Quantum Pathways Interference in Photoemission from Biased Metal Surfaces Induced by Two-color Lasers

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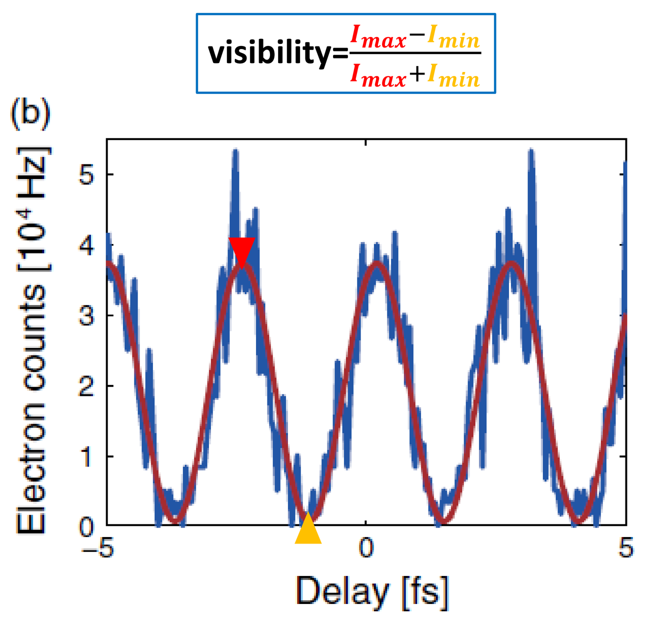
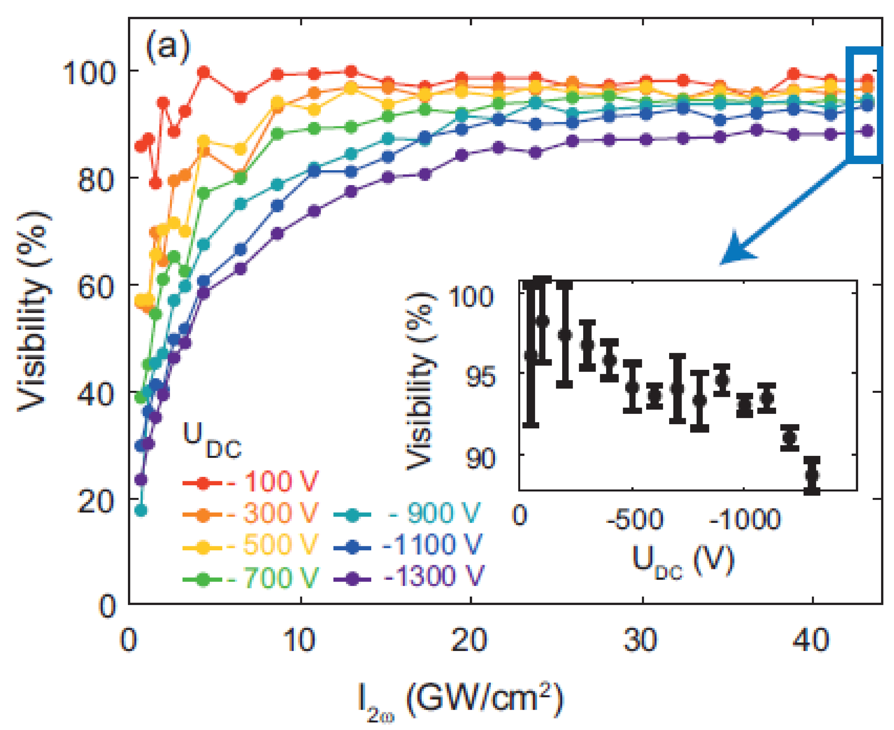
I. Motivation

- Two-color coherent control of photoemission from nanotips has drawn great interest. It has flexibility in manipulating electron dynamics in **ultrashort temporal scale** and **nanometer spatial scale**.
- By tuning the **intensity mixture ratio** and **relative phase difference** between the fundamental laser and its second harmonic, the photoemission current can be modulated with a contrast (called **visibility** or **modulation depth**) of up to 97.5%.
- **DC bias** is another key tuning knob.



