Influence of Temporal Shapes of Femtosecond Laser Pulses on Photoemission from a Metal Surface

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I. Background

Background:

1. Femtosecond laser pulse in various shapes can be generated by techniques, such as mode-locking, chirped pulse amplification (CPA) and birefringent crystals for pulse stacking.
2. Photoemission from metallic nanoparticles induced by femtosecond laser field has many potential applications, including time-resolved photoemission electron microscopy, high frequency electromagnetic radiation generation, and high-speed nanoscale vacuum devices [1].
3. Appropriate temporal shaping of the optical pulses illuminating the photocathode can minimize uncorrelated emittance growth from photoemission electron guns due to space charge forces [2].

We investigate photoemission from an Au surface subjected to a weak DC field and a femtosecond laser pulse in various shapes using an exact quantum model [3-4]. Besides laser intensity, the interaction between solid-state nanostructures and femtosecond laser pulses is impacted by temporal shapes, pulse lengths (τ_p) and phase since they can modify the laser pulse spectrum [5].

II. Theoretical Formulation

➢ **Femtosecond laser**

$$F(t) = F_1 E(t)\cos(\omega t + \phi), \quad (1)$$

where F_1 is the peak laser strength. ω is the angular frequency of the laser carrier and ϕ is the carrier envelope phase (CEP). E(t) stands for the carrier envelops, as shown in Fig. 1, which includes Gaussian (m = 1), super-Gaussian (of different orders m = 3, 5, and 10) of Eq. 2a, hyperbolic secant squared (sech²) of Eq. 2b, and cos² profiles of Eq. 2c,

$$E(t) = \exp[-\left(\frac{t}{\sigma}\right)^{2m}], \quad (2a)$$

$$E(t) = \mathrm{sech}^2(\frac{t}{\sigma}), \quad (2b)$$

$$E(t) = cos^2\left(\frac{\pi t}{2\tau_p}\right)[\mathcal{H}(t+\tau_p) - \mathcal{H}(t-\tau_p)], \quad (2c)$$

we define $-L \le t \le L$, where $T_{pulse} = 2L$ is the time period of the femtosecond laser pulse in a pulse train.

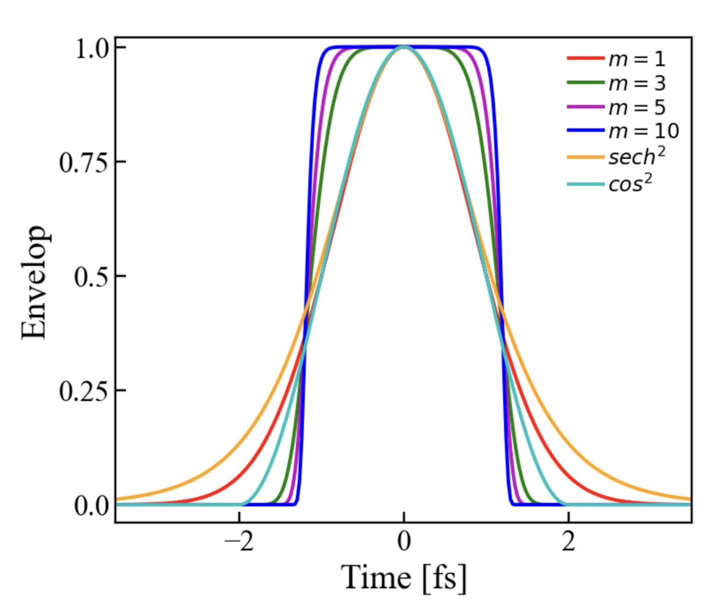


Fig.1 Carrier envelop of E(t)

