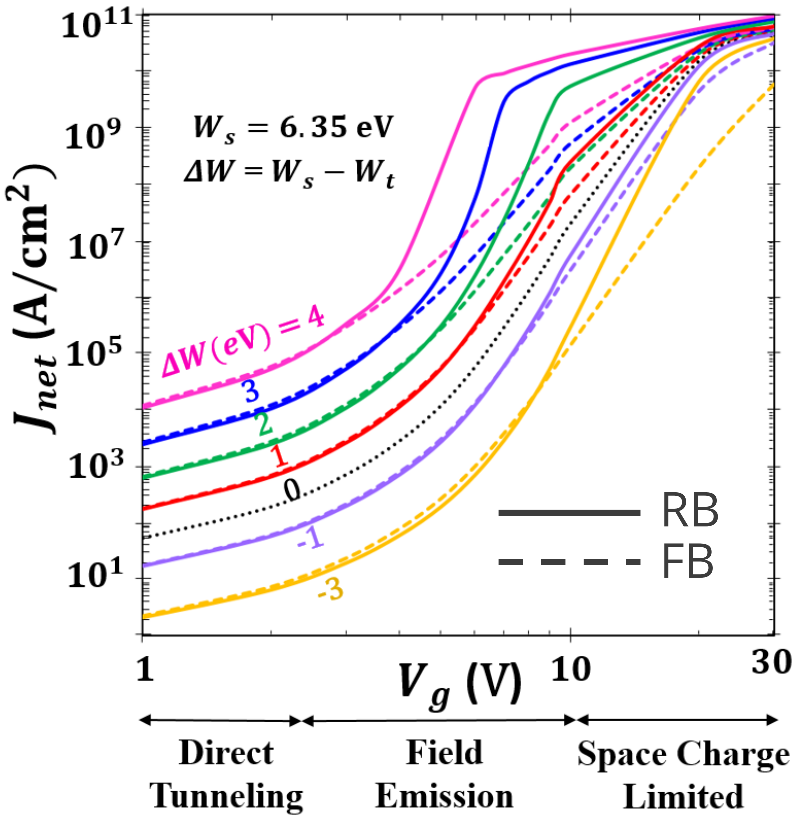
$$N_1(E_x) = \frac{m_e k_B T}{2\pi^2\hbar^3} \ln\left(1 + e^{-(E_x - E_F)/k_B T}\right)$$

$$N_2(E_x) = \frac{m_e k_B T}{2\pi^2\hbar^3} \ln\left(1 + e^{-(E_x + eV - E_F)/k_B T}\right)$$

(free-electron theory of metal)

P. Zhang, **Sci. Rep.** 5, 9826 (2015)
Banerjee and Zhang, **AIP Adv**. 9, 085302 (2019)

J-V Characteristic of a Metal-Insulator-Metal Junction [1]



In field emission regime, the asymmetry between FB and RB currents increases significantly as the work function difference $|\Delta W|$ increases.

Low voltage: Direct tunneling regime → FB current > RB current

High voltage: Field emission regime → RB current >> FB current – Prominent asymmetry

Very high voltage: Space charge limited regime → Quantum Child-Langmuir limit [2]

[1] S. Banerjee and P. Zhang, **AIP Adv.** 9, 085302 (2019)

[2] Lau et al., **Phys. Rev. Lett.** (1991)

Assumptions

- This DC model can be applied to the tip-sample junction of a THz-STM. The electron **tunneling time is much smaller** than the timeperiod (in the scale of picosecond) of the THz field [1-3].
- The operating media is **ultrahigh vacuum** with relative permittivity $\epsilon_r = 1$ and electron affinity $X = 0$
- Although the tip is round-shaped, the majority of the tunneling current flows through the region near the tip-apex. We modeled this **tip-apex** as a **square** for simplicity.
- We assume the THz bias voltage $\boldsymbol{V_{THz}(t)}$ **is readily available** at the junction. We do not examine the generation, propagation, coupling of the THz pulses to the STM tip.

