MICHIGAN STATE UNIVERSITY

Photoemission from Biased Metal Surfaces

Quantum efficiency, Laser heating, Dielectric Coatings, and Quantum Pathways Interference

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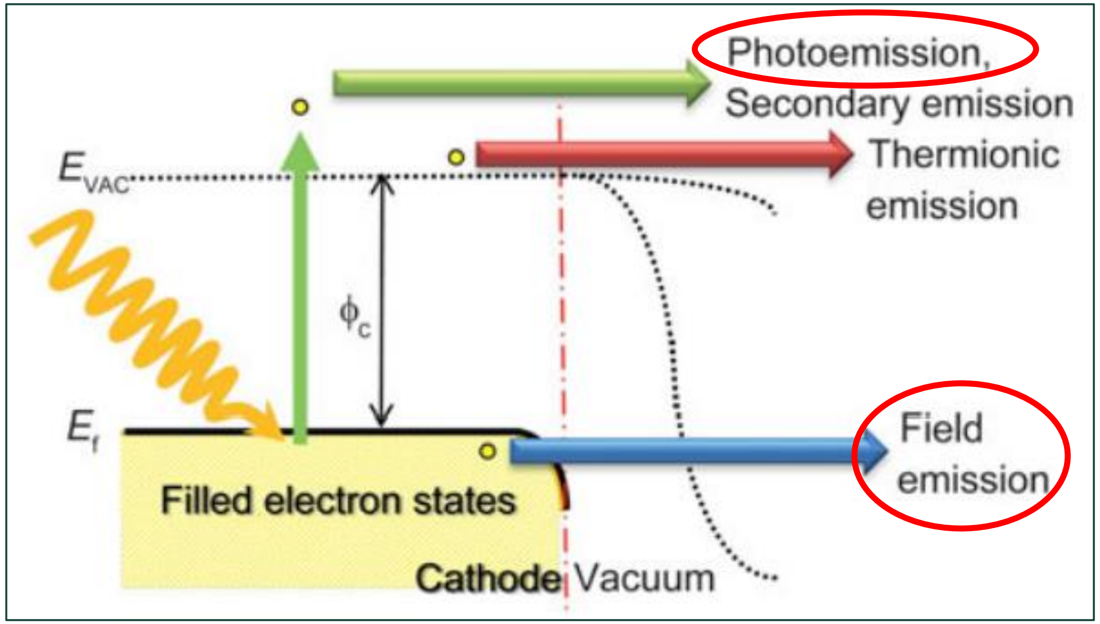
Outline

- **Background & Motivation**
- **Research Overview**
- **Effects of laser wavelength & heating**
- **Effects of cathode surface coating**
- **Summary**

Background and Motivation

- **Electron emission** is important to fundamental research and various applications.



Trucchi, D. M., and Melosh, N. A.. Mrs Bulletin 42, no. 7, 488-492 (2017).

HOME > SCIENCE > VOL. 356, NO. 6333 > REAL-TIME SPECTRAL INTERFEROMETRY PROBES THE INTERNAL DYNAMICS OF FEMTOSECOND SOLITON...

RESEARCH ARTICLE

Real-time spectral interferometry probes the internal dynamics of femtosecond soliton molecules

G. HERINK, F. KURTZ, B. JALALI, D. R. SOLLI, AND C. ROPERS Authors Info & Affiliations

SCIENCE • 7 Apr 2017 • Vol 356, Issue 6333 • pp. 50-54 • DOI: 10.1126/science.aal5326

nature > nature physics > articles > article

Published: 06 November 2017

Phase ordering of charge density waves traced by ultrafast low-energy electron diffraction

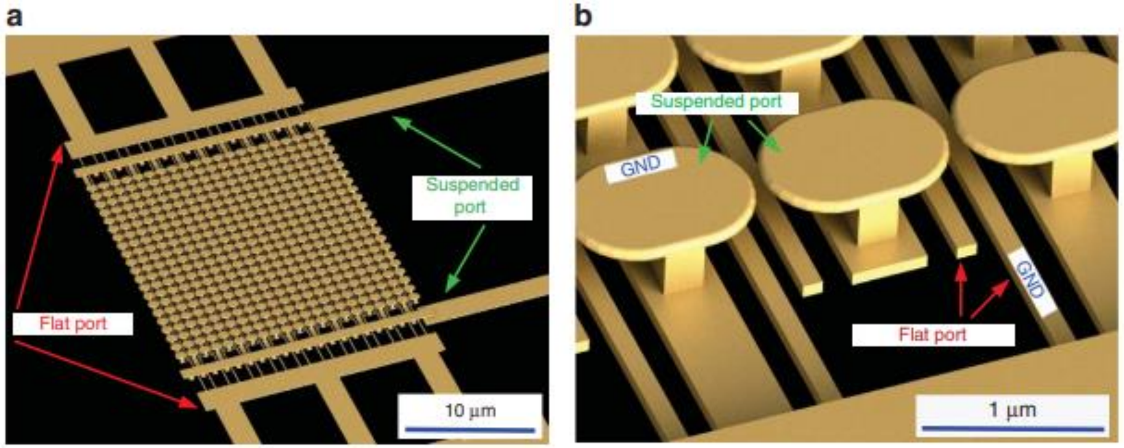
S. Vogelgesang, G. Storeck, J. G. Horstmann, T. Diekmann, M. Sivis, S. Schramm, K. Rossnagel, S. Schäfer & C. Ropers

Nature Physics **14**, 184–190 (2018) | Cite this article

Background and Motivation

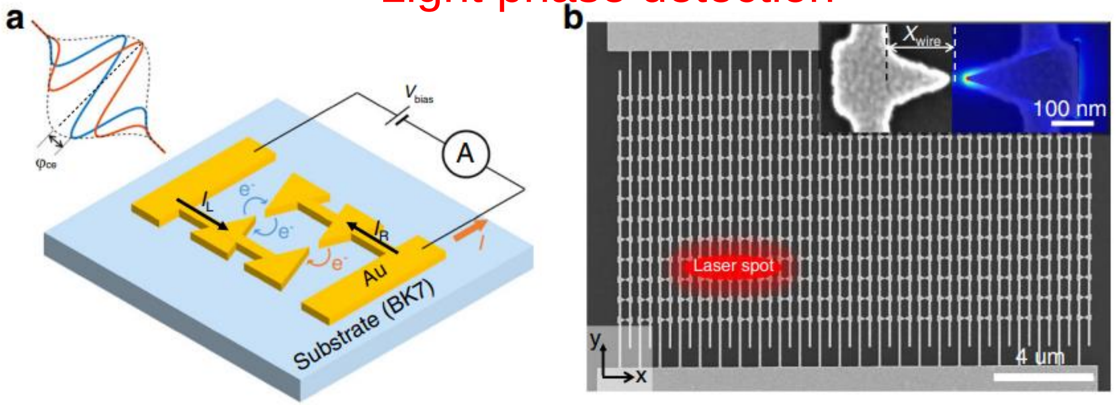
- **Electron emission** is important to fundamental research and various applications.

Photoemission-based device



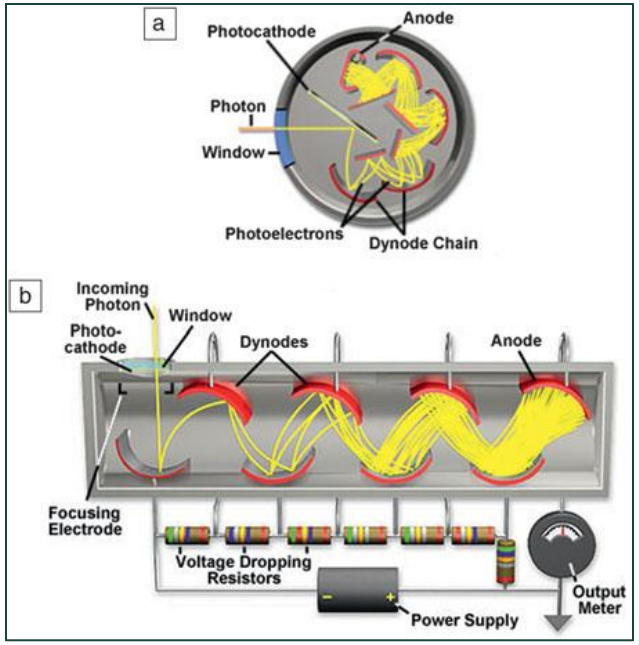
Forati, E., Dill, T.J., Tao, A.R. and Sievenpiper, D. *Nature communications*, *7*(1), p.13399 (2016).

Light phase detection



Yang, Y., et al.. *Nature communications*, *11*(1), p.3407 (2020).

Photomultiplier



Trucchi, D. M., and Melosh, N. A.. Mrs Bulletin 42, no. 7, 488-492 (2017).

Background and Motivation

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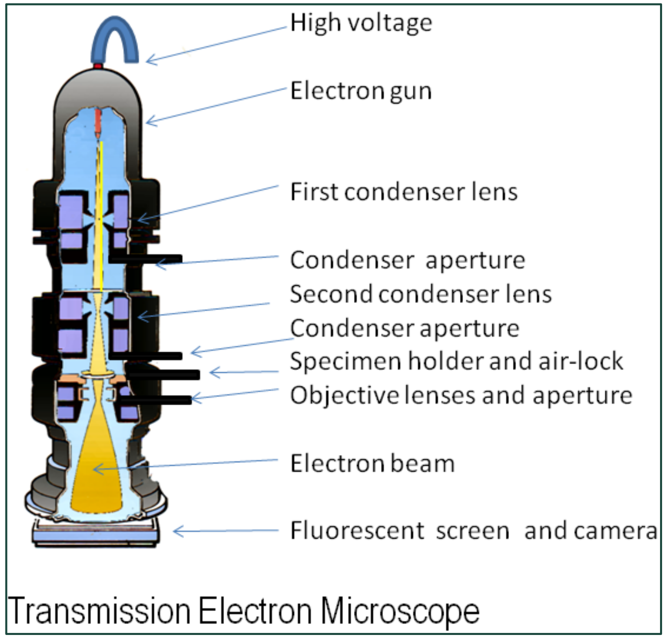
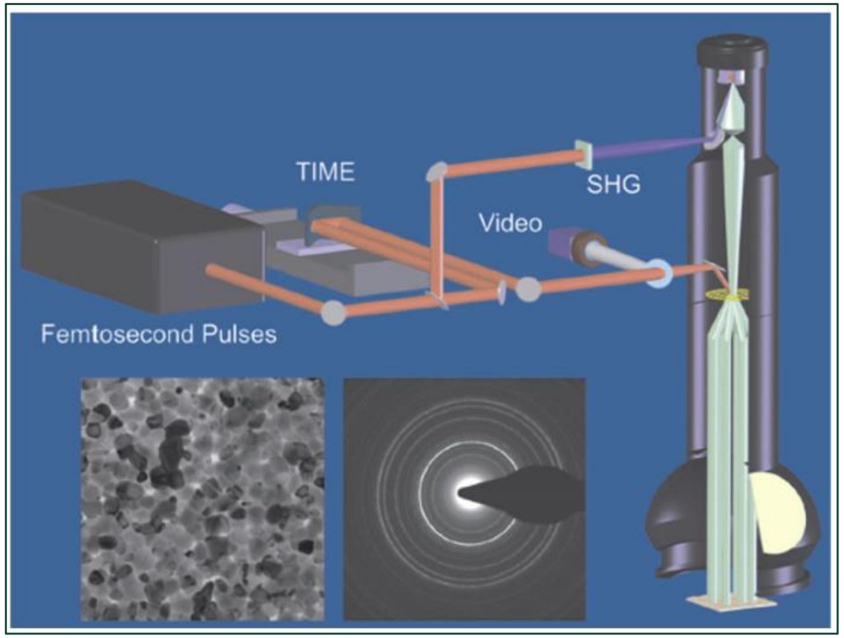


https://en.wikipedia.org/wiki/Electron_microscope#/media/File:Electron_Microscope.png

Ultrafast electron microscope



Grinolds, M.S., et al. *Proceedings of the National Academy of Sciences*, *103*(49), pp.18427-18431 (2006).

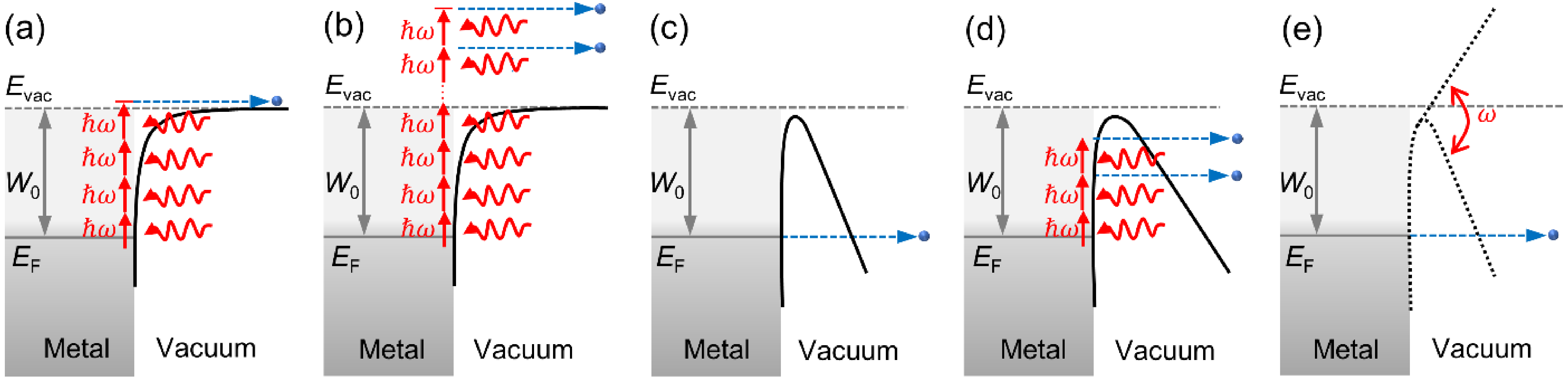
Photocathode for accelerator

https://web.physics.ucsb.edu/~smcbride/photocathodes/

High quantum efficiency, low emittance, long lifetime, and short response time

Background and Motivation

- Laser-matter interaction is primarily affected by **the laser wavelength**
  - a wide range of laser wavelength (~250 nm, ~800 nm, mid-infrared, THz)