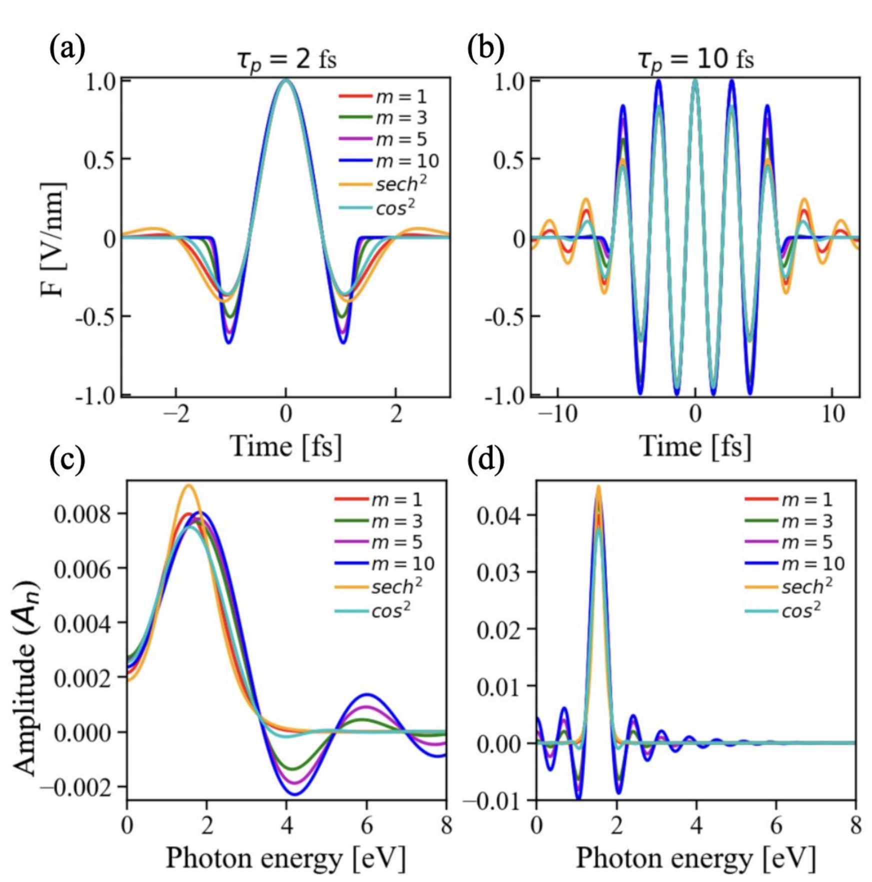


Fig.2 Femtosecond laser field and corresponding frequency spectra

A_n is the Fourier series coefficient (cosine form) of the periodic function derived from Eq. (1).

➢ **One-dimensional time-dependent Schrödinger equation near the metal-vacuum interface is [4],**

$$i\hbar\frac{\partial\psi(x,t)}{\partial t} = -\frac{\hbar^2}{2m}\frac{\partial^2\psi(x,t)}{\partial t^2} + \Phi(x,t)\psi(x,t), \quad (3)$$

$$\text{where } \Phi(x,t) = \begin{cases} 0, & x < 0 \\ E_F + W_{eff} - eF_0x - eF(t)x, & x \ge 0 \end{cases}$$

➢ **Emission current density at electron initial energy ε can be obtained:**

$$w(\varepsilon, x, t) = \frac{J_t}{J_i}, \quad (4)$$

where $J = \frac{i\hbar}{2m}(\psi\nabla\psi^* - \psi^*\nabla\psi)$, $\boldsymbol{J_t}$ is probable transmitted current density and $\boldsymbol{J_i}$ is probable incident current density.