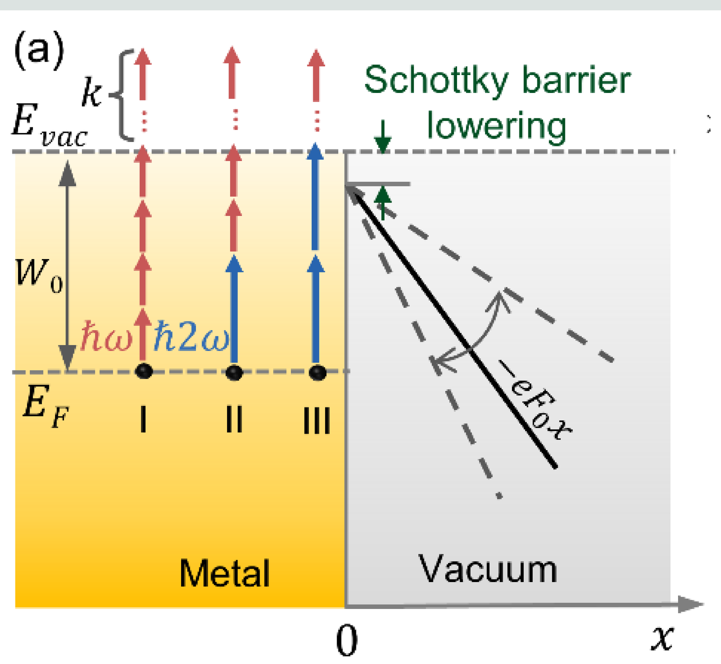
Förster, Michael, et al. *Phys. Rev. Lett.* 117.21 (2016): 217601.



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

- The underlying physics that how laser fields and dc bias field influence two-color coherent control of photoemission is still unclear.

II. Models



W_0: nominal work function
E_F: Fermi energy
E_{vac}: Vacuum level
F_0: DC field
ω: Fundamental angular frequency
$W_{Schottky} = 2\sqrt{e^3F_0/16\pi\varepsilon_0}$: Schottky barrier lowering

- **Potential profile**

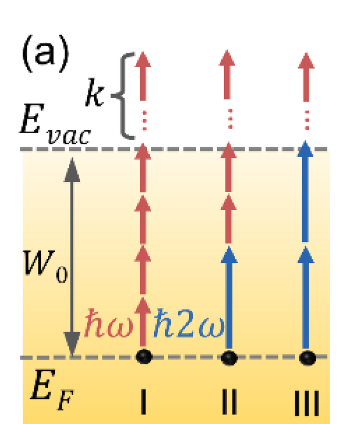
$$\phi(x,t) = \begin{cases} 0, & x<0 \\ V_0 - ef(t)x - eF_0x, & x \ge 0 \end{cases}$$

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

- **Electron transmission probability**

$$D(\varepsilon) = \sum_{l=-\infty}^{\infty} w_l(\varepsilon)$$

- **Quantum pathway interference model**



$D_I \propto \alpha^k (F_1^2)^k (\alpha^4 (F_1^2)^4) = K_I$;

$D_{II} \propto \alpha^k (F_1^2)^k (\zeta^2 (F_1^2)^2 F_2^2) = K_{II} F_2^2$;

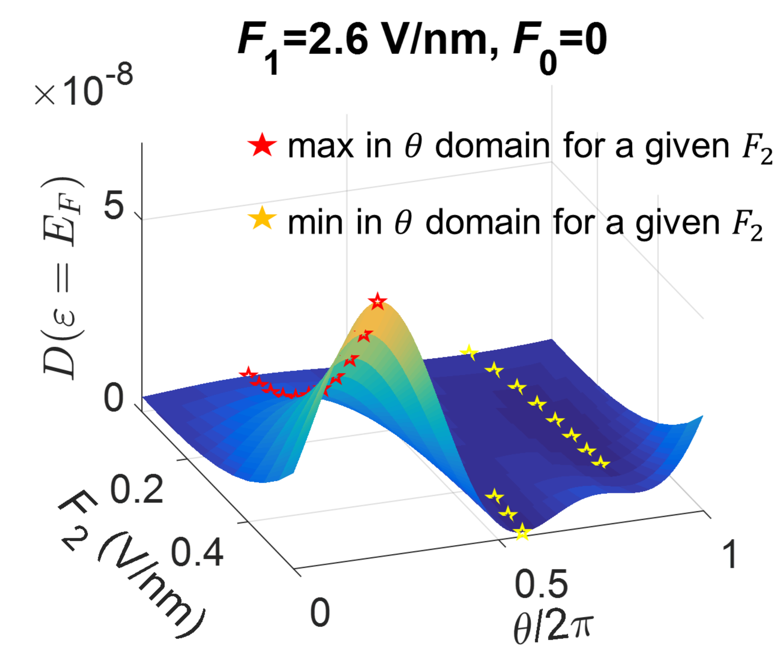
$D_{III} \propto \alpha^k (F_1^2)^k (\beta^2 (F_2^2)^2) = K_{III} (F_2^2)^2$;