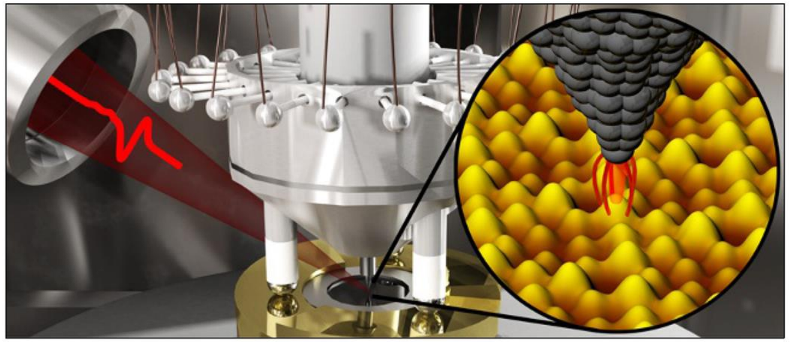
S. Banerjee, and P. Zhang, **Phys. Rev. Applied** 18, 024011 (2022)

Rectification of the Tunneling Junction Depends on:

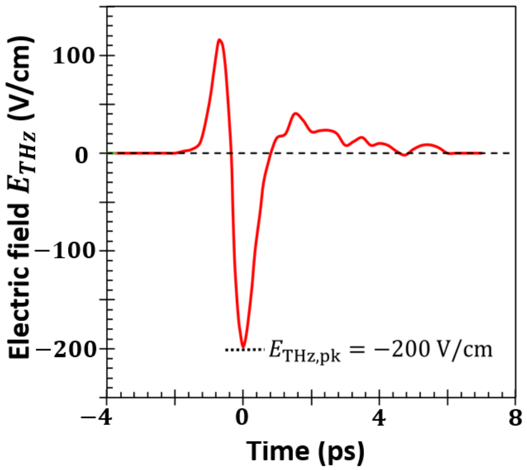
- D.C. bias voltage (polarity, amplitude)
- Incident THz field (polarity, shape, peak amplitude)
- Work functions of STM tip and sample (especially their difference ΔW)
- Tip-sample separation
- Operating media

S. Banerjee, and P. Zhang, **Phys. Rev. Applied** 18, 024011 (2022)

Validation



Experimental set up from [1]

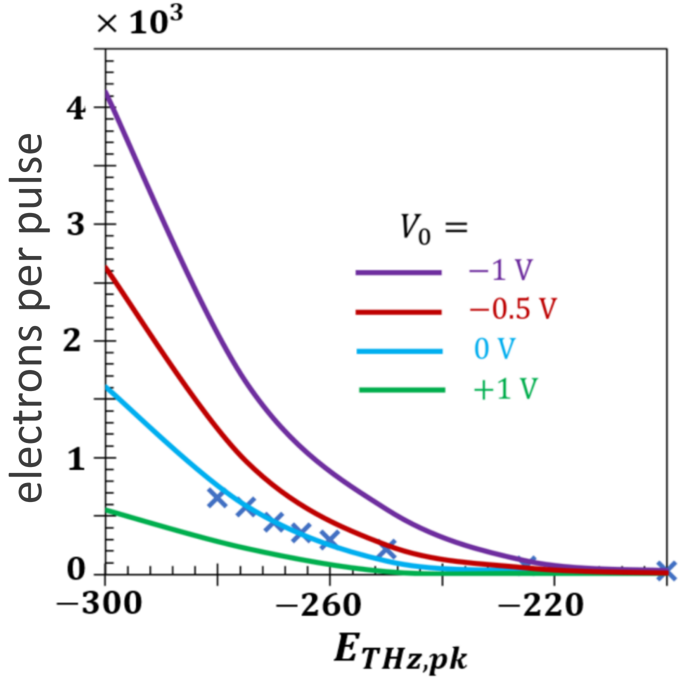


THz electric field from [1]

Number of electrons per pulse $\boldsymbol{N = \int_{t_1}^{t_2} I(t)dt/e}$

$I(t)$ = time dependent tunneling current through the STM tip

$t_2 - t_1$ = pulse duration, e = electron charge



Crossed symbols – experiment [1]