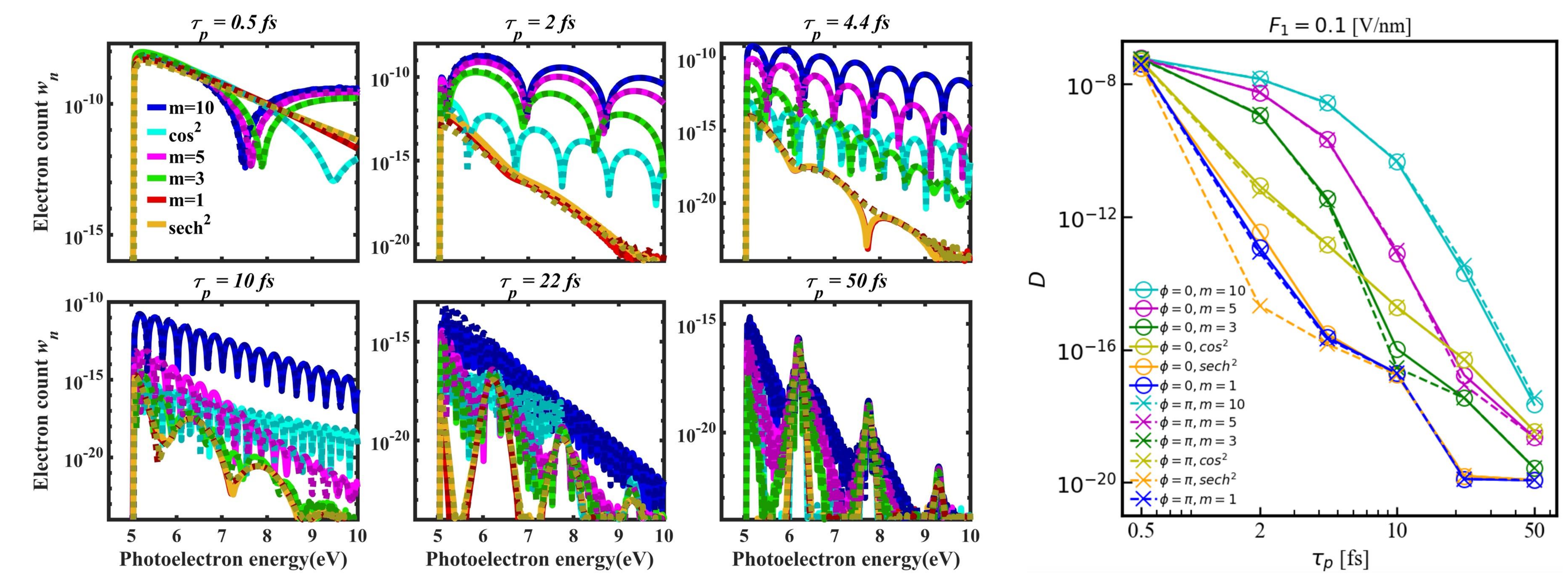
➢ **The total emission current density across one pulse from an initial energy level ε,**

$$D(\varepsilon) = \frac{2L}{\tau_p}\sum_{n=-\infty}^{\infty} w_n(\varepsilon), \quad (5)$$

where $w_n(\varepsilon)$ denotes the time-averaged current density (i.e. electron transmission probability) through the n-photon process.

