

Enhanced quantum efficiency in photoemission driven by few-cycle optical fields

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We theoretically show that the quantum efficiency (QE) of photoemission can be dramatically enhanced by reducing the laser pulse duration to a few optical cycles. Using an exact quantum theory for ultrafast photoemission, we find that QE can increase by several orders of magnitude as the pulse length decreases from tens of cycles to a few cycles, at fixed low laser intensities. The corresponding photon order of the multiphoton process is also significantly reduced, indicating a shift in the underlying emission mechanism. We find that the carrier-envelope phase strongly modulates the photoelectron spectrum and QE in the few-cycle regime; however, counterintuitively, its influence diminishes for subcycle pulses. The results open up opportunities for enhancing and controlling ultrafast electron emission through optical pulse shaping in the low-intensity regime, with potential applications in attosecond science, ultrafast electron sources, and light-wave-driven quantum devices. The macroscopic enhancement of photoemission efficiency driven by few-cycle optical fields may be considered as yet another example of macroscopic quantum phenomena.

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I. INTRODUCTION

Photoemission driven by ultrafast optical fields from solids enables the generation and control of spatiotemporally coherent electron bunches within femtosecond and nanometer scales, which has attracted significant interest due to its potential applications in a variety of areas, including time-resolved electron microscopy and streaking spectroscopy [1–5], advanced particle accelerators and light sources [6], high-power electromagnetic sources [7,8], strong-field nano-optics [9], and high-speed light-field-driven electronics [10,11].

Experiments and, more importantly, practical realization of photoemission-enabled applications all hinge on the photoemitter's quantum efficiency (QE), a key parameter that measures the yield of photoelectrons per incident photon. While commonly used in the characterization of photocathodes, particularly for particle accelerators [12–15], QE is barely studied under ultrashort pulsed condition. A key question arises as to how QE varies with laser parameters, especially when the optical pulse length shrinks to few-cycle duration, which is essential for effectively advancing photoemitters driven by few-cycle optical pulses for various advanced ultrafast applications.

Extensive studies have been conducted to understand the underlying emission processes of photoelectrons driven by pulsed lasers, including the transition from multiphoton absorption at low laser intensities to optical field tunneling at

high laser intensities [16–19], the sensitivity of the total emitted current to the carrier-envelope phase (CEP) [20–23], and precise probing and control of electron dynamics [1–5,24,25]. While the nonlinear dependence of QE of photoemission on the laser wavelength and intensity was previously characterized [26,27] and confirmed with experiments [27,28], its dependence on the laser pulse length is not well understood. Previous studies indicate that pulse length plays a significant role in controlling the emission current and electron energy distribution, as well as the possible shift of photon order in the multiphoton regime [16,29,30]; however, the influence of pulse length on the photoemission mechanism and its corresponding QE, particularly in the few-cycle duration, has not been quantitatively analyzed in detail. Although qualitative trends can be inferred from strong-field ionization models such as the Yudin-Ivanov rate [31], a quantitative assessment of the QE scaling of photoemission from solids remains largely unexplored. Furthermore, the majority of the existing works on photoemission from solids induced by few-cycle lasers almost exclusively focus on optical field emission using high-intensity lasers [10,19,23,32,33], as the efficiency of the multiphoton process at low laser intensities is believed to be exceedingly smaller.

In this work, we report the significant increase of QE as the pulse length reduces to a duration of a few optical cycles at low laser intensities. The results imply that photoemission induced by few-cycle optical fields may be practically detectable in the multiphoton regime at very low laser intensities, thus offering insights into the development of practical ultrafast electronics enabled by few-cycle optical fields. The physics behind this phenomenon lies in the broadening of the photon energy (optical frequency) as the laser pulse width shortens, where photons with higher energy facilitate the emission of photoelectrons from the solid surface. Using a quantum photoemission model by exactly solving

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the one-dimensional (1D) time-dependent Schrödinger equation (TDSE) [30], we found a substantial increase in the photoemission current density accompanied by a broadening of the emitted photoelectron spectrum as the laser pulse width reduces to the few-cycle regime. Consequently, an increase of the corresponding QE by several orders of magnitude is obtained as the laser pulse length reduces from ~ 10 cycles to subcycle duration. The photon order in the multiphoton emission process is also reduced. Furthermore, we found that the influence of CEP becomes less pronounced when the laser pulse is reduced to subcycle duration, in contrast to optical field emission at high laser intensities.