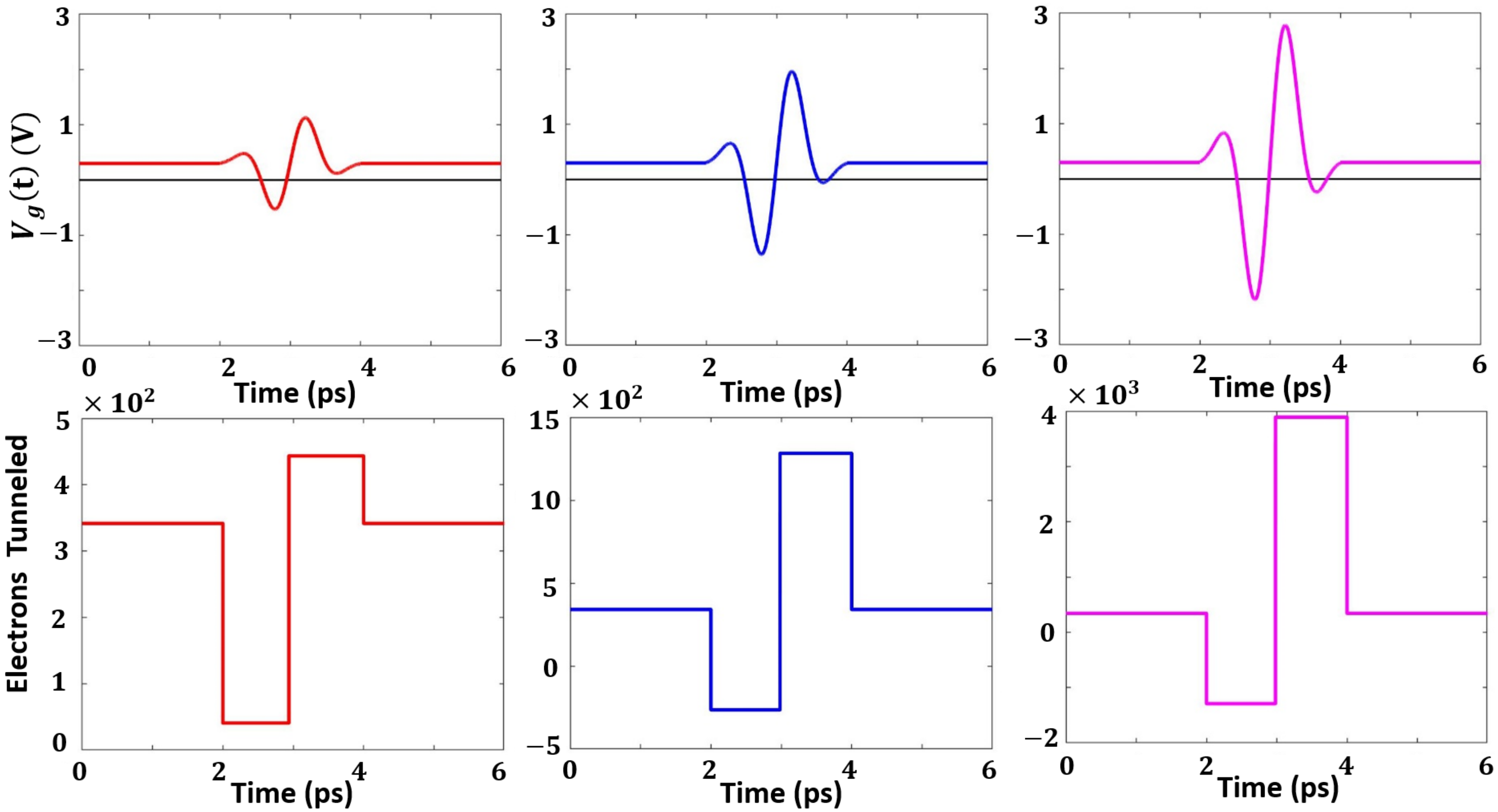
Solid Lines – our theory [2]

Highly non-linear I-V characteristics leads to asymmetry in rectification

[1] V. Jelic et al., **Nat. Phys.** 13, 591 (2017)

[2] S. Banerjee, and P. Zhang, **Phys. Rev. Applied** 18, 024011 (2022)

Effects of time-dependent bias voltage $V_g(t) = V_0 + V_{THz}(t)$



$V_0 = 0.3$ V

$W_s = W_t = 4.5$ eV

Tip height = 0.5 nm

Electrons tunneled = $\int_{t_1}^{t_2} I(t)dt/e$

The rectified electrons per pulse in a THz-STM can be manipulated by varying the bias voltage (polarity, amplitude) and THz field (polarity, shape, peak THz field $E_{THz,pk}$)