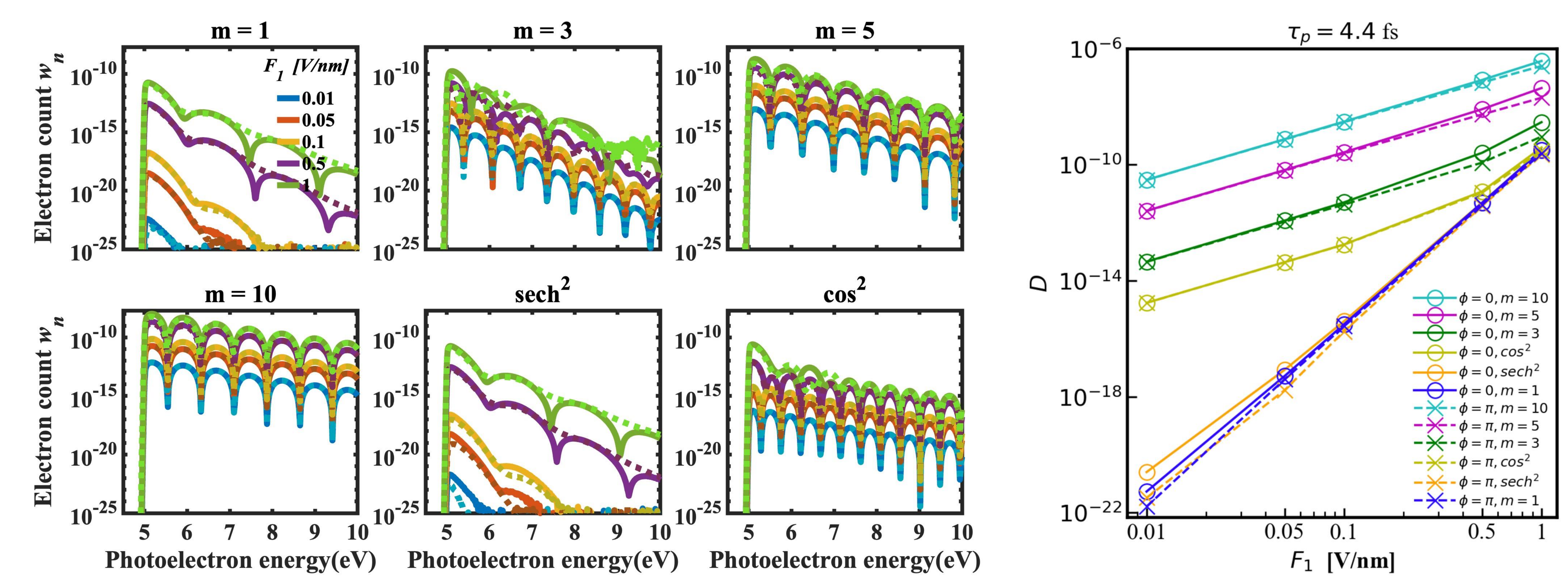
III. Results

➢ **Emission current density through nth channel for various pulse duration τ_p**