II. MODEL

We consider photoelectron emission from the interface of metal-vacuum subjected to a dc electric field F_0 and a Gaussian profile femtosecond optical field

$$F(t) = F_1 e^{-\frac{t^2}{\sigma^2}} \cos(\omega t + \phi), \quad (1)$$

where F_1 is the peak optical field, $\sigma = \frac{\tau_p}{2\sqrt{\ln 2}}$ with τ_p being the full width at half maximum (FWHM) of the field envelope, ω is the angular frequency of the optical carrier, and ϕ is the CEP. It is worthwhile to note that the optical field described in Eq. (1) may represent the local optical field near the emitter surface, which is not necessarily propagating in space [10], and effects such as local field enhancement factor, light-surface diffraction, and plasmonic resonance could all be captured by the effective optical field strength [34]. By solving the 1D TDSE subject to the time-varying potential barrier influenced by both F_0 and $F(t)$, the exact analytical solution for the total emission probability due to a single laser pulse from an initial electron energy level ε is found to be

$$D(\varepsilon) = \sum_{n=-\infty}^{\infty} \langle w_n(\varepsilon) \rangle, \quad (2)$$

where $\langle w_n(\varepsilon) \rangle$ denotes the electron transmission probability through the n -photon process, with $n < 0$ representing multiphoton emission, $n = 0$ direct tunneling, and $n > 0$ multiphoton absorption process. The detailed expression for $\langle w_n(\varepsilon) \rangle$ can be found in Ref. [30]. The total emission current density J during a Gaussian laser pulse can be calculated by considering electron emission from all energy levels in metal:

$$\begin{aligned} J &= e \int_0^{\infty} D(\varepsilon) N(\varepsilon) d\varepsilon \\ &= \int_{-\infty}^{\infty} J_{\text{spectrum}}(\varepsilon_{\text{emitted}}) d\varepsilon_{\text{emitted}}, \end{aligned} \quad (3)$$

where $N(\varepsilon) = \frac{m_e k_B T}{2\pi^2 \hbar^3} \ln[1 + \exp(\frac{E_F - \varepsilon}{k_B T})]$ is the electron supply function with $N(\varepsilon)d\varepsilon$ specifying the electron flux within the metal that normally impinges on the metal surface [26,28,35,36] with initial longitudinal energy between ε and $\varepsilon + d\varepsilon$, assuming Fermi-Dirac distribution of the electrons with m_e being the mass of the electron, k_B the Boltzmann constant, T the temperature, and E_F the Fermi energy. In Eq. (3), $J_{\text{spectrum}}(\varepsilon_{\text{emitted}})$ is the differential current density per energy [$\text{A}/(\text{cm}^2 \text{eV})$], which represents the emitted

photoelectron energy spectrum with $\varepsilon_{\text{emitted}} = \varepsilon + n\hbar\omega$ the emitted electron energy.

The QE is defined as the ratio of the number of emitted electrons to that of the incident photons per pulse,

$$\text{QE} = \frac{J\tau_p/e}{E_l/\hbar\omega}, \quad (4)$$

where $E_l = \frac{\epsilon_0 c F_1^2 \tau_p \sqrt{\pi}}{4\sqrt{2} \ln 2}$ is the laser energy per unit area per pulse. Note here that, for simplicity, we assume the local optical field near the emitter surface is the same as the input laser field, i.e., a unity local field enhancement factor. In this work, we consider gold as the cathode material with a nominal work function $W_0 = 5.1$ eV and Fermi energy $E_F = 5.53$ eV at $T = 300$ K. The laser wavelength is 800 nm, corresponding to a photon energy of 1.55 eV and a laser period of 2.67 fs.