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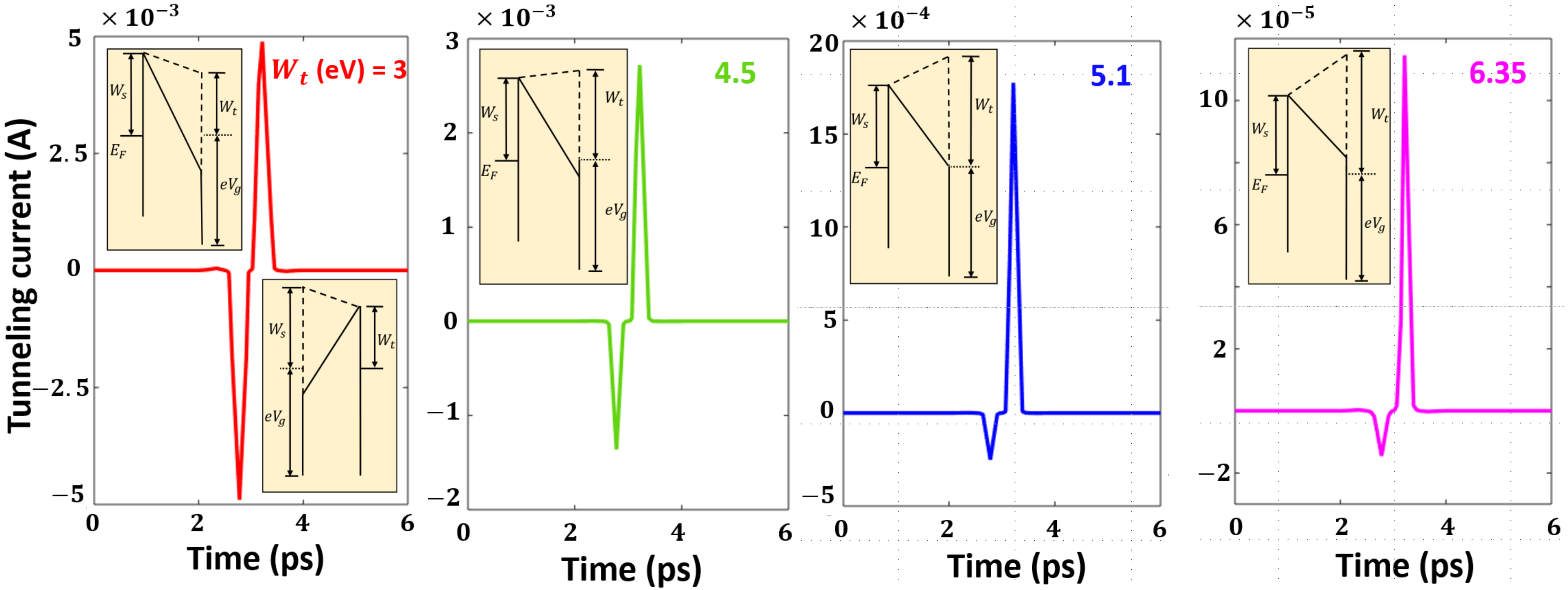
Effects of Different Tip Metals for Al ($W_s = 4.08\text{eV}$) Sample