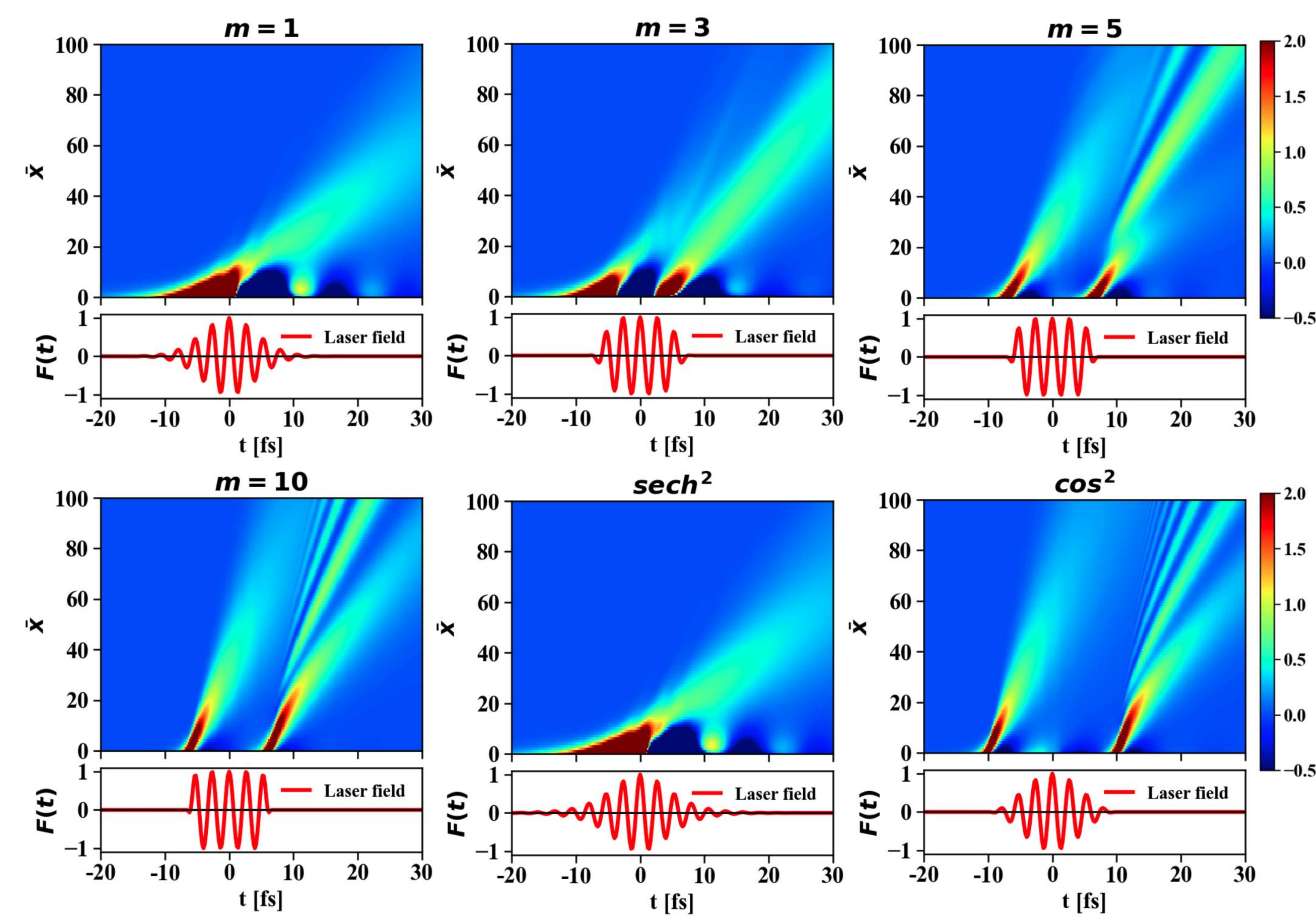
- DC field $F_0 = 0.001$ V/nm

➢ **Emission current density through nth channel for various peak laser strength F_1**



- DC field $F_0 = 0.005$ V/nm
- Right figure: $aF_1{}^b$ fitting from $F_1 = 0.01$ to 0.5 V/nm (not shown)
  - $\phi = 0$: b=5.99 (m=1), 2.44 (m=3), 2.12 (m=5), 2.05 (m=10), 2.61 (cos²), 5.85 (sech²)
  - $\phi = \pi$: b=6.00 (m=1), 2.07 (m=3), 1.92 (m=5), 1.96 (m=10), 2.55 (cos²), 6.18 (sech²)

➢ **Time/space-dependent emission current density $w(\varepsilon, x, t)$/<w>**



- Pulse length $\tau_p = 10$ fs.
- Red line is the corresponding laser field for reference.

IV. Conclusion

1. The emission spectrum shape correlates with the laser frequency spectrum shape: shorter pulse lengths (broader spectra) produce broader emission spectra, while longer pulse lengths (narrower spectra) result in narrower emission spectra.
2. For very short pulse lengths with broad spectra that approaches the effective work function, differences in pulse shapes have little impact on the total emission current density.
3. For longer pulse lengths with narrower spectra, much smaller than the effective work function, the influence of pulse shape on electron emission decreases as pulse length increases.
4. With a fixed pulse length, the emission current induced by narrower spectra laser rises more rapidly than by broader spectra laser as the intensity increases.
5. The effect of the CEP is influenced by both pulse length and laser intensity. The laser intensity required to observe CEP effects on emission current is lower for narrower spectra laser than for wider spectra laser.
6. The spatiotemporal emission current density is different for various femtosecond pulse profiles.

References

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Acknowledgements

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