III. RESULTS

The effect of pulse length on the optical field frequency spectra is shown in Fig. 1(a), where the amplitude is calculated from $A_\omega = \int_{-\infty}^{\infty} F(t) \cos(\omega t) dt$, which is the Fourier transform of the optical field pulse in Eq. (1). As the pulse length decreases, the spectrum widens and its peak at the laser photon energy of 1.55 eV decreases. This means that as optical pulse length shrinks to a value comparable to or even smaller than the laser period, the pulse would effectively consist of a distribution of photons with a broad range of photon energies, where photons with higher effective energy would contribute to photoemission and significantly enhance the emission yield. Notably, the spectral amplitude for $\phi = \pi$ is identical to that of $\phi = 0$ but with a negative sign for a given pulse length. In Fig. 1(b), we compare the emitted photoelectron energy spectra from electrons with initial energy at the Fermi level for different pulse lengths under CEP of $\phi = 0$ and $\phi = \pi$, with $F_1 = 0.3$ V/nm and $F_0 = 0.00075$ V/nm. The emission spectra show stronger similarity between $\phi = 0$ and π when the pulse length is either much smaller or much larger than one laser cycle (2.67 fs for an 800 nm laser). For $\tau_p = 1.8$ and 2.54 fs, noticeable shift between the $\phi = 0$ and π curves exists, indicating the strong effect of CEP when τ_p is close to the laser period. Also, when the pulse length decreases, the distinct multiphoton peaks become broadened and eventually smeared to form a smooth spectrum with an overall higher electron yield. This is consistent with the broadened laser spectra at short pulse length as shown in Fig. 1(a). Figure 1(c) shows the effect of optical field pulse length on the total emission probability per pulse, $D(\varepsilon = E_F)$. As the pulse length decreases from 40 fs (15 cycles) to 20 fs (7 cycles), D increases slightly. However, as τ_p further decreases from 20 fs to the subcycle regime (0.2 fs), D increases by approximately 10 orders of magnitude. Such a dramatic increase in emission current density is attributed to the presence of high-energy photons for shorter laser pulses. This may also be seen from the uncertainty principle, $\Delta\omega\tau_p \gtrsim 0.5$, in which a laser pulse of 0.2 fs would correspond to a variation in the photon energy $\gtrsim 1.65$ eV, which is even higher than the fundamental laser photon energy of 1.55 eV at 800 nm wavelength. The lower left inset shows the difference $\delta = D_{\phi=0} - D_{\phi=\pi}$ (blue) and the relative difference $\delta/D_{\phi=0}$ (red) of the electron