multiphoton photoemission

above-threshold photoemission

field emission

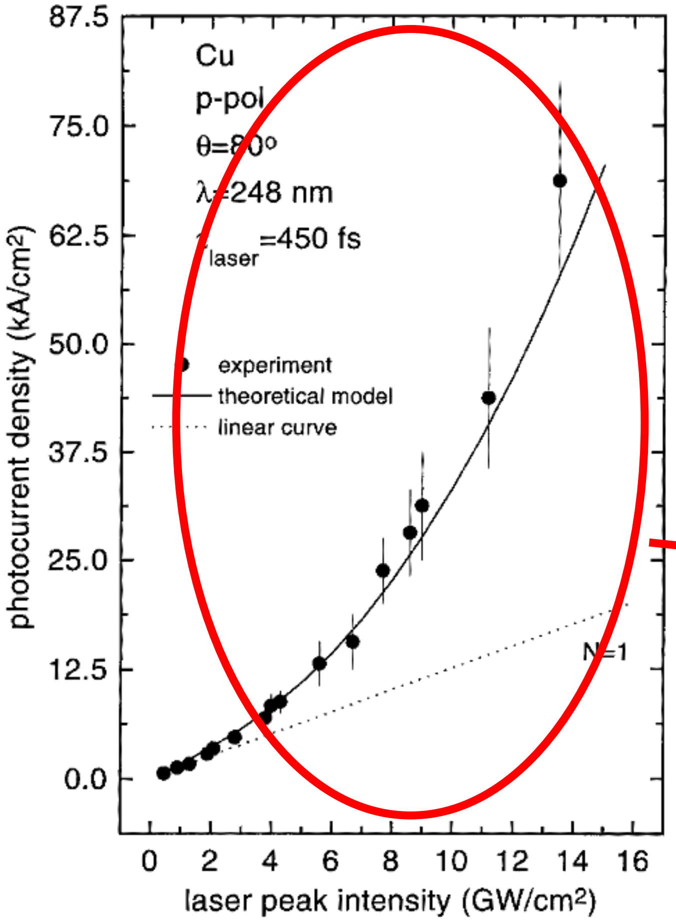
photo-assisted field emission

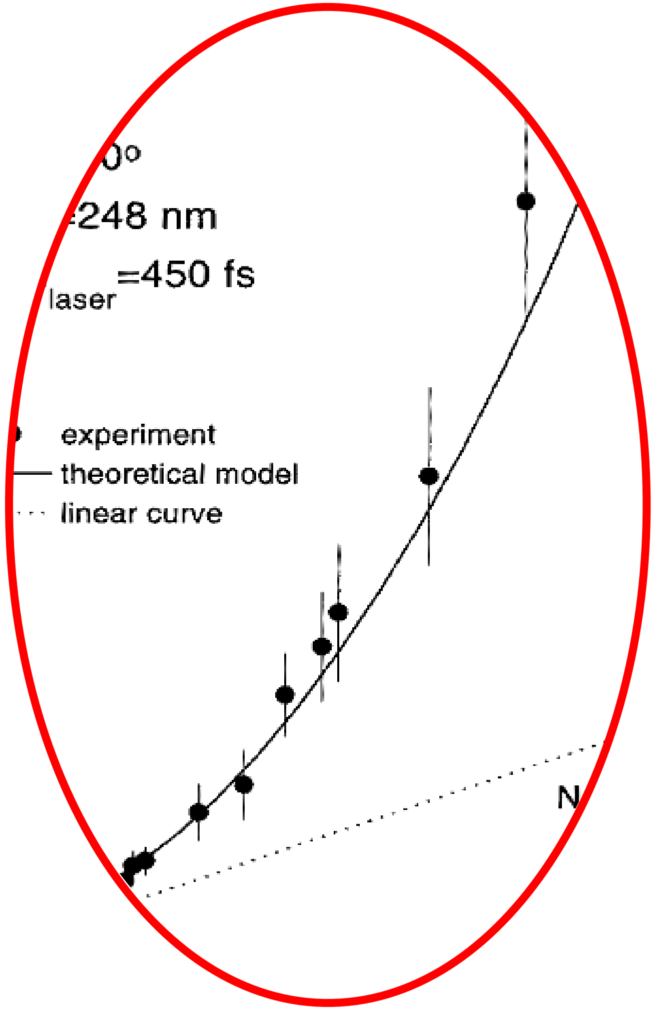
optical field emission

electron emission mechanisms

Background and Motivation

- Laser acts as a heat source



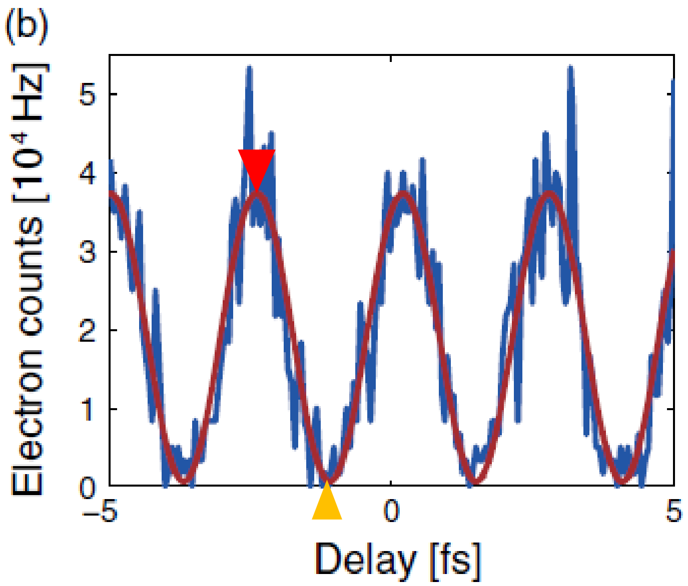


Papadogiannis, N. A., S. D. Moustaïzis, and J. P. G rardeau-Montaut. *J. Phys. D: Appl. Phys* 30(17), 2389 (1997).

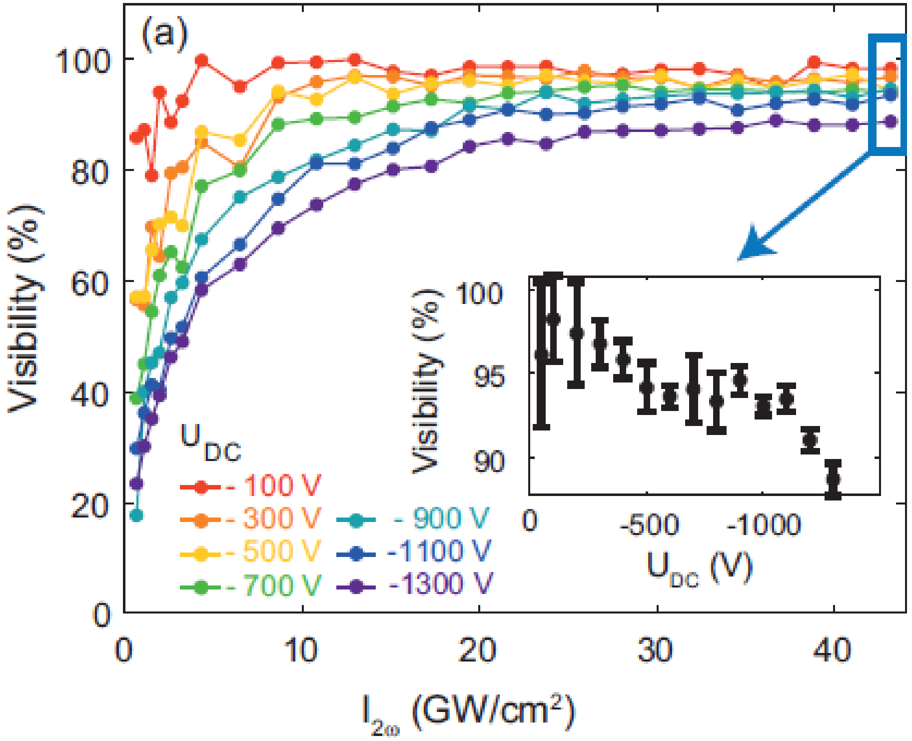
Background and Motivation

- Two-color laser coherent control of photoemission

$$\frac{I_{max} - I_{min}}{I_{max} + I_{min}}$$



Förster, M., et al. *Phys. Rev. Lett.* 117.21 (2016): 217601.

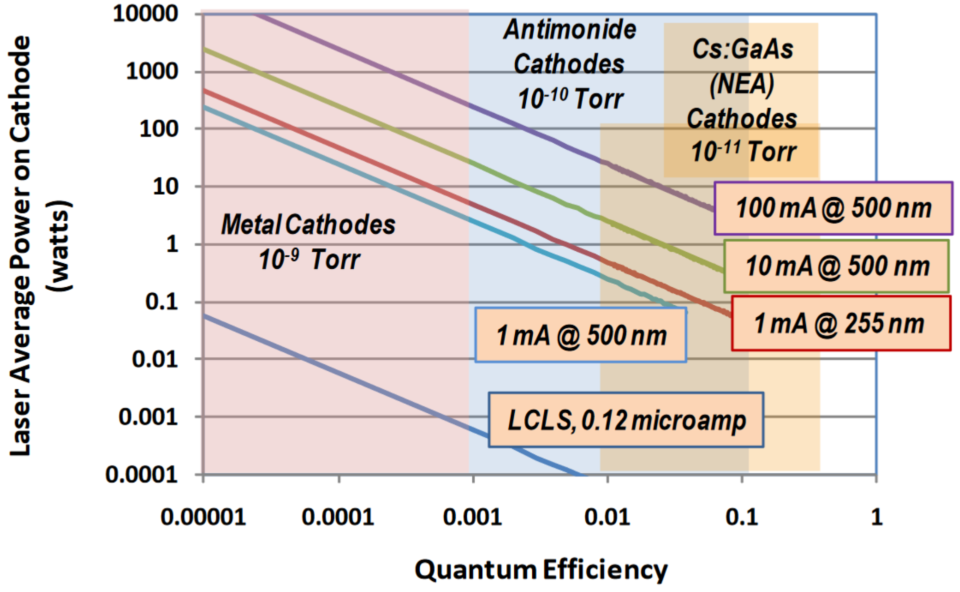


Dienstbier, Philip, Timo Paschen, and Peter Hommelhoff. *J. Phys. B: Atomic, Molecular and Optical Physics* 54.13 (2021): 134002.

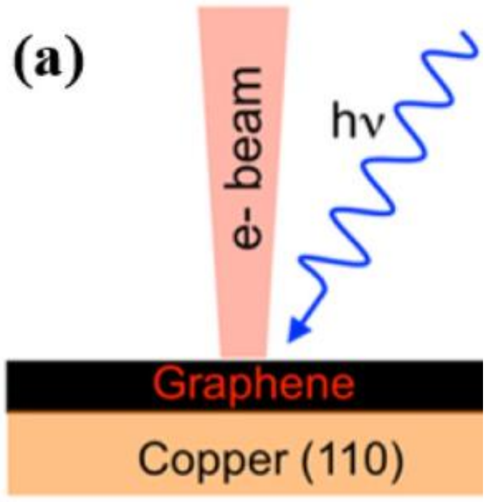
Two-color laser, DC field?

Background and Motivation

- Cathode surface status influences the fundamental mechanisms of photoemission
  - **Metals** and semiconductors
  - Artificial **coatings** (graphene, nano-diamond, silicon dioxide, and zinc dioxide)
  - Natural **coatings**: oxide



Dowell, D. H., et al. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 622(3), 685-697 (2010).



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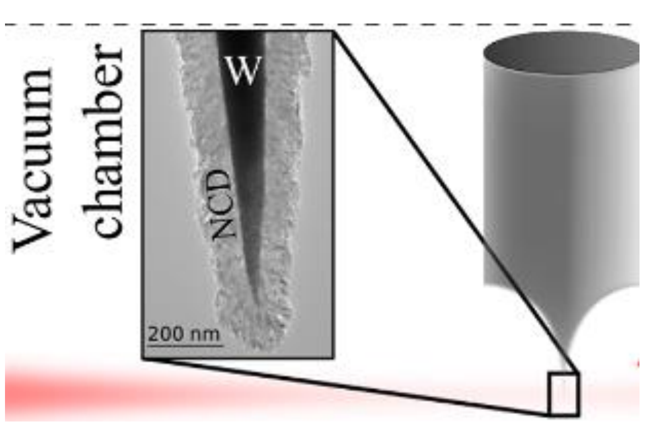

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Research Overview

Photoemission

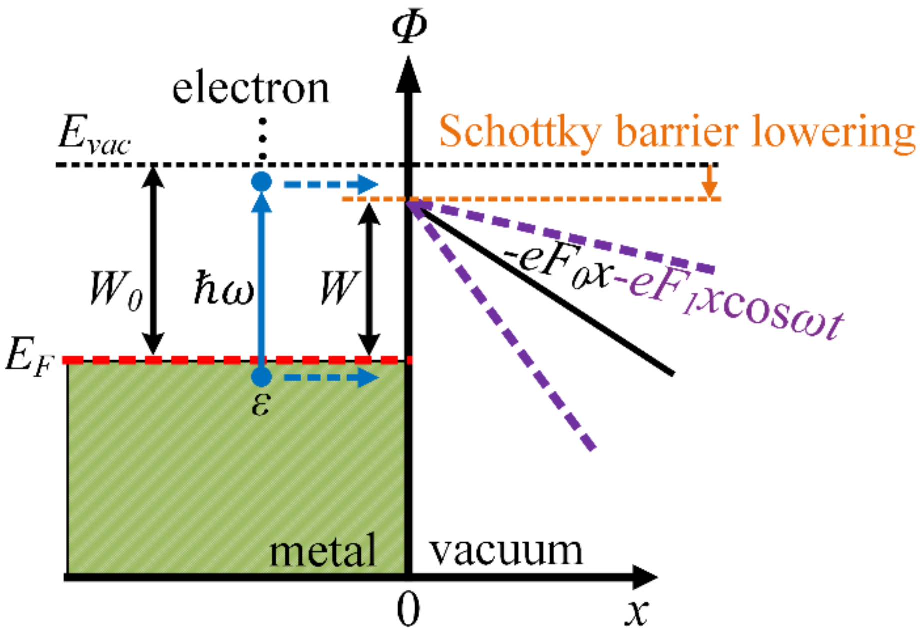
Laser effects

- Effects of laser wavelength and **accompanying heating**
- **Two-color** laser coherent control of photoemission

Status of metallic photocathode

- **Dielectric coatings** effects
- **Plasmon** resonance enhancement
- Quantum **well** effects

Effects of laser wavelength & heating



E_{vac}: vacuum level
W_0: work function of metal
E_F: Fermi energy
ε: electron initial energy
F_0: dc field
F_1: laser field
ω: laser frequency
$W=W_0-W_{Schottky}$: effective work function

- Potential profile $$\phi(x,t) = \begin{cases} 0, & x<0 \\ E_F + W - eF_0x - eF_1x\cos\omega t, & x>0 \end{cases}$$
- Schrödinger equation $$i\hbar\frac{\partial\psi(x,t)}{\partial t} = -\frac{\hbar^2}{2m}\frac{\partial^2\psi(x,t)}{\partial x^2} + \phi(x,t)\psi(x,t)$$
- Electron waves $$\psi_I(x,t) = e^{-i\frac{\varepsilon}{\hbar}t+i\sqrt{\frac{2m\varepsilon}{\hbar^2}}x} + \sum_{n=-\infty}^{n=\infty} \boldsymbol{R_n} e^{-i\frac{\varepsilon+n\hbar\omega}{\hbar}t-i\sqrt{\frac{2m(\varepsilon+n\hbar\omega)}{\hbar^2}}x}, x<0$$