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

$D_{I\&III} \propto 2\sqrt{D_I D_{III}}\cos 2\theta \propto \alpha^k (F_1^2)^k (2\alpha^2 (F_1^2)^2 \beta F_2^2 \cos 2\theta) = K_{I\&III} F_2^2 \cos(4\omega\tau)$;

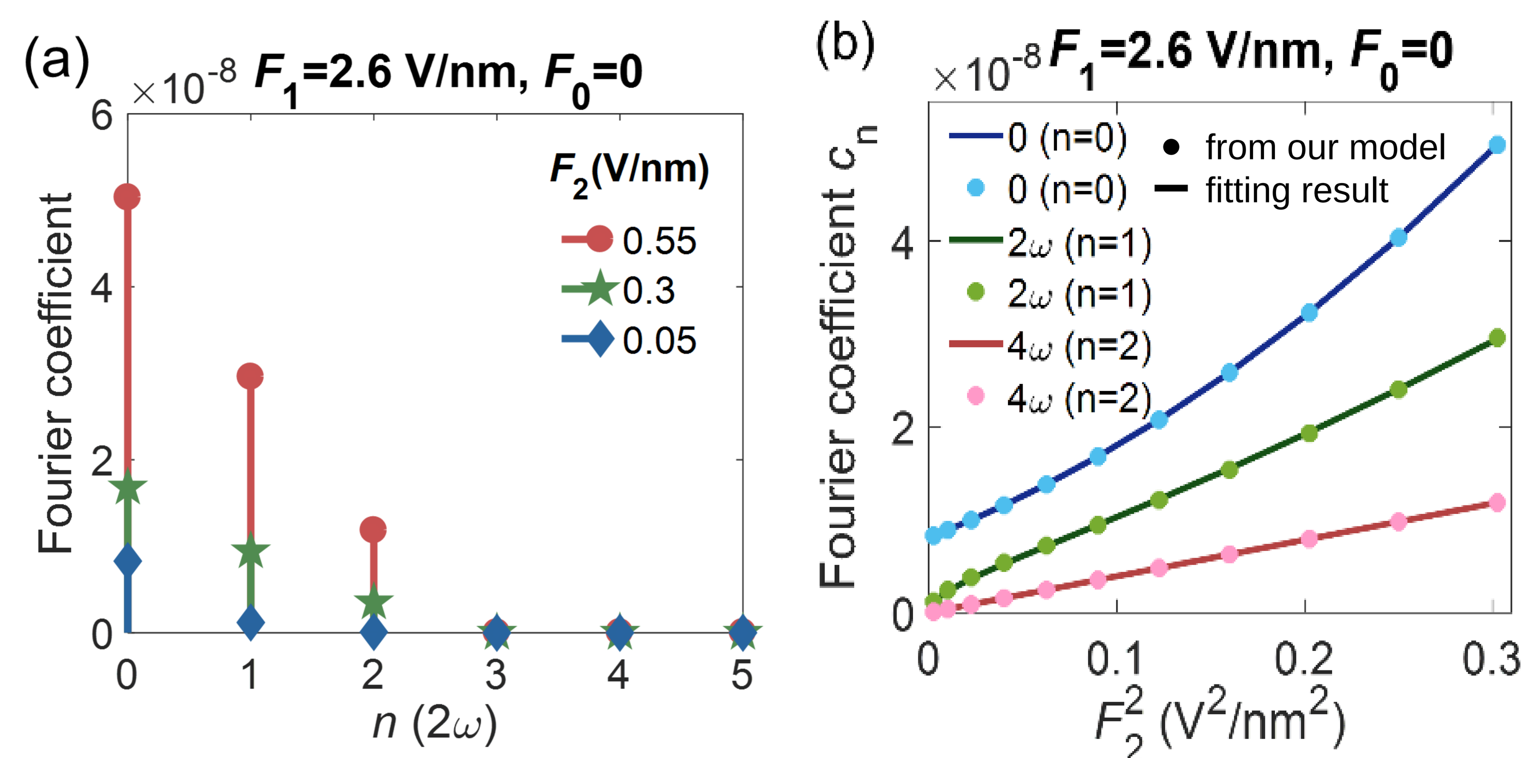
$D_{II\&III} \propto 2\sqrt{D_{II} D_{III}}\cos\theta \propto \alpha^k (F_1^2)^k \left(2\zeta F_1^2 \beta \sqrt{(F_2^2)^3}\cos\theta\right) = K_{II\&III}\&\sqrt{(F_2^2)^3}\cos(2\omega\tau)$.