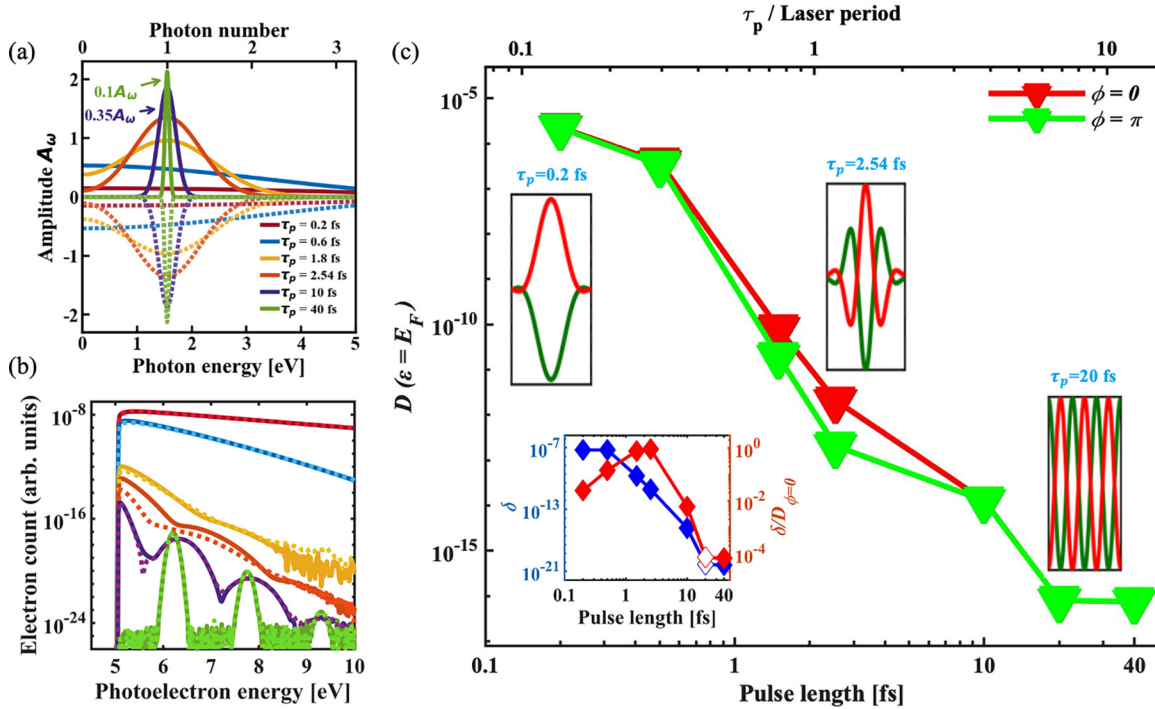


FIG. 1. (a) Optical field spectra $A_\omega = \int_{-\infty}^{\infty} F(t) \cos(\omega t) dt$ for varying pulse lengths with CEP of 0 (solid) and π (dotted). A scaling factor of 0.35 and 0.1 is employed to the cases of $\tau_p = 10$ and 40 fs, respectively, to facilitate the plotting for comparison. (b) Photoelectron emission spectra per pulse D ($\varepsilon = E_F$) for varying pulse lengths at $\phi = 0$ (solid) and $\phi = \pi$ (dotted). (c) Time-averaged emission probability current density per pulse D ($\varepsilon = E_F$) for varying pulse lengths. Lower left inset shows the difference (δ) and difference ratio ($\delta/D_{\phi=0}$) of D between $\phi = 0$ and $\phi = \pi$, where a filled diamond represents a positive value, while an empty diamond indicates a negative value. The other insets show the laser field profile with $\phi = 0$ (red) and $\phi = \pi$ (green) for $\tau_p = 0.2, 2.54$, and 20 fs, respectively.

emission yield for $\phi = 0$ and $\phi = \pi$. The emission current density difference δ increases as the pulse length decreases, while the relative difference peaks at 2.54 fs, near the laser period of 2.67 fs. This indicates that the CEP effect becomes most apparent when the optical field pulse length approaches a single cycle of the laser. However, when the optical pulse length is in the deep subcycle regime [$\ll$ laser period; cf. inset for 0.2 fs in Fig. 1(c)], the effect of CEP becomes less important; instead, the electron emission characteristics are mainly determined by the substantial frequency broadening. Note here we only consider photoemission due to low-intensity lasers, where the Keldysh parameter [37] $\gamma = \omega\sqrt{2mW_0}/eF_1 \gg 1$ and optical field emission becomes irrelevant. Note also that while a pulse duration of 0.2 fs at 800 nm lies beyond the capabilities of present near-infrared laser systems, it is used here as a theoretical limiting case to reveal the QE scaling in the subcycle regime and to emphasize the theoretical strength of the predicted effect.

Figure 2(a) depicts the total transmission probability per pulse, D ($\varepsilon = E_F$), as a function of peak laser strength F_1 . As the pulse length increases from 0.2 to 1000 fs, D decreases and approaches that of continuous wave (CW) laser excitation (black line). It is clear that the differences in electron emission resulting from variations in pulse lengths decrease as the laser intensity increases. In the multiphoton emission regime with low laser intensity (i.e., below the threshold intensity determined by the Keldysh parameter), it is known that the emission current scales as $J \sim F_1^{2n}$, where the value of n indicates the dominant n -photon process. The dotted lines in

Fig. 2(a) represent the fitting $\sim F_1^{2n}$. Notably, the exponent n decreases from 4 to 1 as the pulse length decreases, as shown in Fig. 2(b), reflecting a shift in the dominant emission process from four-photon to an effective one-photon absorption. The former is expected by considering the ratio of work function to photon energy at 800 nm, which predicts that absorbing four photons is required to overcome the surface

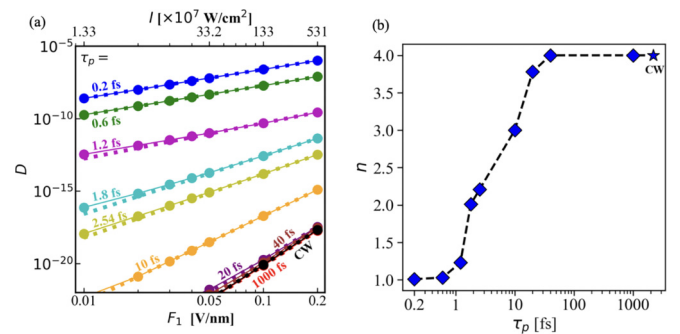


FIG. 2. (a) The total transmission probability per pulse D ($\varepsilon = E_F$) as a function of F_1 for varying pulse lengths from 1000 to 0.2 fs. The CW result is also included for comparison. The CEP for all the laser pulses is $\phi = 0$. The dc field is $F_0 = 0.001$ V/nm. The dotted line represents the fitting curve F_1^{2n} , with $n = 1.01, 1.03, 1.23, 2.01, 2.21, 3.00, 3.78, 4.00, 4.00$, and 4.00, respectively, from top to bottom. (b) The fit parameter n from panel (a), plotted as a function of pulse length.