Y. Zhou, and P. Zhang, J. Appl. Phys. 127, 164903 (2020).

Effects of laser wavelength & heating

- Electron waves

$$\psi_{II}(x,t) = \sum_{n=-\infty}^{n=\infty} \boldsymbol{T_n} e^{i\sqrt{\frac{2mE_n}{\hbar^2}}\xi} e^{-i\frac{\varepsilon+n\hbar\omega}{\hbar}t} e^{\frac{ieF_1 \sin \omega t}{\hbar\omega}x+\frac{ie^2F_1^2 \sin 2\omega t}{8\hbar m\omega^3}}, x \geq 0 \qquad \boldsymbol{F_0 = 0}$$

$$\psi_{II}(x,t) = \sum_{n=-\infty}^{n=\infty} \boldsymbol{T_n}[Ai(-\eta_n) - iBi(-\eta_n)]\boldsymbol{\Gamma}, x \geq 0 \qquad \boldsymbol{F_0 \neq 0}$$

with $\boldsymbol{\Gamma} = e^{i\sqrt{\frac{2mE_n}{\hbar^2}}\xi} e^{-i\frac{\varepsilon+n\hbar\omega}{\hbar}t} e^{\frac{ieF_1 \sin \omega t}{\hbar\omega}x+\frac{ie^2F_1^2 \sin 2\omega t}{8\hbar m\omega^3}} e^{-\frac{ie^2F_0F_1 \sin \omega t}{\hbar m\omega^3}}$

- Boundary conditions: ψ and $\partial\psi/\partial x$ continuous at the interface→ $\boldsymbol{R_n}$ & $\boldsymbol{T_n}$
- **Electron transmission probability** at electron initial energy ε is defined as $\boldsymbol{w(\varepsilon,x,t)} = \frac{\boldsymbol{j_t}}{\boldsymbol{j_i}}$, with $j = \frac{i\hbar}{2m}(\psi\nabla\psi^* - \psi^*\nabla\psi)$
- Time-averaged **electron transmission probability**

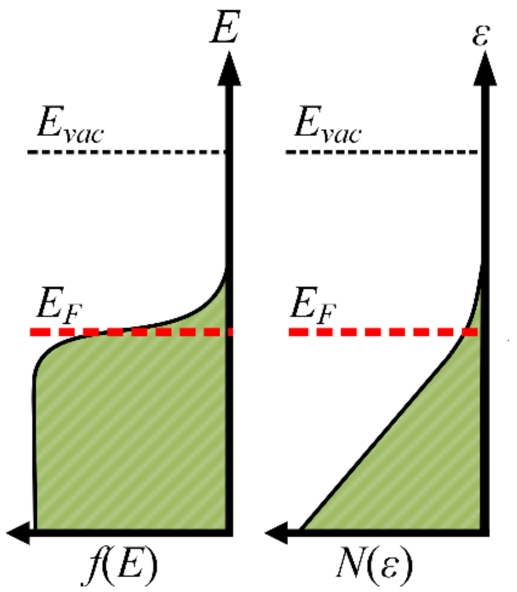
$$w_n(\varepsilon) = \begin{cases} Im\left(i\frac{\sqrt{\varepsilon+n\hbar\omega-V_0}}{\sqrt{\varepsilon}}|T_n|^2\right), \boldsymbol{F_0 = 0} \\ \frac{1}{\pi}\left[\frac{eF_0\hbar}{\sqrt{2m}}\right]^{1/3}\frac{W^{1/3}}{\varepsilon}, \boldsymbol{F_0 \neq 0} \end{cases}$$

- **Electron transmission probability**:

$$D(\varepsilon) = \sum_{n=-\infty}^{\infty} w_n(\varepsilon)$$

Y. Zhou, and P. Zhang, J. Appl. Phys. 127, 164903 (2020).

Effects of laser wavelength & heating



E_{vac}: vacuum level
E_F: Fermi energy
ε: electron longitudinal energy
E: electron energy, including both traverse and longitudinal energy

$f(E)$: Fermi-Dirac distribution $$f(E) = \frac{1}{\exp\left(\frac{E - E_F}{k_B T}\right) + 1}$$

$N(\varepsilon)$: supply function $$N(\varepsilon) = \frac{m k_B T}{2\pi^2 \hbar^3} \ln\left[1 + \exp\left(\frac{E_F - \varepsilon}{k_B T}\right)\right]$$

- Emission current density

$$J = e \int_0^\infty D(\varepsilon) N(\varepsilon) d\varepsilon$$

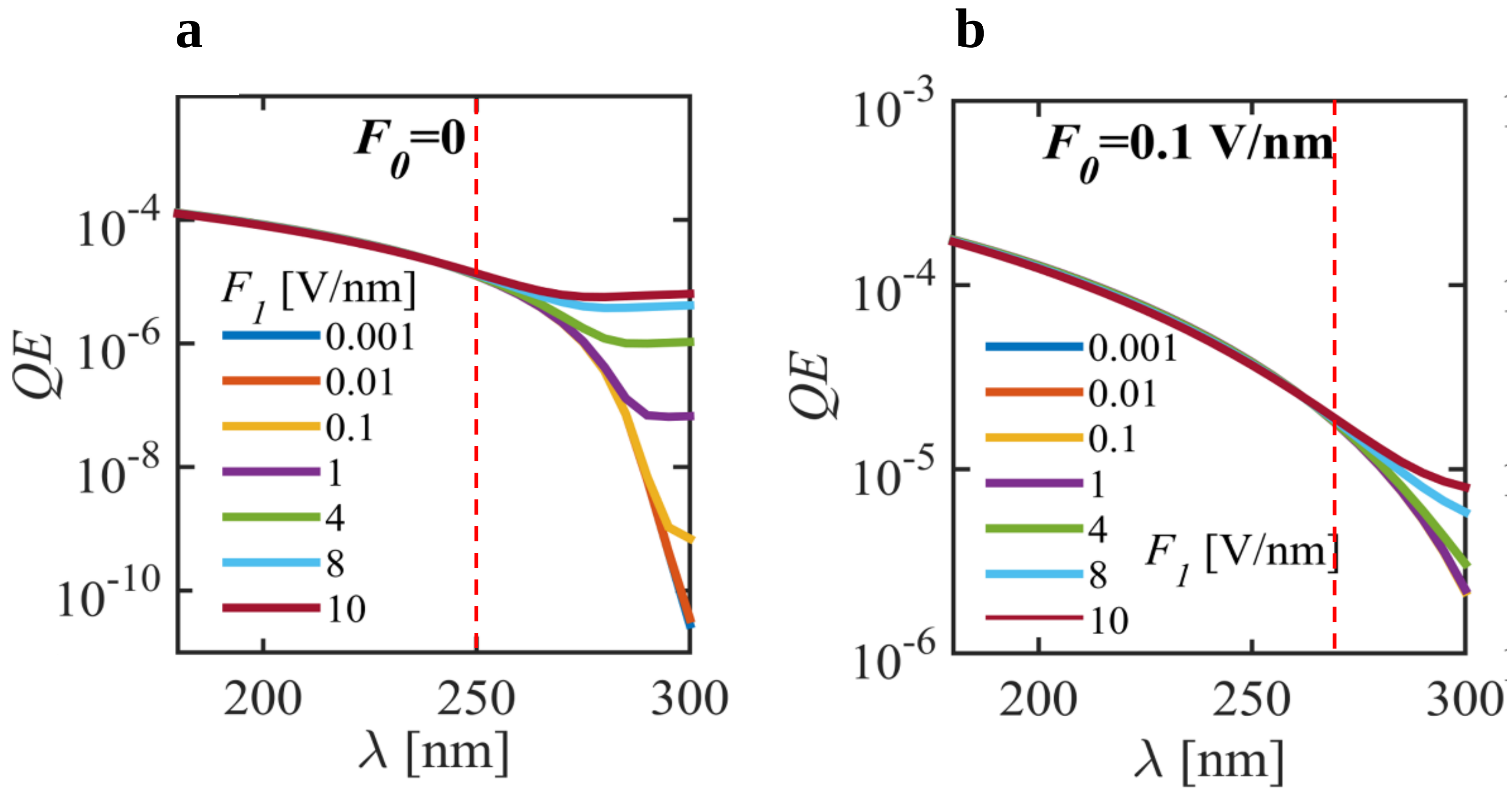
where $N(\varepsilon)d\varepsilon$ is the number density of electrons inside metal with longitudinal energy between ε and $\varepsilon + d\varepsilon$ impinging on the surface of metal per unit time.

- The quantum efficiency (QE)

$$QE = \frac{J/e}{I/\hbar\omega}$$

Y. Zhou, and P. Zhang, J. Appl. Phys. 127, 164903 (2020).

Effects of laser wavelength & heating

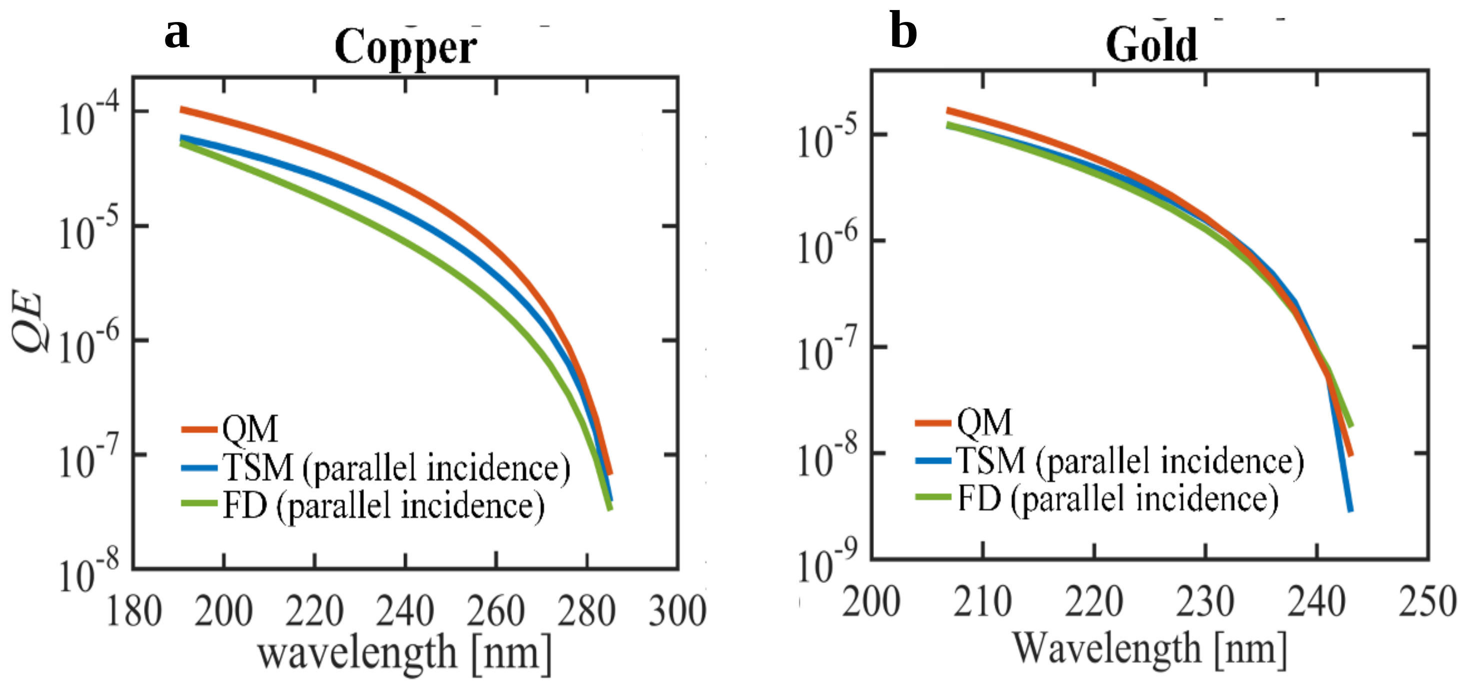


Copper
E_F = 7eV
W_0 = 4.31eV
T=300 K.

$$I \propto F_1^{2n},\ QE \propto F_1^{2(n-1)},\ n = \left\langle \frac{W}{\hbar\omega} + 1 \right\rangle$$

Y. Zhou, and P. Zhang, J. Appl. Phys. 127, 164903 (2020).

Effects of laser wavelength & heating



$F_1 = 0.01$ V/nm
$F_0 = 0$

FD: $QE(\omega) = a_1 \alpha A T^2 F(\frac{\hbar\omega - W}{k_B T})$

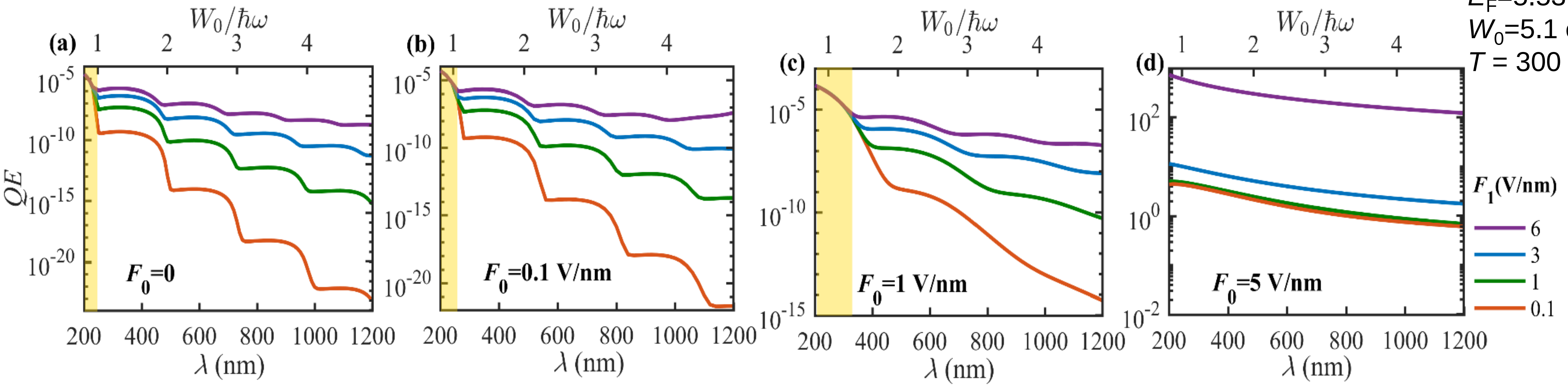
TSM: $QE(\omega) = \alpha F_{e-e}(\omega) \frac{(E_F + \hbar\omega)}{2\hbar\omega}\left[1 + \frac{E_F + W}{E_F + \hbar\omega} - 2\sqrt{\frac{E_F + W}{E_F + \hbar\omega}}\right]$

$\alpha = \frac{\pi \delta_s}{\lambda}$ for **parallel incidence**

Y. Zhou, and P. Zhang, J. Appl. Phys. 127, 164903 (2020).

Effects of laser wavelength & heating

Au:
E_F=5.53 eV
W_0=5.1 eV
T = 300 K



$$QE \propto F_1^{2(n-1)},\ n = \left\langle \frac{W}{\hbar\omega} + 1 \right\rangle$$

- F_0 **shifts** step points and **smooths** curves
- Dominant **direct tunneling** for $F_0 = 5$ V/nm