III. Fourier series expansion

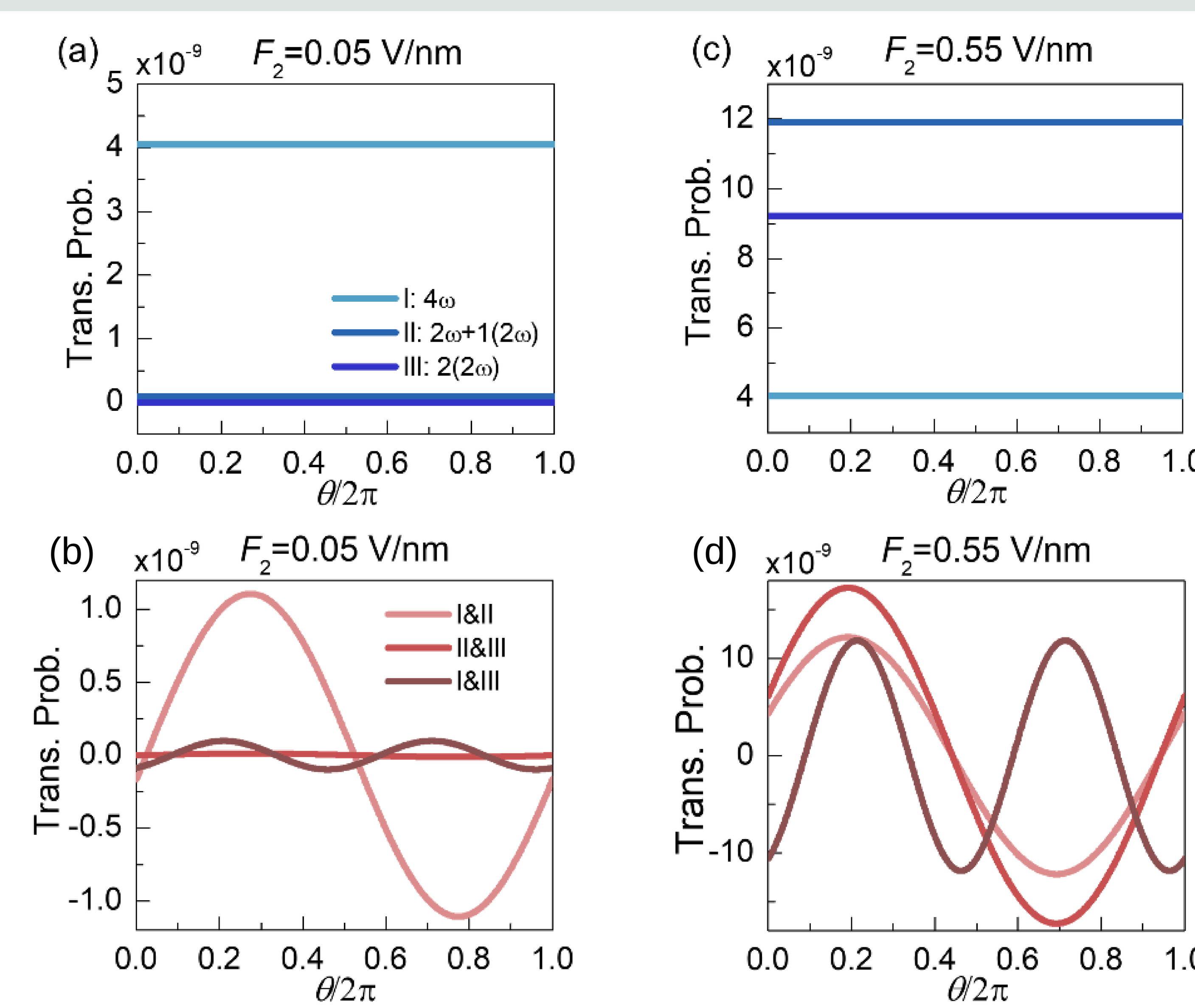


$$D(\tau) = \frac{c_0}{2} + \sum_{n=1}^{N} c_n \sin(n(2\omega)\tau + \varphi_n)$$