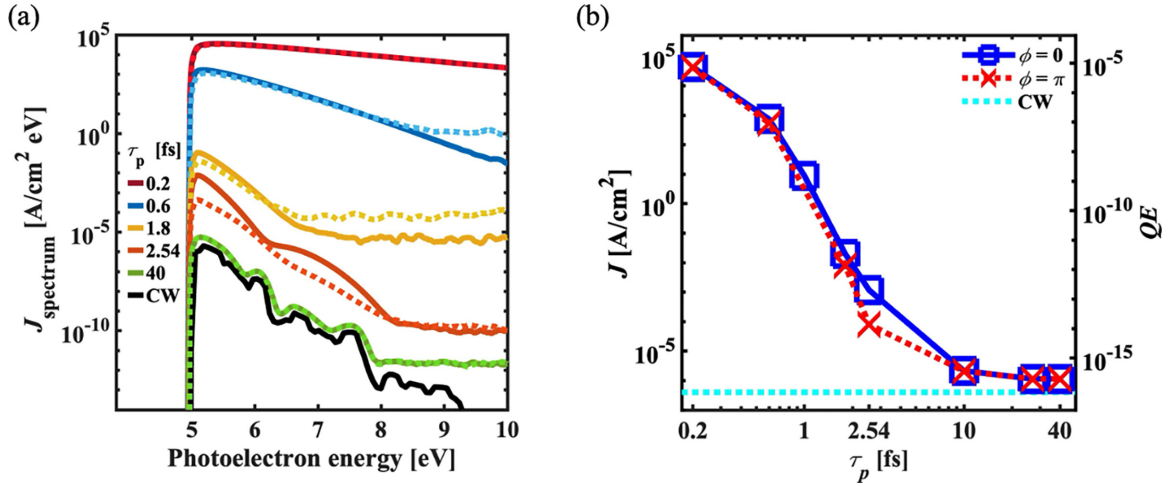


FIG. 3. (a) Emitted electron energy spectra J_{spectrum} as a function of photoelectron energy (with respect to Fermi energy) in varying pulse lengths of 0.2, 0.6, 1.8, 2.54, and 40 fs with CEP of 0 (solid) and π (dotted). The black line is for CW excitation. (b) Emission current density and QE per pulse as a function of pulse length for $\phi = 0$ (solid blue) and $\phi = \pi$ (dotted red). Here, the dc field at $F_0 = 0.00075$ V/nm and peak laser strength at $F_1 = 0.3$ V/nm, corresponding to a peak intensity $I = \epsilon_0 c F_1^2 / 2 = 1.19 \times 10^{10}$ W/cm².

barrier. This shift correlates with variations in the laser spectra width illustrated in Fig. 1(a), substantiating the changes in photoemission dynamics. For shorter pulses, photoemission originates from photons with larger energy in the laser pulse spectrum, resulting in the absorption of fewer photons under these conditions. The shift of dominant photon order is also reflected in the shift of the photoelectron energy peaks as shown in Fig. 1(b), where the dominant peak starts to appear around the Fermi level of 5.1 eV (instead of the four-photon peak of $1.55 \text{ eV} \times 4 = 6.2 \text{ eV}$) when the laser pulse length becomes 10 fs or less. These findings are consistent with the results reported in Refs. [28,38] for the dependence of QE on laser wavelengths and Ref. [39] on the influence of the laser pulse shaping.