Y. Zhou and P. Zhang, J. Appl. Phys. 130, 064902 (2021).

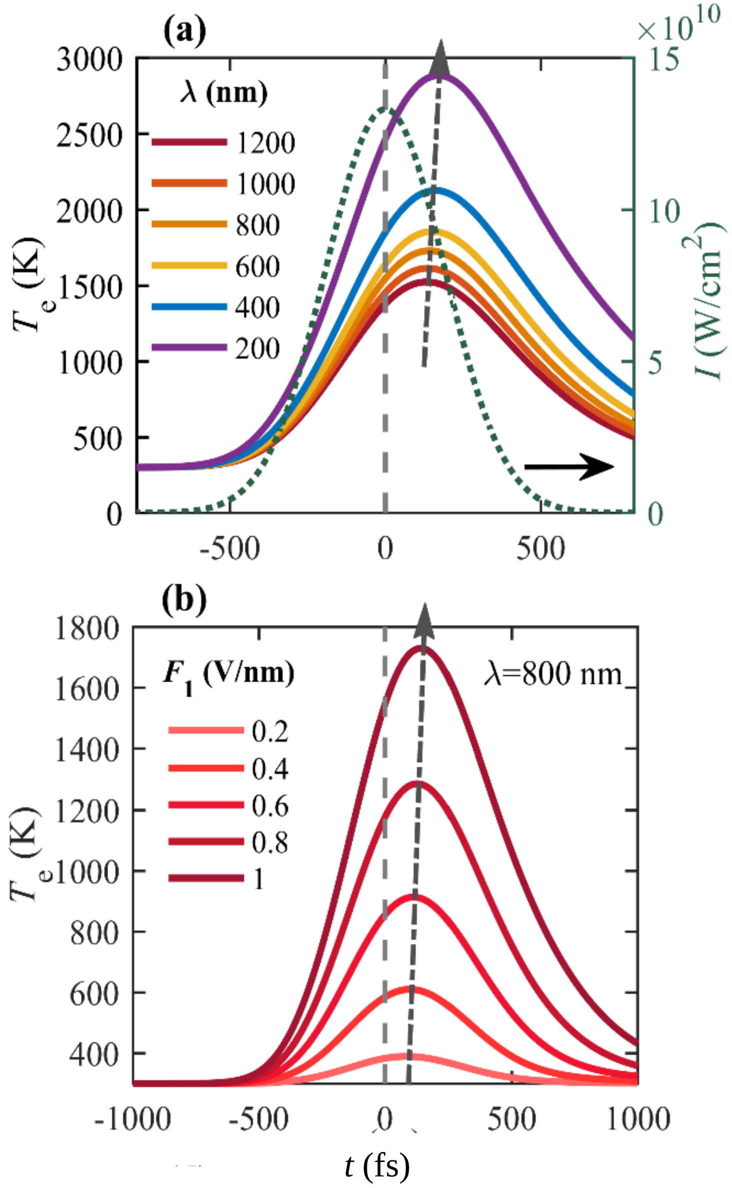
Effects of laser wavelength & heating

Two-temperature model

$$C_e \frac{\partial T_e}{\partial t} = \frac{\partial}{\partial x} \kappa \frac{\partial T_e(x,t)}{\partial x} - g(T_e - T_l) + G(x,t)$$

$$C_l \frac{\partial T_l}{\partial t} = g(T_e - T_l)$$

- C_e and C_l are the electron and lattice capacity;
- T_e and T_l are the electron and lattice temperature;
- g is the electron-lattice coupling coefficient;
- $G(x,t) = I(t) P_{abs} \alpha \exp(-\alpha x)$ is the energy absorbed by the metal;
- κ is the thermal conductivity.



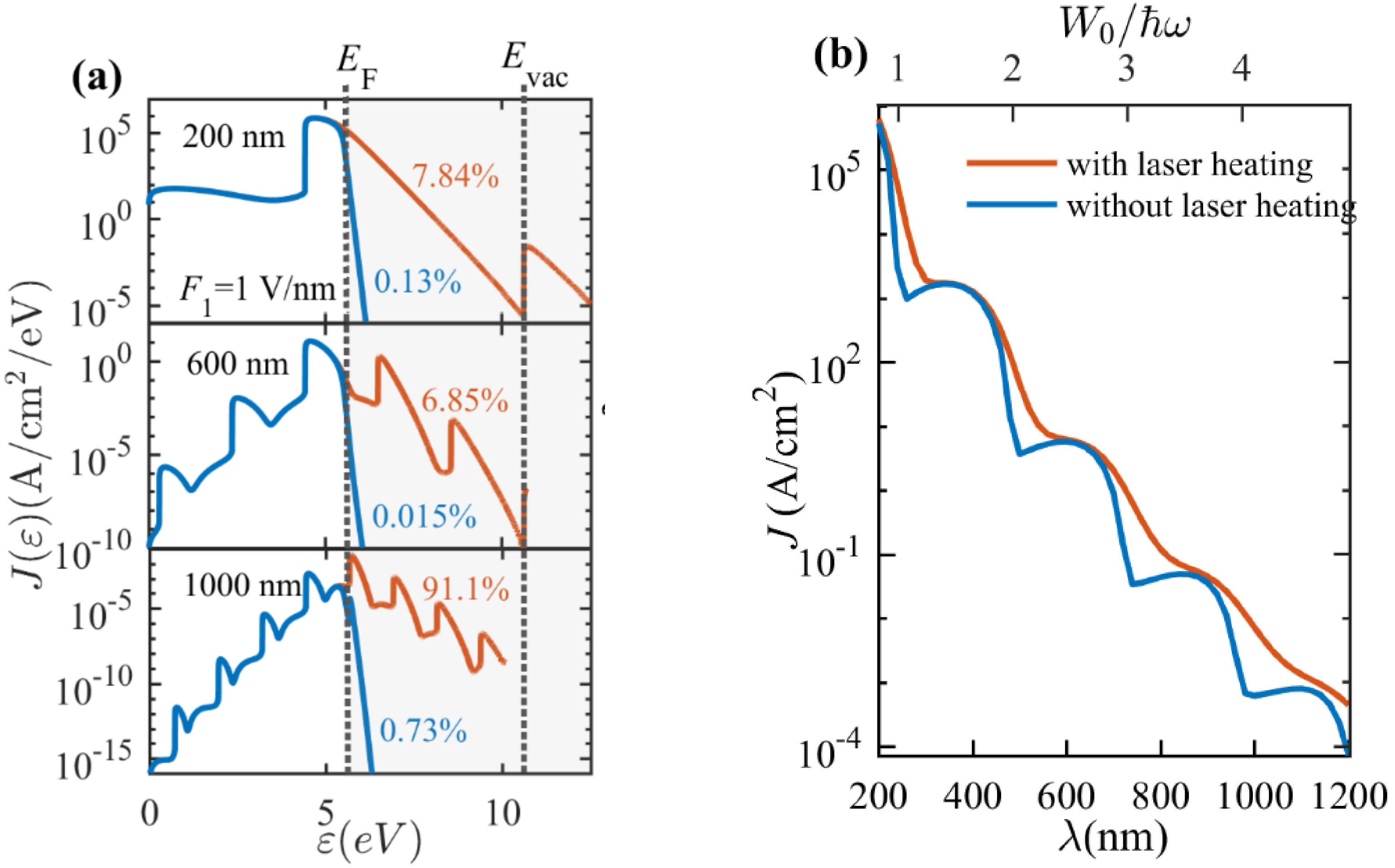
[1] S. I. Anisimov, et al., Sov. Phys. JETP 39, 375 (1974). [2] Y. Zhou and P. Zhang, J. Appl. Phys. 130, 064902 (2021).

Effects of laser wavelength & heating

$$J(\varepsilon) = eD(\varepsilon)N(\varepsilon)$$

$$J = \int_0^\infty J(\varepsilon)d\varepsilon$$

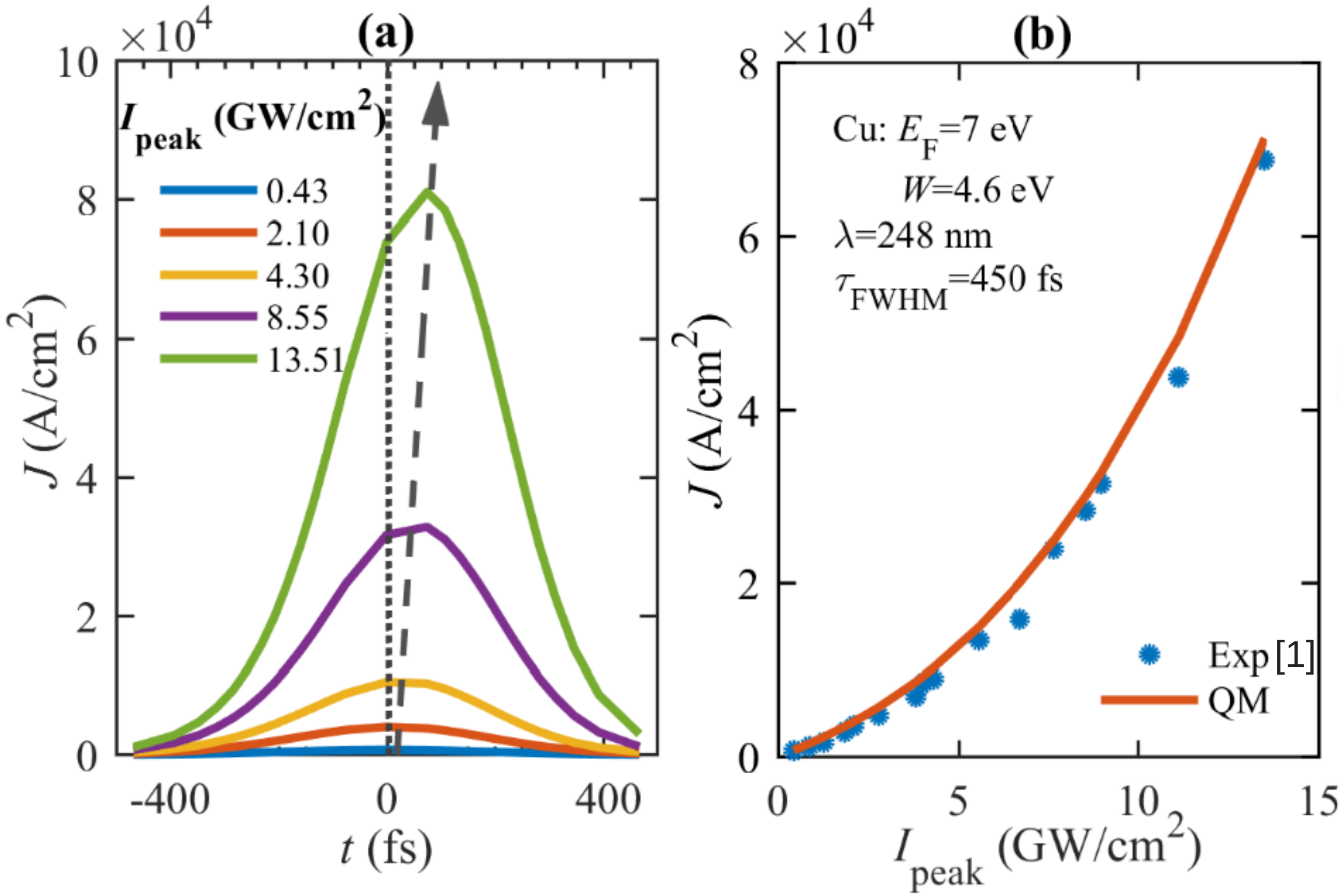
$$t = 0$$



- Laser heating results in more electron emission from initial energy **above Fermi level.**
- Laser heating has greater effects at **step points** and **long laser wavelengths**.

[1] Y. Zhou and P. Zhang, J. Appl. Phys. 130, 064902 (2021).

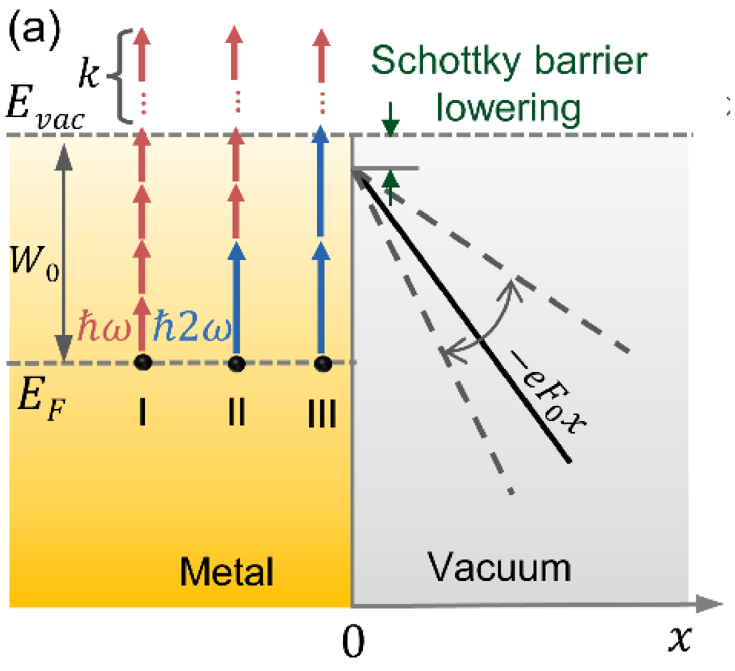
Effects of laser wavelength & heating

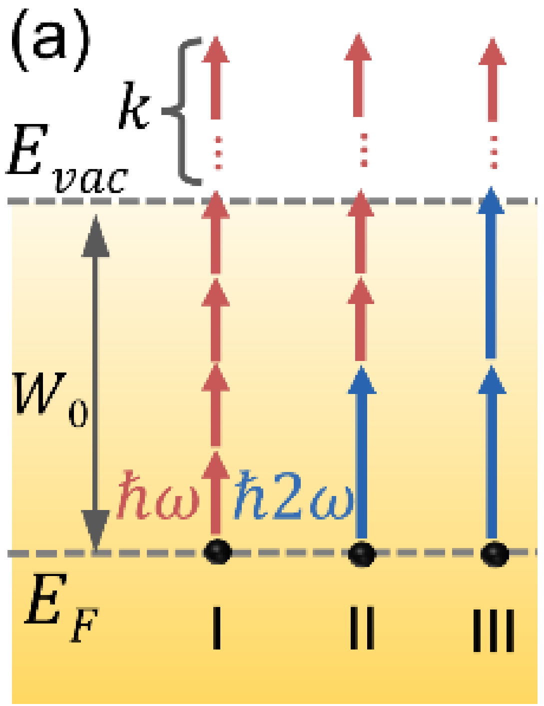


- Good **agreement** with experimental results.
- **Lag** between emission current peak and laser intensity peak.

[1] N. A. Papadogiannis et al., J. Phys. D: Appl. Phys. 30, 17 (1997). [2] Y. Zhou and P. Zhang, J. Appl. Phys. 130, 064902 (2021).