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

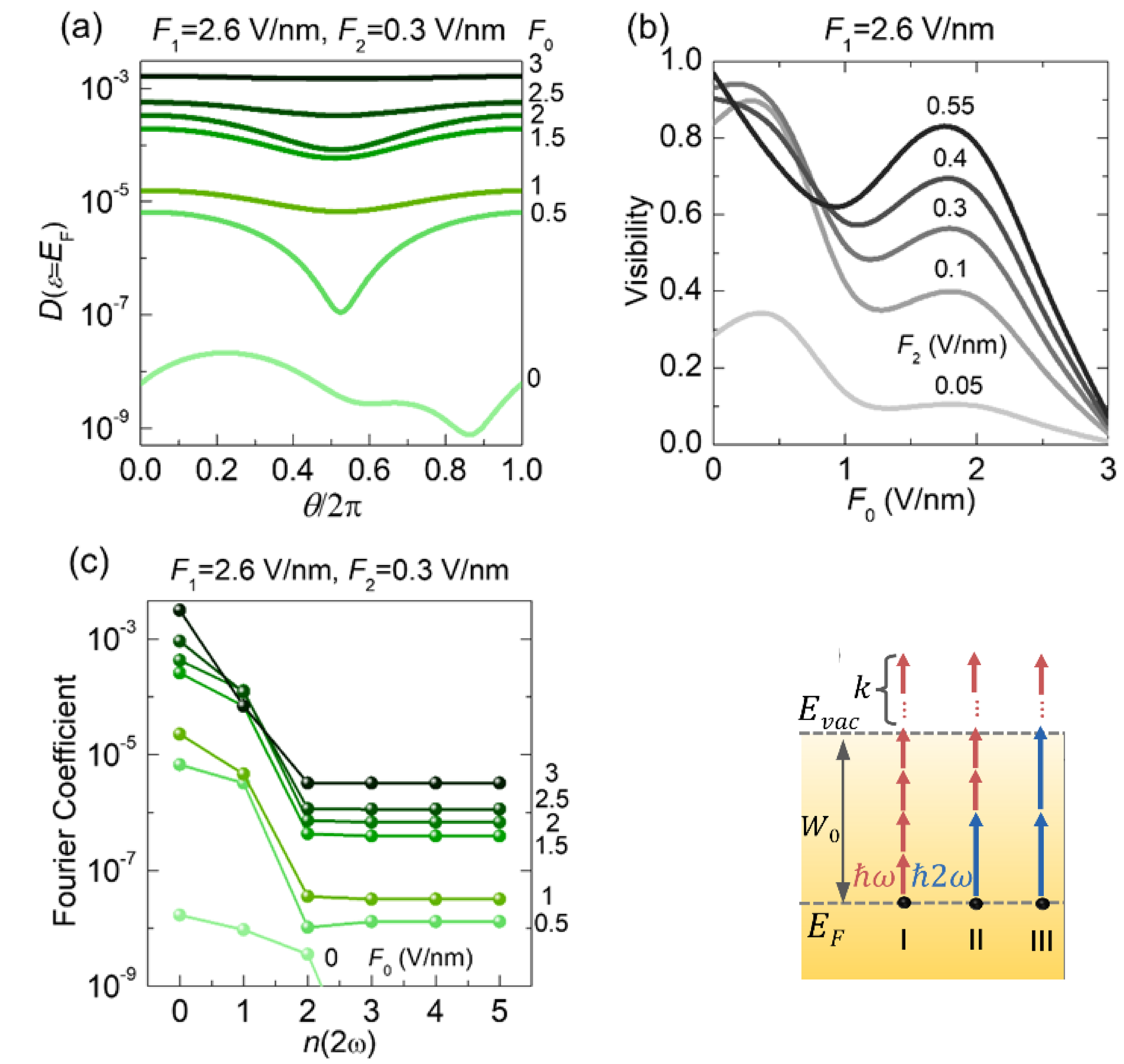


IV. Effects of laser fields



When F_2/F_1 increases, emissions through **pathway II** and **interference II&III** become the dominant terms.

V. Effects of dc field



- Dc field enhances electron emission
- There are two peaks on visibility
- Interference is sequentially suppressed as dc field increases

VI. Conclusion

- We analyzed the **quantum pathways interference** in two-color coherent control of photoemission using exact analytical solutions of the TDSE including dc bias.
- Increasing the intensity ratio of the second harmonic to fundamental lasers would result in more contribution from **multicolor pathway** and **multiphoton absorption of ℏ(2ω).**
- Increasing bias voltages sequentially decreases the weights of **higher order 4ω and then 2ω components**, resulting in two peaks in the visibility as a function of bias voltage.

References

[1] Y. Luo and P. Zhang, Phys. Rev. B 98, 165442 (2018).
[2] Y. Luo and P. Zhang, Phys. Rev. Applied 12, 044056 (2019).
[3] Y. Zhou and P. Zhang, Phys. Rev. B 106, 085402 (2022).

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