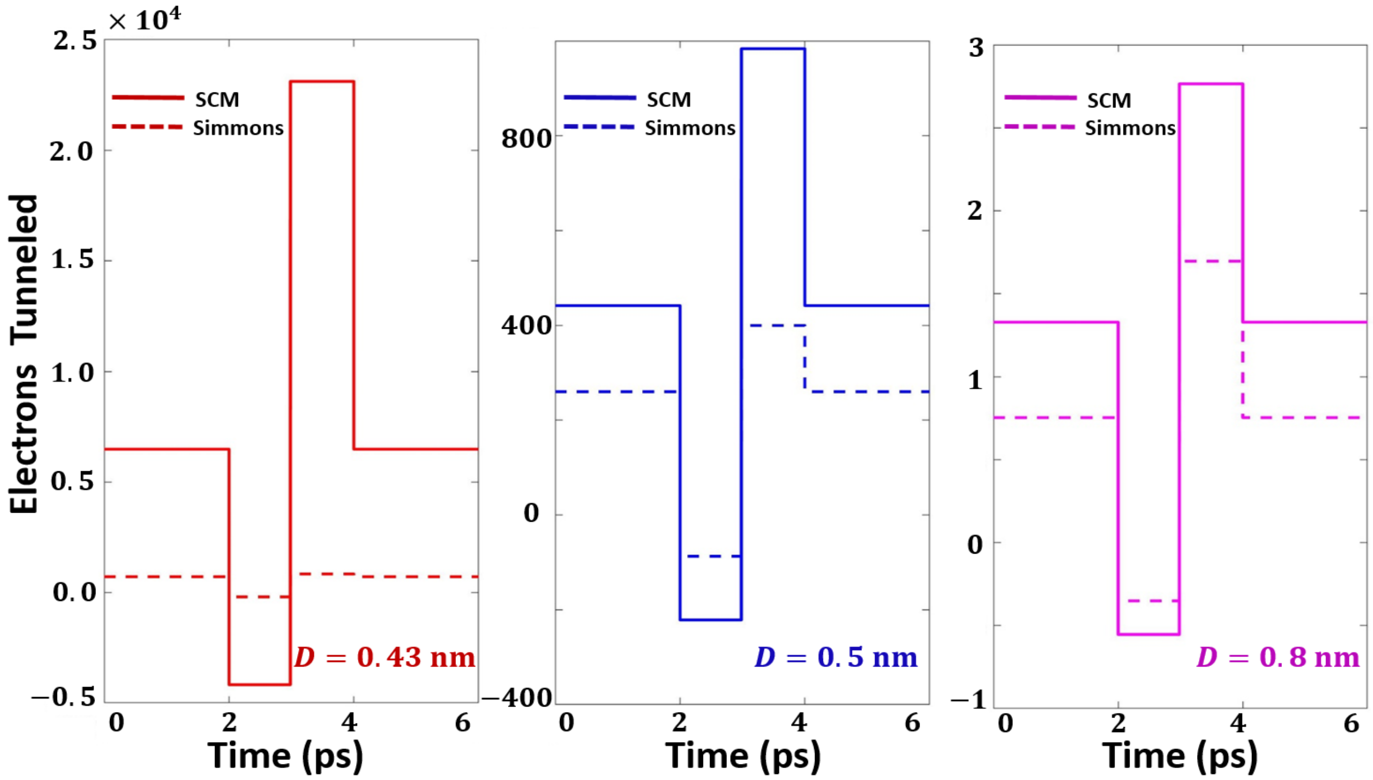


- Work function difference between tip and sample → Asymmetry → **Forward and Reverse Bias**
- In high voltage regime, current is more when lower work function electrode is negatively biased
- Asymmetry increases with the work function difference ΔW

S. Banerjee, and P. Zhang, **Phys. Rev. Applied** 18, 024011 (2022)

Effects of different tip-heights *D* [1]

and Comparison with the Simmons Formula



$V_0 = 0.3$ V

$W_s = W_t = 4.5$ eV

Electrons tunneled = $\int_{t_1}^{t_2} I(t)dt/e$

- For same THz field, the number of tunneled electrons increases with decreasing tip height.
- The difference between Self Consistent Model and Simmons formula [2] increases with decreasing tip height since the **effects of exchange correlation potential** increases as tip height decreases [3].

[1] S. Banerjee, and P. Zhang, **Phys. Rev. Applied** 18, 024011 (2022)
[2] J. G. Simmons, **J. Appl. Phys.** 34, 2581 (1963)
[3] Ang et al., **Phys. Rev. Lett.** 91, 208303 (2003)

Summary

Self-consistent quantum model [1,2,3]
- includes exchange correlation potential & space charge potential
- DC but can be applied here since electron tunneling time << THz timeperiod [4]

Field emission voltage regime
- effects of exchange correlation potential are important
- existing models (Simmons, Bardeen) are unreliable

Asymmetry
- different work-functions of tip and sample
- polarity dependent tunneling current
- reverse bias and forward bias

Rectification depends on:
- d.c. bias voltage (polarity, amplitude)
- incident THz field (polarity, shape, $E_{THz,pk}$)
- work functions of STM tip and sample (especially their difference ΔW)
- tip-sample separation.

→

Manipulation of electron transport
- better rectification can be obtained by enhancing the asymmetry
- asymmetrical THz field, reverse bias, larger workfunction difference improves rectification

[1] P. Zhang, **Sci. Rep.** 5, 9826 (2015)

[2] S. Banerjee and P. Zhang, **AIP Adv**. 9, 085302 (2019)

[3] S. Banerjee, and P. Zhang, **Phys. Rev. Applied** 18, 024011 (2022)

[4] G. Nimtz, and H. Aichmann, **Z. Für Naturforschung A** 76, 295–297 (2021)

Future Work

Tunneling Probability

- although widely used, WKBJ approximation need to be examined
- time-independent Schrödinger equation may be solved numerically to get a more accurate transmission probability.

Tip-sample geometry

- we considered the tip and sample surface to be flat and the problem is one-dimensional
- the effects of their geometry may be included in future works

Time dependence

- time-dependent analysis of the electron wave function might be performed
- results might be compared to define regimes where the proposed simple DC model is applicable

S. Banerjee, and P. Zhang, **Phys. Rev. Applied** 18, 024011 (2022)

Acknowledgement

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Thank you!