Two-color laser coherent control of photoemission





- Potential profile

$$\phi(x,t) = \begin{cases} 0, & x < 0 \\ V_0 - ef(t)x - eF_0x, & x \geq 0 \end{cases}$$

where $V_0 = E_F + W$, $W = W_0 - W_{Schottky}$, $f(t) = F_1\cos(\omega t) + F_2\cos(2\omega t + \theta)$

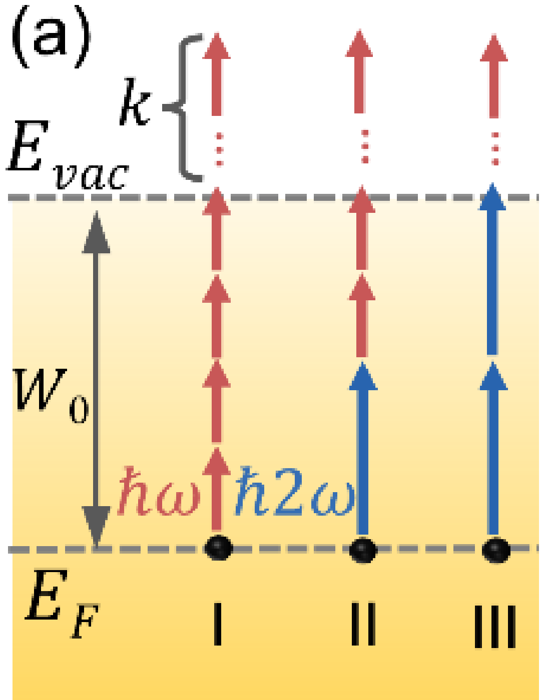
- Time-dependent Schrödinger equation

$$i\hbar\frac{\partial\psi(x,t)}{\partial t} = -\frac{\hbar^2}{2m}\frac{\partial^2\psi(x)}{\partial x^2} + \phi(x,t)\psi(x)$$

Y. Zhou, and P. Zhang, Phys. Rev. B. 106, 085402 (2022).

Two-color laser coherent control of photoemission

- $D_I \propto \alpha^k (F_1^2)^k \left(\alpha^4 (F_1^2)^4\right) = K_I;$
- $D_{II} \propto \alpha^k (F_1^2)^k \left(\zeta^2 (F_1^2)^2 F_2^2\right) = K_{II} F_2^2;$
- $D_{III} \propto \alpha^k (F_1^2)^k \left(\beta^2 (F_2^2)^2\right) = K_{III} (F_2^2)^2;$
- $D_{I\&II} \propto 2\sqrt{D_I D_{II}} \cos\theta \propto \alpha^k (F_1^2)^k \left(2\alpha^2 \zeta (F_1^2)^3 \sqrt{F_2^2} \cos\theta\right) = K_{I\&II} \sqrt{F_2^2} \cos(2\omega\tau);$
- $D_{I\&III} \propto 2\sqrt{D_I D_{III}} \cos 2\theta \propto \alpha^k (F_1^2)^k \left(2\alpha^2 (F_1^2)^2 \beta F_2^2 \cos 2\theta\right) = K_{I\&III} F_2^2 \cos(4\omega\tau);$
- $D_{II\&III} \propto 2\sqrt{D_{II} D_{III}} \cos\theta \propto \alpha^k (F_1^2)^k \left(2\zeta F_1^2 \beta \sqrt{(F_2^2)^3} \cos\theta\right) = K_{II\&III} \& \sqrt{(F_2^2)^3} \cos(2\omega\tau).$

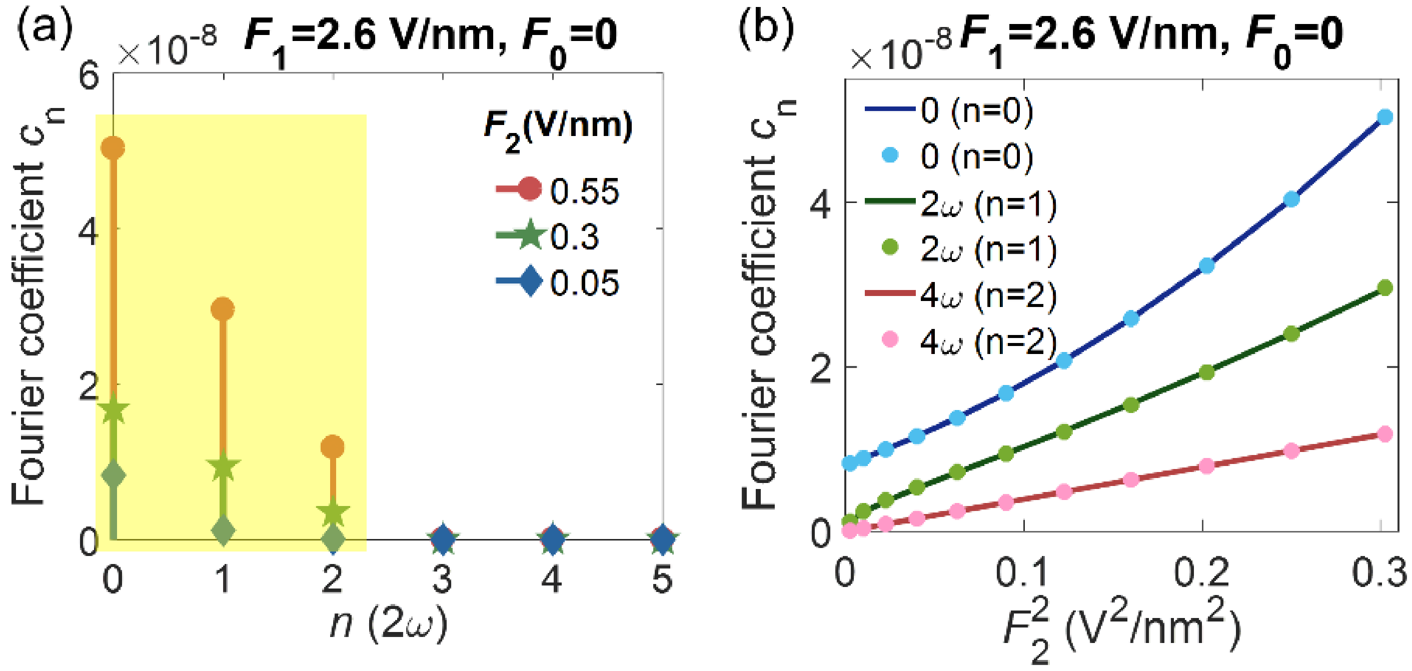


Y. Zhou, and P. Zhang, Phys. Rev. B. 106, 085402 (2022).

Two-color laser coherent control of photoemission

$$D(\tau) = \frac{c_0}{2} + \sum_{n=1}^{N} c_n \sin(n(2\omega)\tau + \varphi_n)$$

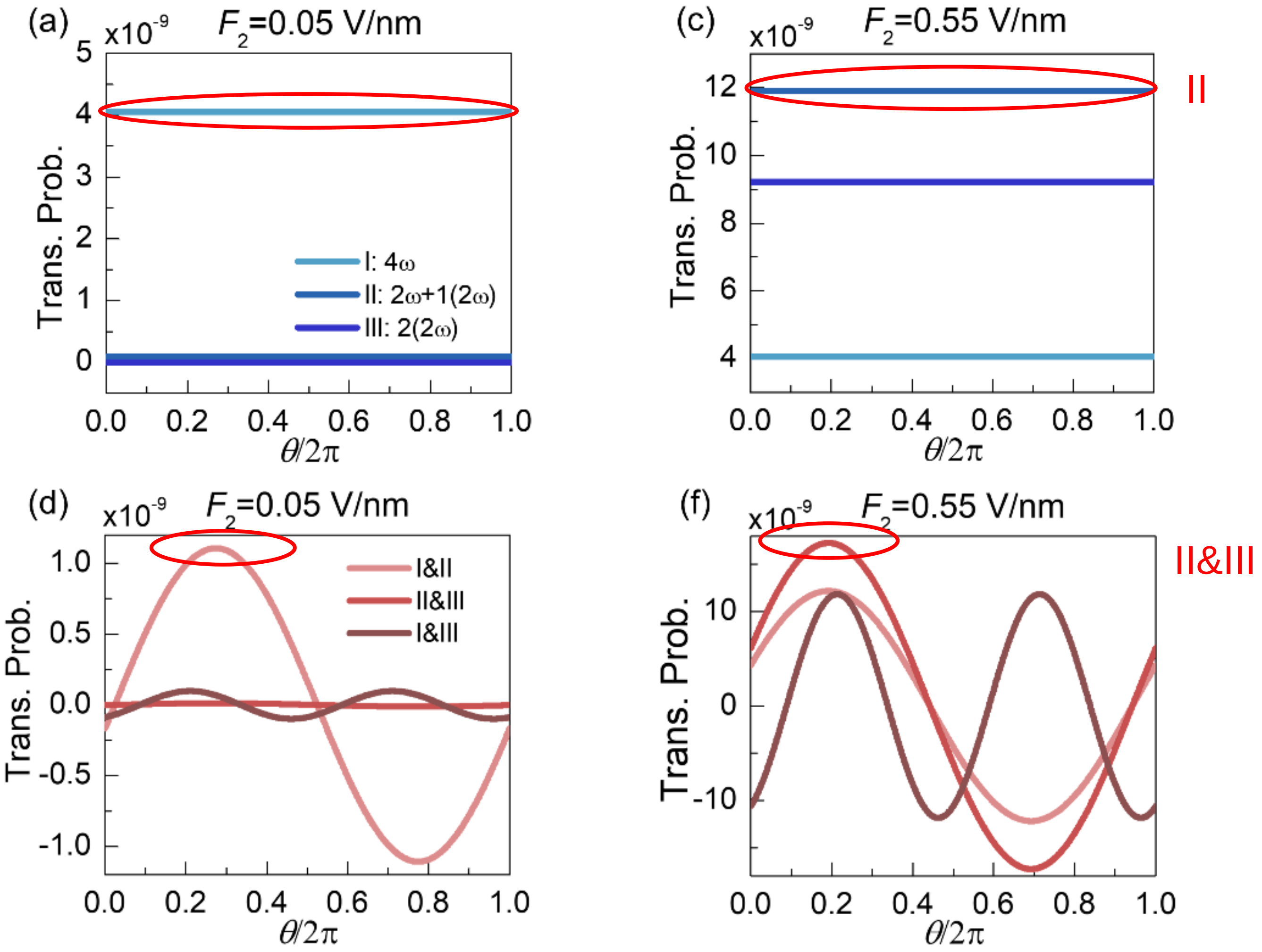
- with $c_0 = \frac{2}{T}\int_0^T D(\tau)d\tau$, $c_n = \sqrt{a_n^2 + b_n^2}$, $a_n = \frac{2}{T}\int_0^T D(\tau)\cos\big(n(2\omega\tau)\big)\,d\tau$, $b_n = \frac{2}{T}\int_0^T D(\tau)\sin(n(2\omega)\tau)\,d\tau$, $T = \frac{2\pi}{2\omega}$, and $\varphi_n = \tan^{-1}\left(\frac{a_n}{b_n}\right)$.

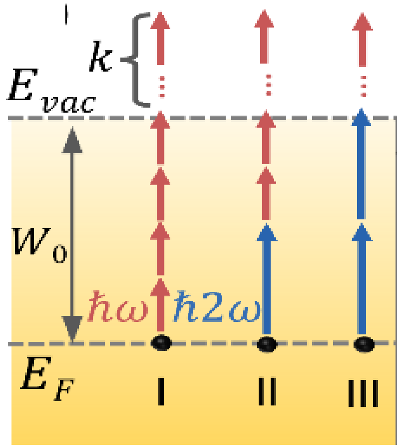


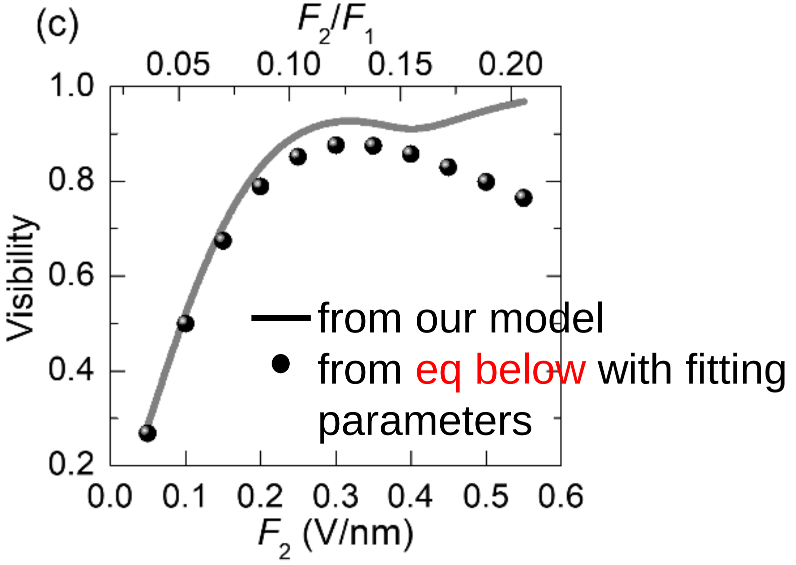
Y. Zhou, and P. Zhang, Phys. Rev. B. 106, 085402 (2022).