Figure 3(a) shows the emitted electron energy spectra from Eq. (3) by considering contributions from all the energy levels inside the cathode under various laser pulse lengths, with CEP $\phi = 0$ (solid line) and $\phi = \pi$ (dotted line). For the long laser pulses (e.g., 40 fs and CW), while multiphoton peaks are still visible in the emission energy spectrum, the spectrum exhibits a jagged shape in contrast to the distinct peaks as in Fig. 1(b), indicating emission contributions from multiple initial energy levels in the cathode. As the pulse length decreases, the electron energy spectra smooth out and become more similar to those in Fig. 1(b), where emission only from the Fermi level is considered. The resemblance of the photoelectron energy spectra between Figs. 1(b) and 3(a) for laser pulses shorter than 40 fs indicates that electron emission occurs predominantly near the Fermi level under shorter laser pulse conditions. In Fig. 3(b), we plot the emission current density and QE as a function of laser pulse length. At 40 fs, both current density J and QE approach those of CW excitation. When the pulse length decreases from 40 fs (15 laser cycles) to 27 fs (10 cycles), both J and QE increase only slightly as the emission process is similar to that of CW laser-induced electron emission, consistent with previous studies [30,38]. When the optical field pulse further decreases from 27 to 0.2 fs, we observe a remarkable increase of about 10 orders

of magnitude in emission current density and QE at an optical field $F_1 = 0.3$ V/nm, corresponding to a peak laser intensity of $I = \epsilon_0 c F_1^2 / 2 = 1.19 \times 10^{10}$ W/cm², for both $\phi = 0$ (solid) and $\phi = \pi$ (dashed). The corresponding number of emitted electrons per pulse $J\tau_p/e$ increases from $2.8 \times 10^{-9} \mu\text{m}^{-2}$ at $\tau_p = 40$ fs to $0.5 \mu\text{m}^{-2}$ at $\tau_p = 0.2$ fs. This significant enhancement in QE indicates that photoemission can become practically detectable with few-cycle or subcycle optical field excitation, without increasing the optical intensity. Note again that the CEP effect becomes only apparent when the optical pulse length is close to one laser cycle, highlighting the dominance of the optical field spectrum broadening effect over the pulse shape difference induced by CEP.

It is important to note that all our calculations so far are for Fourier-limited laser pulses with a constant spectral phase [see Eq. (1)]. We also repeated the analysis using laser pulses with random spectral phase (RSP) while keeping the spectral amplitude and bandwidth unchanged. The RSP pulses strongly suppress the peak electric field due to the loss of temporal coherence and therefore reduce the total emission rate relative to transform-limited pulses. Nevertheless, the trend of increasing emission current density and QE with decreasing pulse duration remains unchanged, indicating that the scaling behavior reported here is not restricted to the transform-limited pulses.

Moreover, to further assess the plausibility of the calculated QE values, we compare our model predictions with representative experimental observations. In our previous work (Ref. [28]), without using any fitting parameters, the quantum model yielded emission current densities and QE values in quantitative agreement with experiment [27]. In addition, using the experimental parameters reported in Ref. [40] and assuming a tip radius of 6.5 nm, our model predicts emitted electrons per pulse of $\mu_e = 0.05$ and $\mu_e = 0.12$ at near-field intensities of $I_{\text{NF}} = 8.07 \times 10^{12}$ and 9.59×10^{12} W/cm², respectively. These values fall within the range of experimentally measured values, $\mu_e \approx 0.04 - 0.22$ for near-field intensities $I_{\text{NF}} \approx (7.6 - 11.3) \times 10^{12}$ W/cm² in Fig. 2(c) of Ref. [40]. These comparisons support the validity

of our model and the physical plausibility of the QE values obtained in this work.

IV. CONCLUSION

In conclusion, we have presented a substantial enhancement of the QE for photoemission by decreasing the optical pulse length to a few cycles. As the optical pulse length decreases, the emitted electron energy spectra change from distinct multiphoton peaks to a smeared broadened distribution. Power-law fitting of emission current as a function of laser intensity reveals a transition from four-photon to an effective one-photon absorption process. These effects are attributed to the broadening of the laser frequency spectrum as the laser pulse shortens, enabling the contributions of higher-energy photons in the photoemission process. Furthermore, we have shown that the impact of CEP on QE and photoelectron energy spectra is significantly dependent on pulse length, with the most profound effect when the pulse length approaches one optical cycle but diminishing with subcycle pulses. The findings enhance our understanding of photoemission mechanisms driven by few-cycle optical field pulses and provide insights into practical detection of photoemission

at low laser intensities, which are critical for advancing the development of ultrafast optics-enabled devices and systems. The macroscopic enhancement of photoemission efficiency due to few-cycle optical fields may be regarded as yet another example of a macroscopic effect arising from quantum physics. We hope that this study will inspire experimental investigations to validate and further explore the predicted phenomena.

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DATA AVAILABILITY

The data supporting this study's findings are available within the article.

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