Two-color laser coherent control of photoemission

$F_1 = 2.6\ \mathrm{V/nm},\ F_0 = 0$



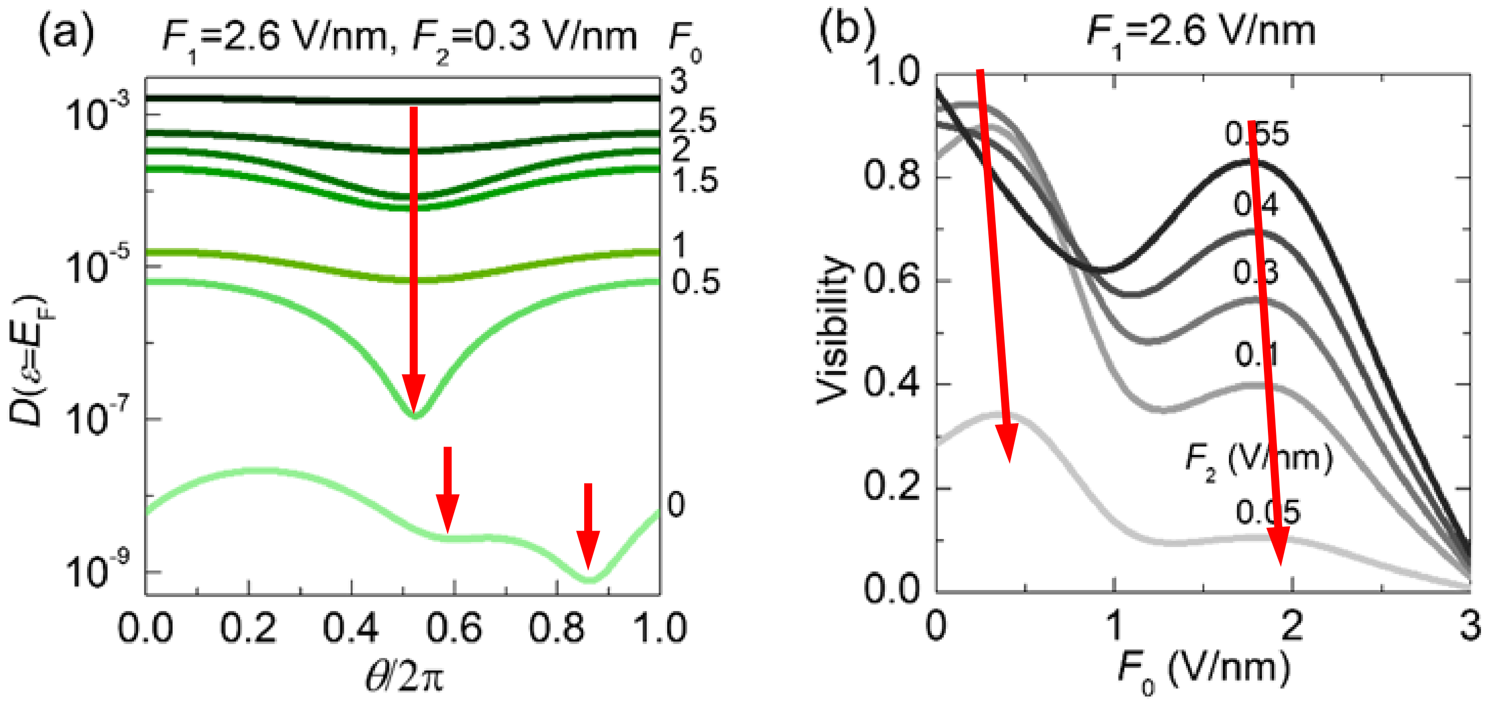




Dots: $\dfrac{D_{I\&II}}{D_I + D_{II}}$

Y. Zhou, and P. Zhang, Phys. Rev. B. 106, 085402 (2022).

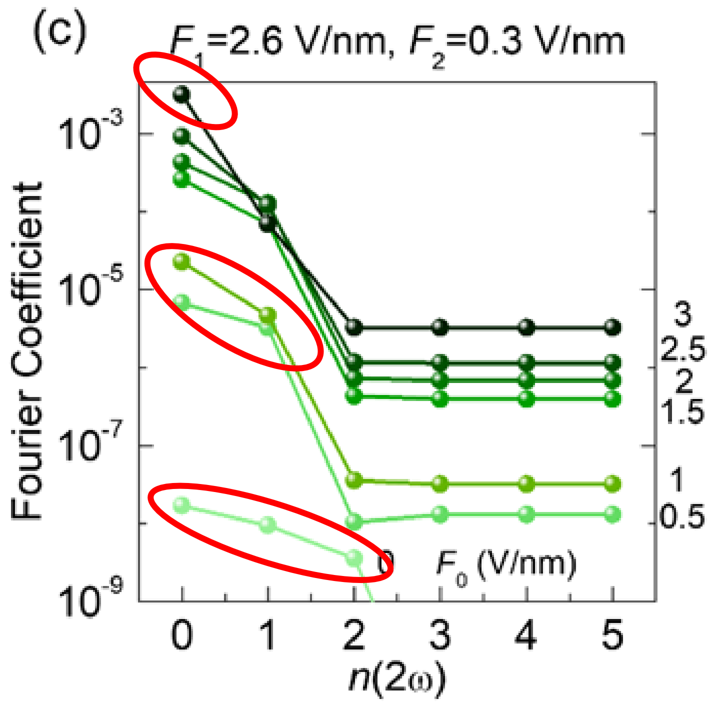
Two-color laser coherent control of photoemission

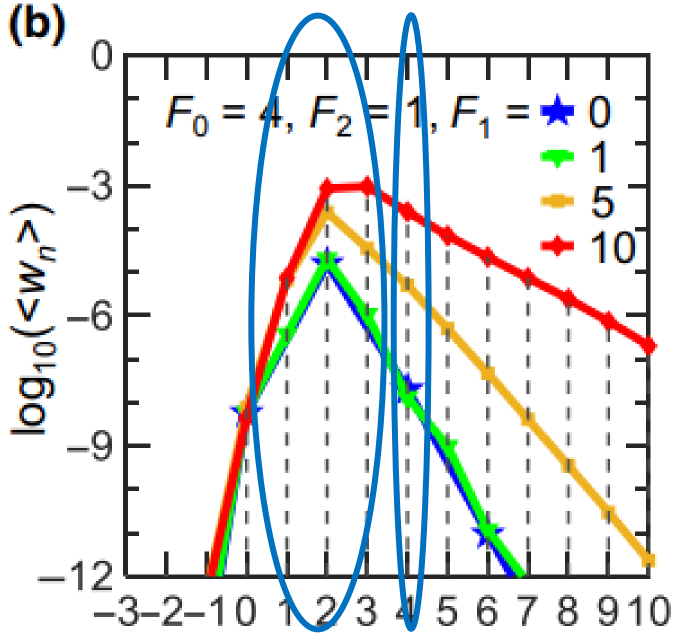


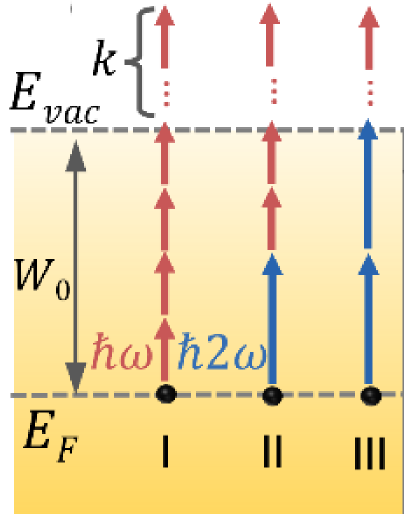
- DC field enhances electron emission
- Two peaks on visibility vs DC field

Y. Zhou, and P. Zhang, Phys. Rev. B. 106, 085402 (2022).

Two-color laser coherent control of photoemission



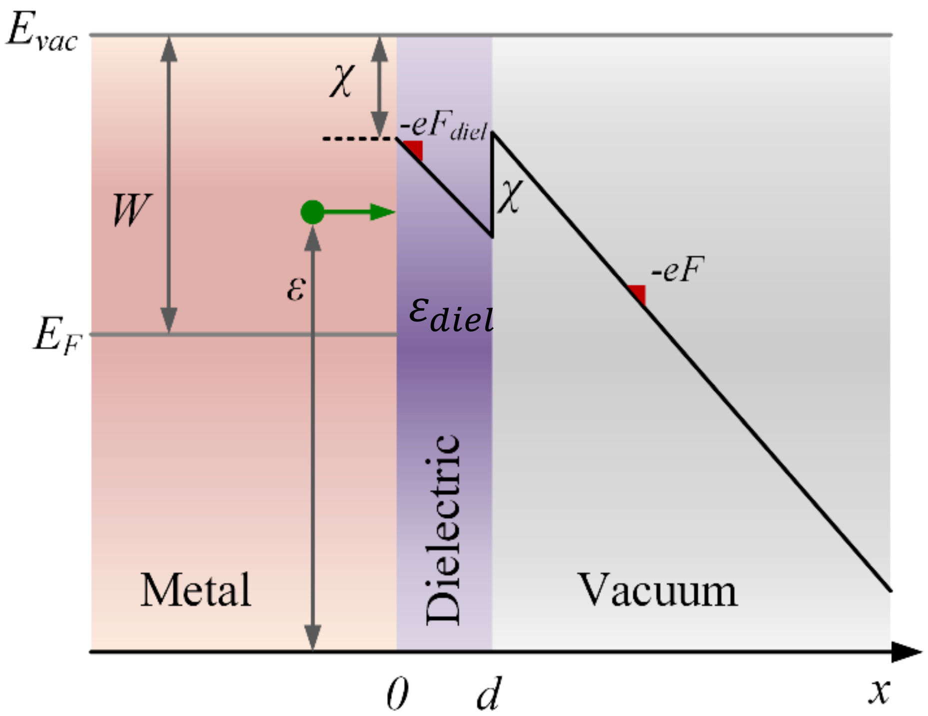




- Interference is sequentially suppressed as DC field increases

[1] Y. Zhou, and P. Zhang, Phys. Rev. B. 106, 085402 (2022). [2] Y. Luo, and Peng Zhang, Phys. Rev. Applied 12, 044056 (2019).

Effects of cathode surface coating



Potential profile

$$V(x) = \begin{cases} 0, & x < 0, \\ E_F + W - \chi - eF_{diel}x, & 0 \le x < d, \\ E_F + W + e(F - F_{diel})d - eFx, & x \ge d. \end{cases}$$

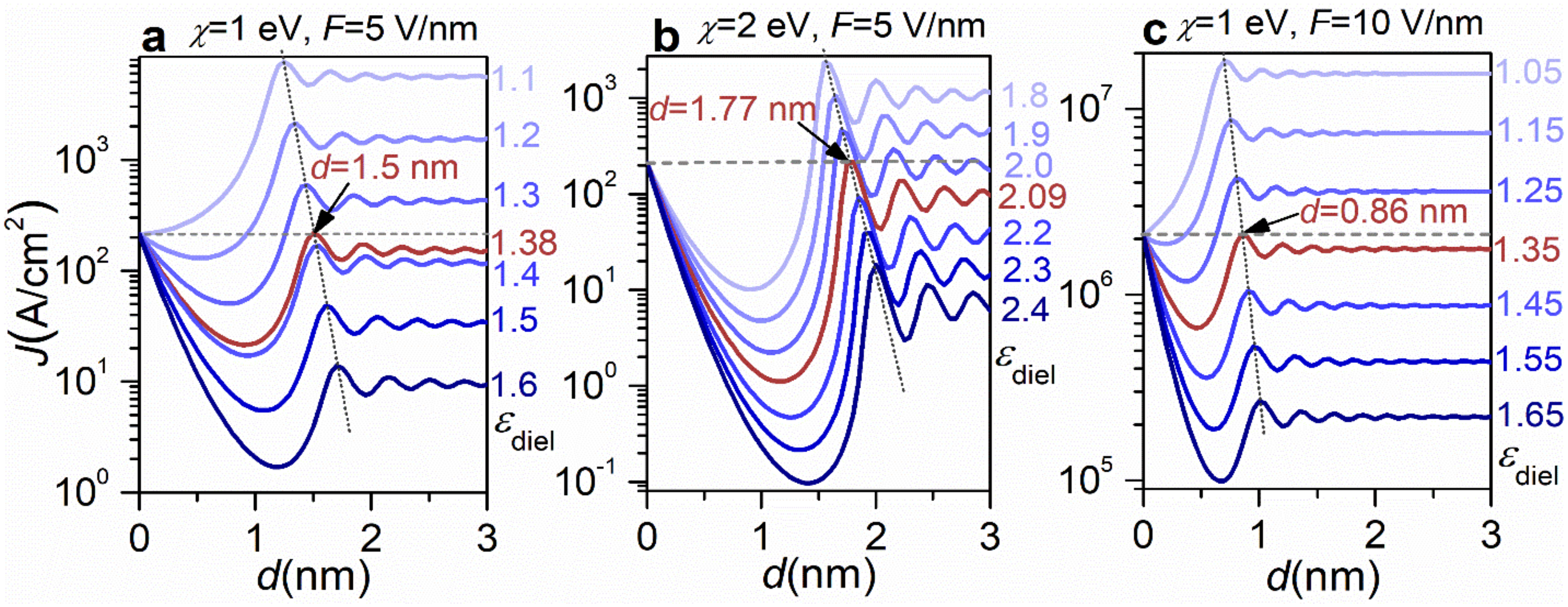
Schrödinger equation

$$-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial x^2}\psi(x) + [V(x) - \varepsilon]\psi(x) = 0$$

Complex wave

Y. Zhou and P. Zhang, Phys. Rev. Res., **2**, 043439 (2020)

Effects of cathode surface coating



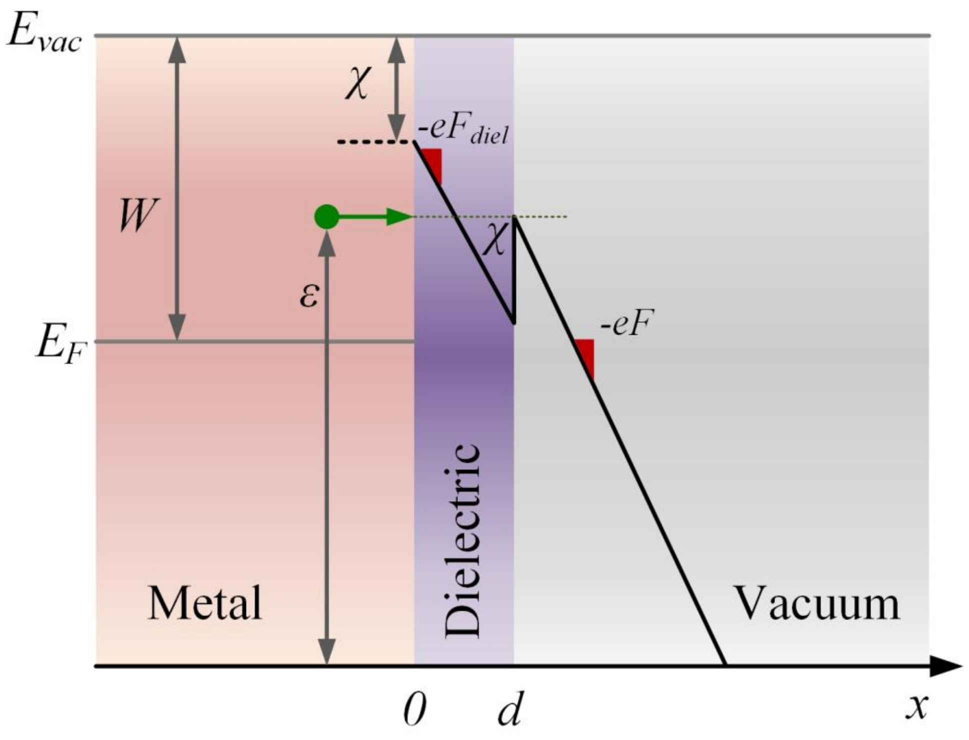
enhanced field emission

Y. Zhou and P. Zhang, Phys. Rev. Res., **2**, 043439 (2020)

Effects of cathode surface coating

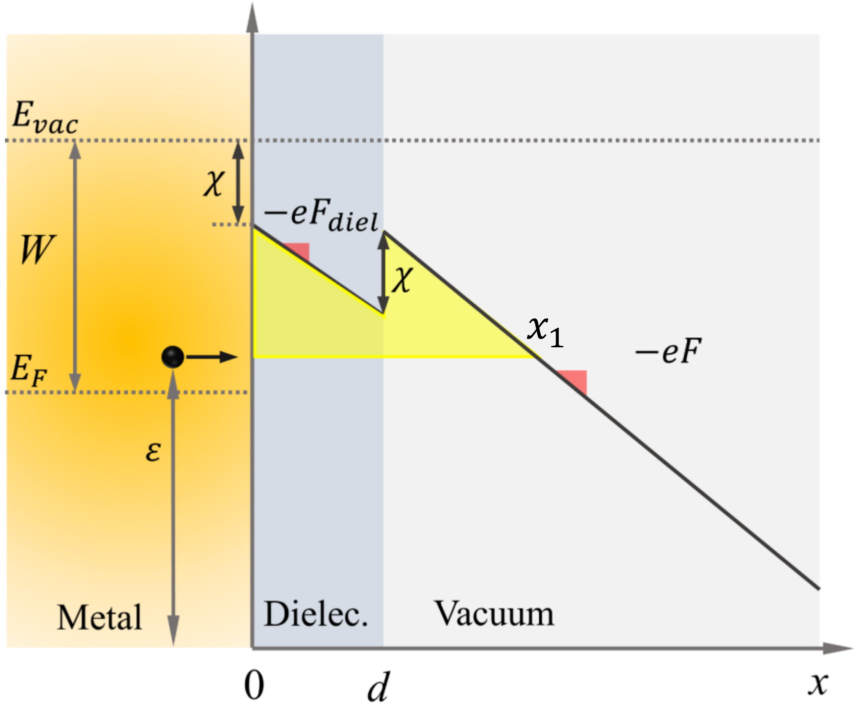
- An empirical relation between **thickness threshold** and **dielectric constant threshold** at room temperature

$$d_{th}[\mathrm{nm}] = \frac{\varepsilon_{diel}^{th} W}{eF}$$



Y. Zhou and P. Zhang, Phys. Rev. Res., **2**, 043439 (2020)

Effects of cathode surface coating



- Electron transmission probability

$$D(\varepsilon) = \exp[Q(\varepsilon)]$$

with $Q(\varepsilon) = -2\int_0^{x_1}\sqrt{\frac{2m}{\hbar^2}[V(x)-\varepsilon]}dx$ by WKBJ approximation

- Electron emission current density

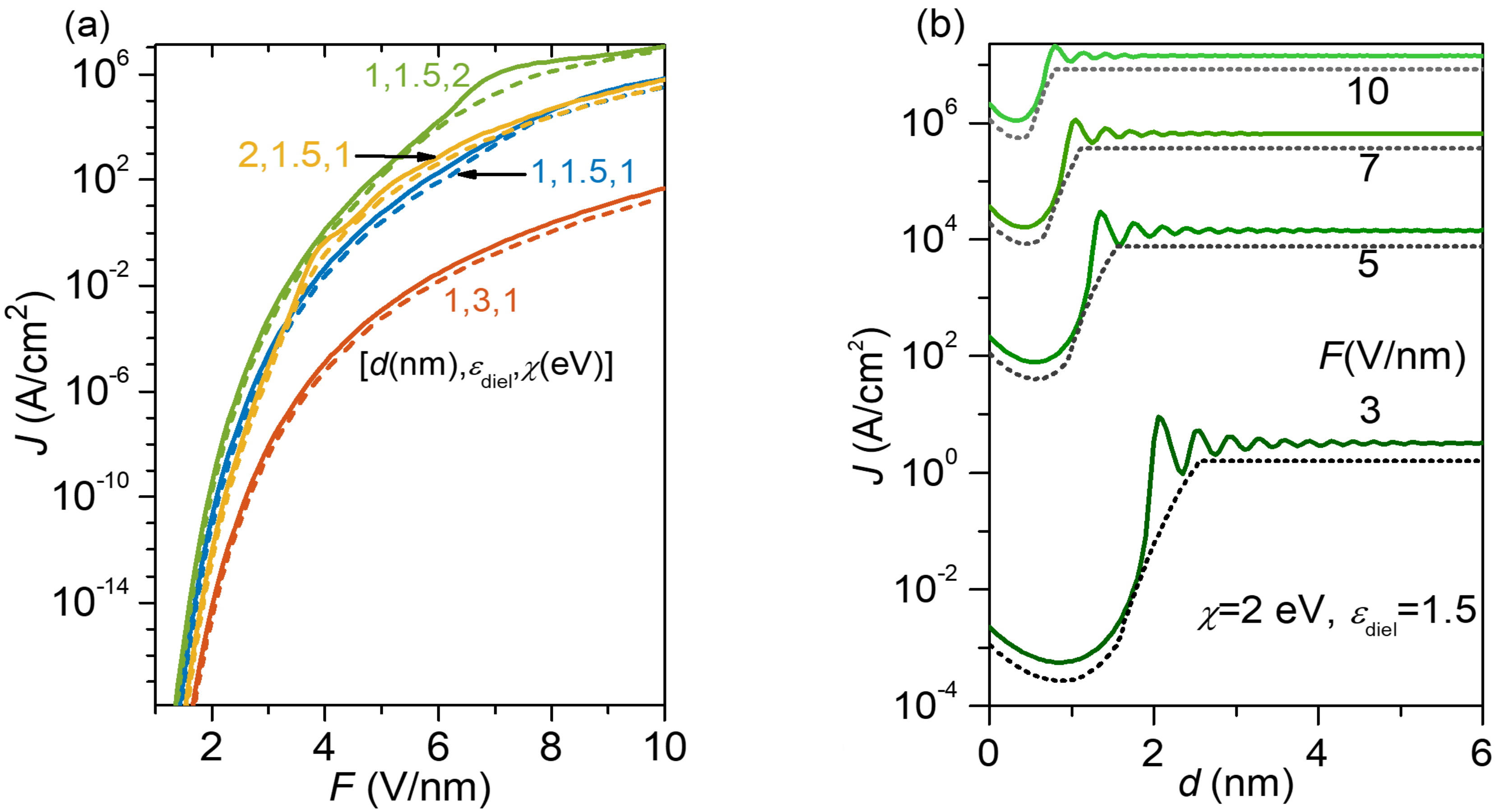
$$J = e\int_0^{\infty} D(\varepsilon)N(\varepsilon)\,d\varepsilon$$

with $N(\varepsilon)$ the supply function

$$N(\varepsilon) = \frac{mk_BT}{2\pi^2\hbar^3}\ln\left[1+\exp\left(\frac{E_F-\varepsilon}{k_BT}\right)\right]$$

[1] Q.-A. Huang, J. Appl. Phys. 79(7), 3703-3707, (1996). [2] P. D. Keathley et al., Ann. Phys., 525(1–2), 144–150 (2013).
[3] K. L. Jensen, et al., J. Appl. Phys. 127(23), 235301, (2020).

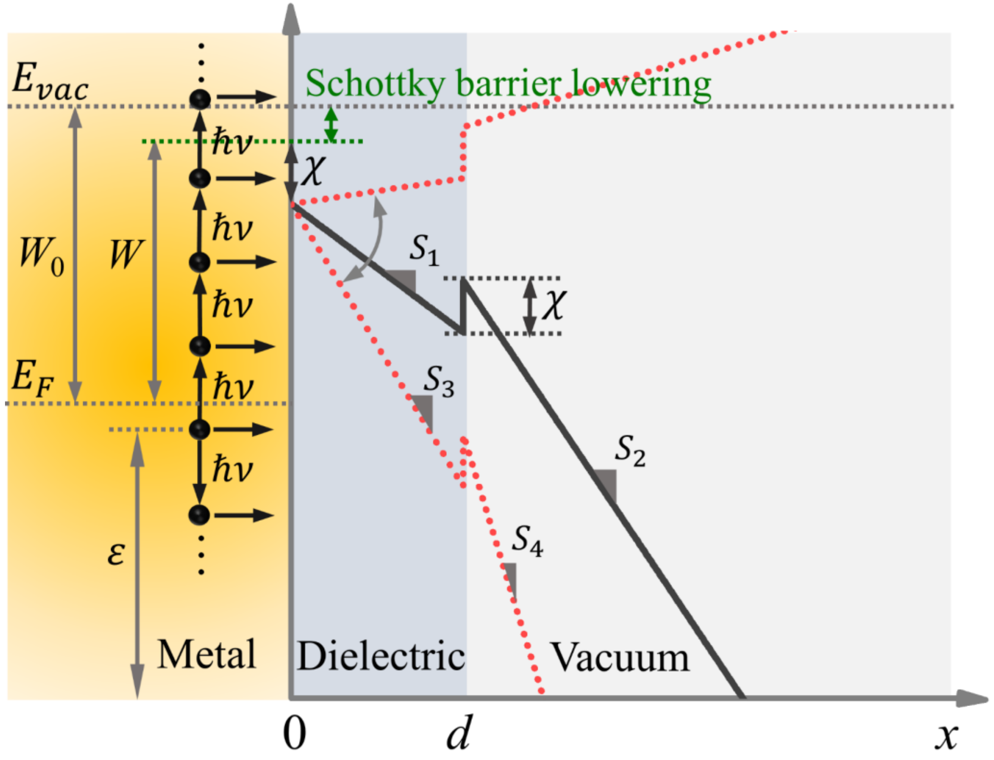
Effects of cathode surface coating



- Good **agreement** in the scaling
- Resonance behavior in J vs. d cannot be revealed by the modified FN equation

Y. Zhou and P. Zhang, Phys. Rev. Res., **2**, 043439 (2020)

Effects of cathode surface coating



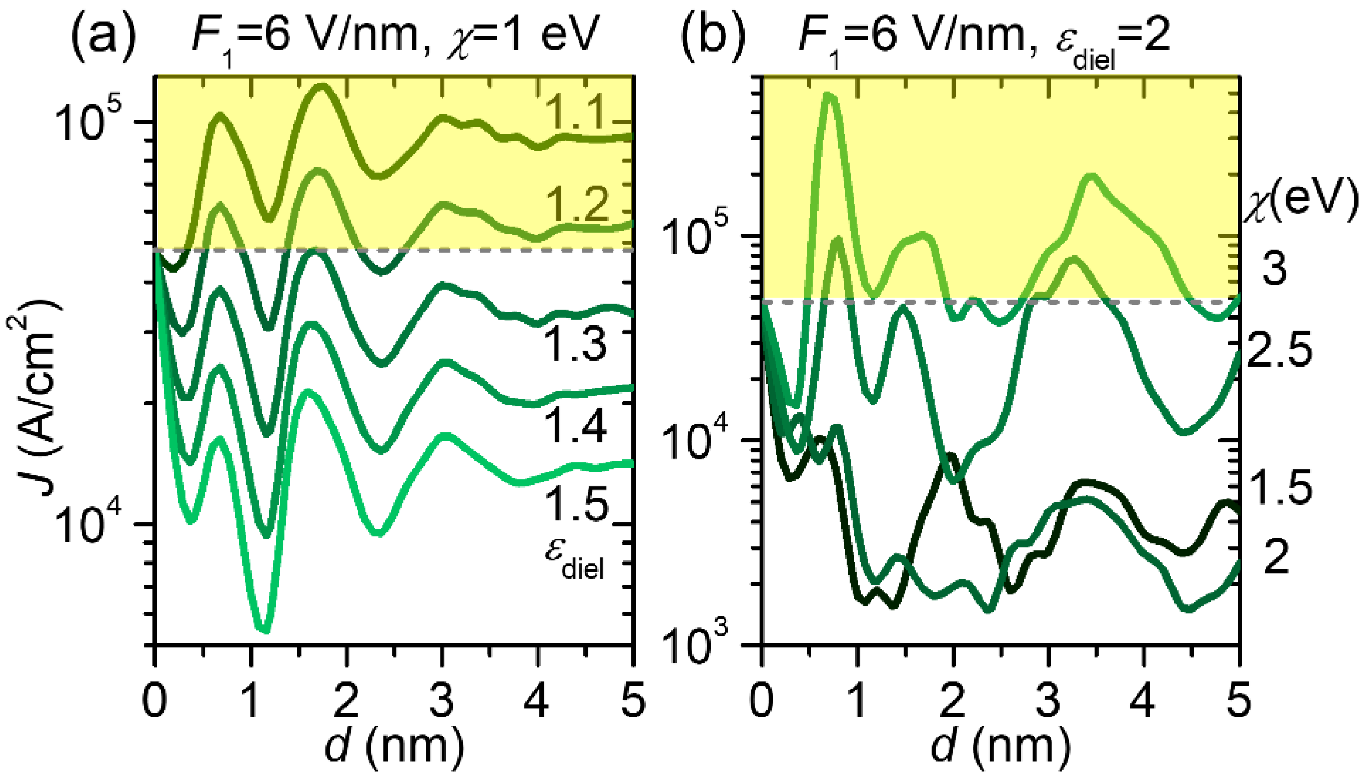
- Potential profile

$$\phi(x,t) = \begin{cases} 0, & x < 0, \\ E_F + W - \chi - eF_0^{diel}x - eF_1^{diel}x\cos\omega t, & 0 \le x < d, \\ E_F + W + e\left(F_0 - F_0^{diel}\right)d + e\left(F_1 - F_1^{diel}\right)d\cos\omega t - eF_0x - eF_1x\cos\omega t, & x \ge d. \end{cases}$$

$W = W_0 - \Delta W$, ΔW is the Schottky barrier lowering. $F_1^{diel} = F_1/\varepsilon_{diel}$, $F_0^{diel} = F_0/\varepsilon_{diel}$ for a perfectly flat surface

Y. Zhou and P. Zhang, J. Appl. Phys. 131, 064903 (2022).

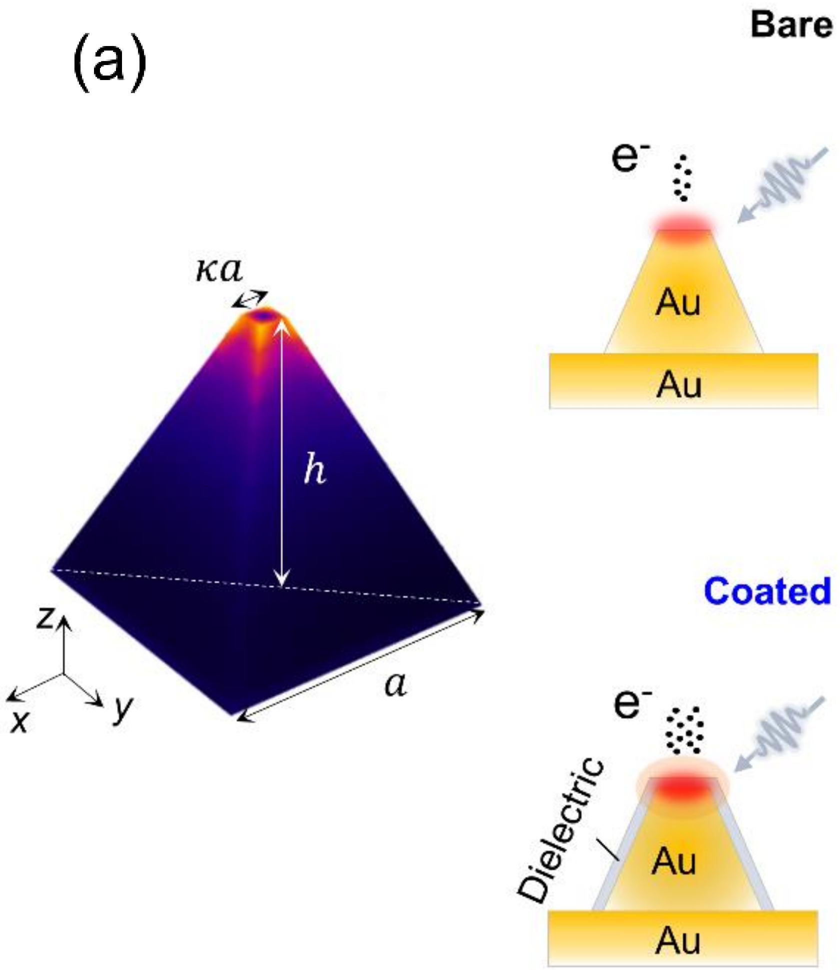
Effects of cathode surface coating



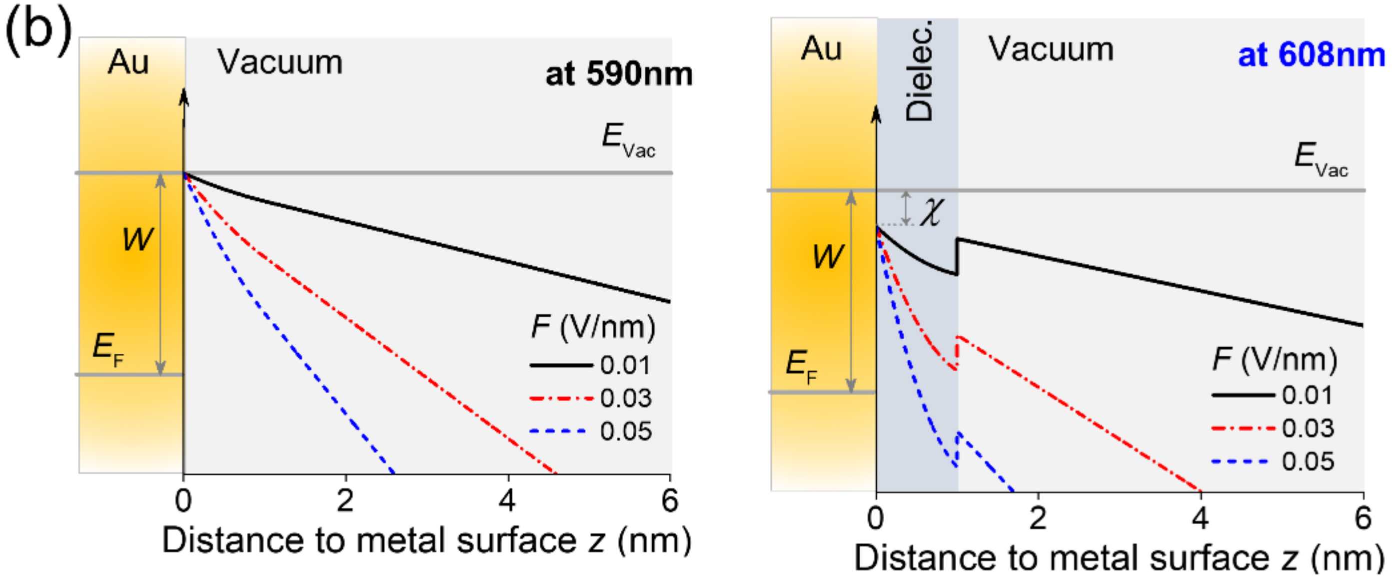
enhanced photoemission

Y. Zhou and P. Zhang, J. Appl. Phys. 131, 064903 (2022).

Plasmon resonance enhancement



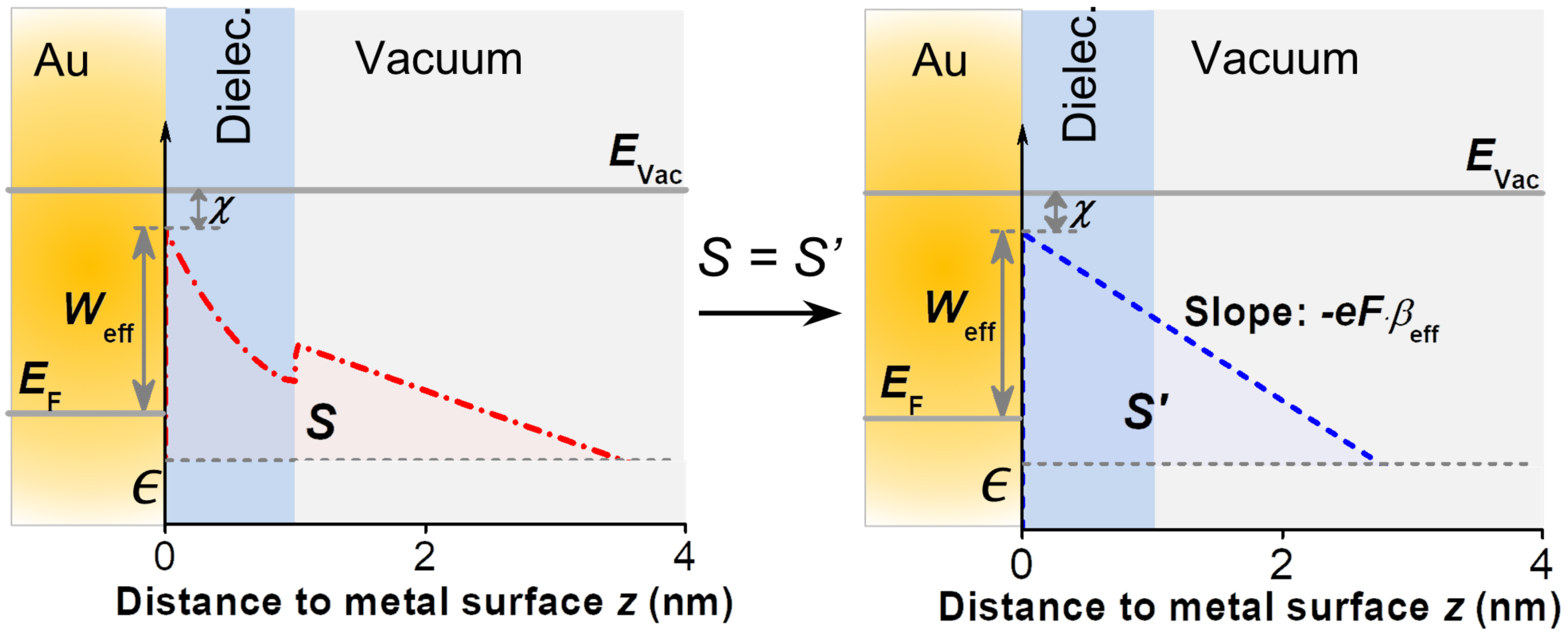
Au-nanopyramid emitter



potential profile

X. Xiong, Y. Zhou, Y. Luo, X. Li, M. Bosman, L. K. Ang, P. Zhang, and L. Wu, ACS Nano, 14, 8806 − 8815 (2020).

Plasmon resonance enhancement



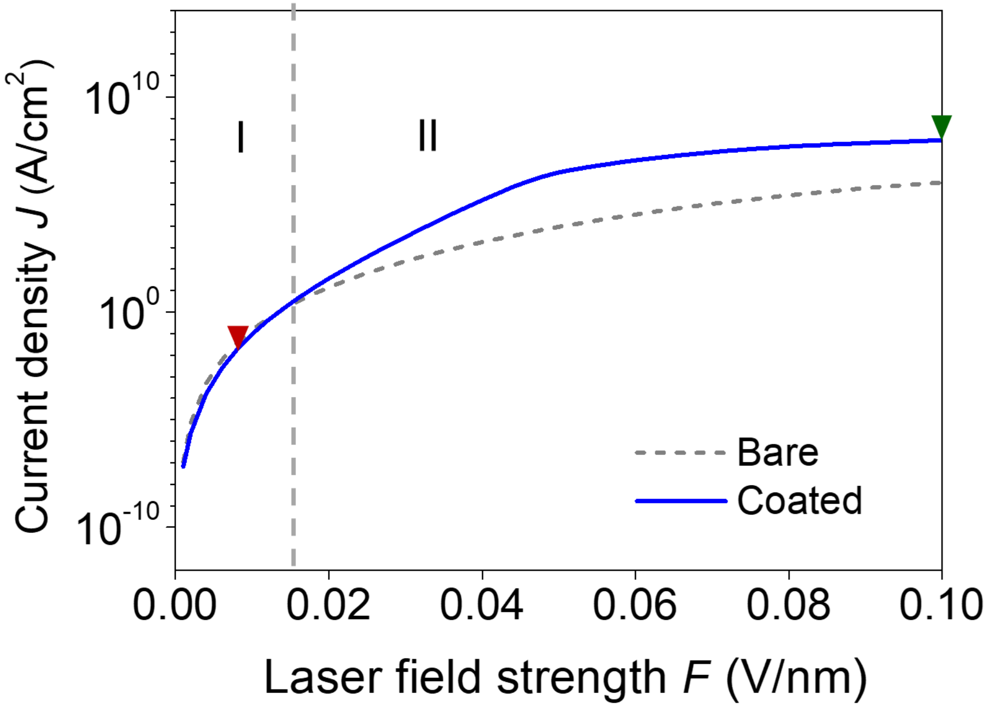
Effective barrier profile approximation

$$\phi(z,t) = \begin{cases} 0, & z < 0 \\ E_F + W_{\text{eff}} - e\beta_{\text{eff}} F z \cos \omega t\,, & z > 0 \end{cases},$$

with $W_{\text{eff}} = W - \chi$

X. Xiong, Y. Zhou, Y. Luo, X. Li, M. Bosman, L. K. Ang, P. Zhang, and L. Wu, ACS Nano, 14, 8806 − 8815 (2020).

Plasmon resonance enhancement

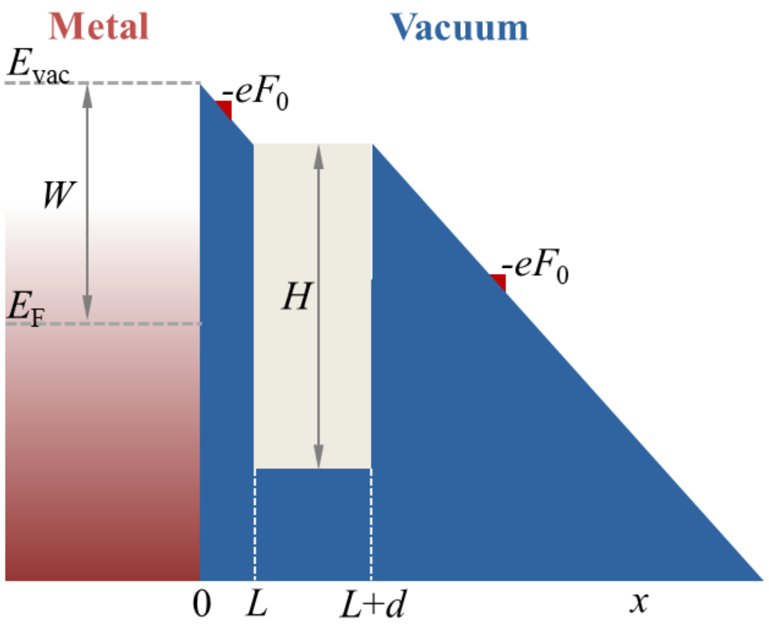


~2 orders of magnitude enhancement

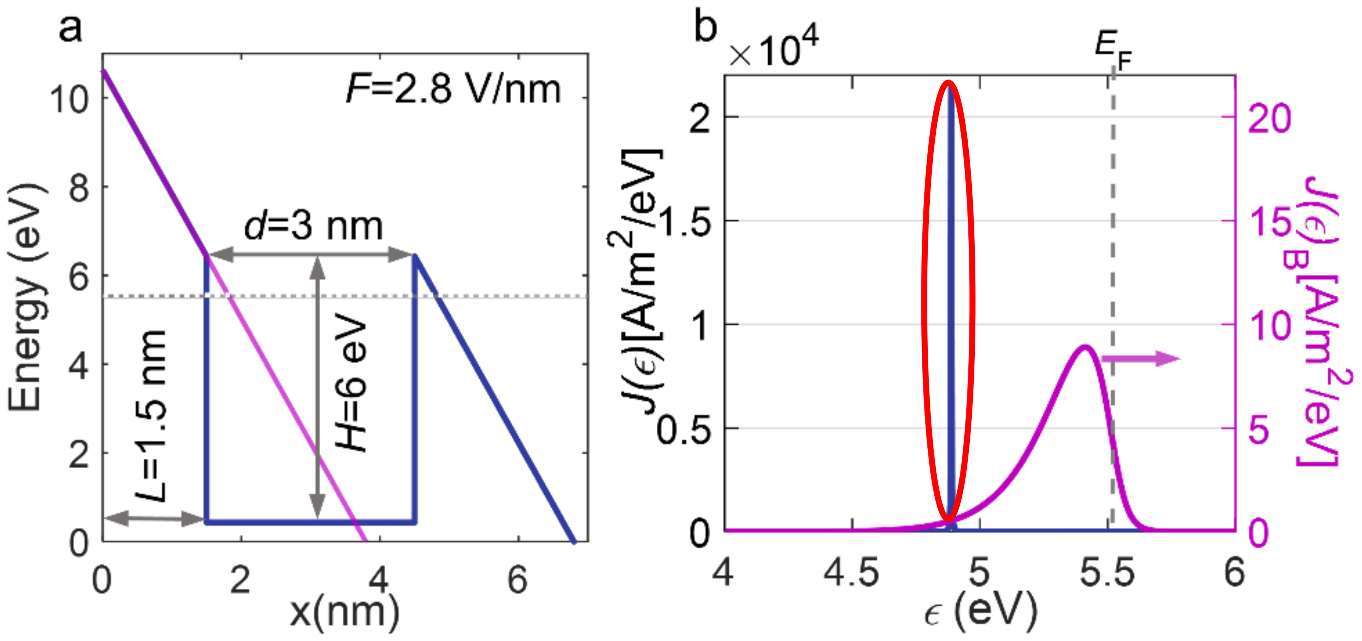
X. Xiong, Y. Zhou, Y. Luo, X. Li, M. Bosman, L. K. Ang, P. Zhang, and L. Wu, ACS Nano, 14, 8806 − 8815 (2020).

Metal Surfaces with a Quantum Well

- Nanostructures or adsorbates (ions, atoms, or molecules etc.) on cathodes



Resonant Tunneling Enhanced Field Emission



3.07 A/m^2

105 A/m^2

- Potential to produce highly intense and highly collimated electron beams

Summary

- Photoemission
  - Laser effects
    - Effects of laser wavelength and **accompanying heating**
    - **Two-color laser** coherent control of photoemission
  - Status of metallic photocathode
    - Dielectric coatings effects
    - Quantum well effects

- Photo-/field/thermionic emission
- Integer of $W_0/\hbar\omega$
- Contribution from each pathway

- Enhanced field/photo- emission
- Plasmon-enhanced photoemission
- High-intensity and highly collimated electron beam

Q&A

Thank you!