

Dual-mechanism tuned double-walled carbon nanotube cold-cathode for ultrafast electron emission

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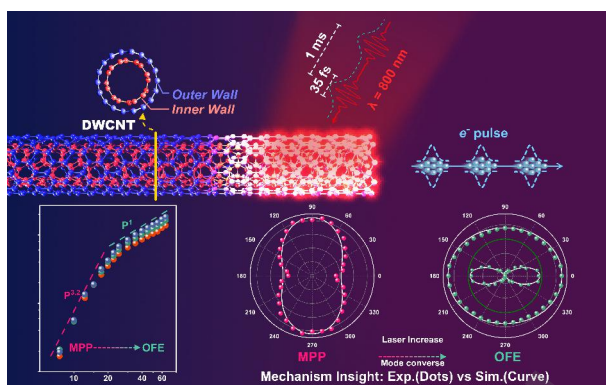
Nano Res., **Just Accepted Manuscript** • <https://doi.org/10.26599/NR.2026.94909024>

<https://www.sciopen.com/journal/1998-0124> on Jul. 13, 2026

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A semiconducting DWCNT cold-cathode is fabricated via in-situ nanomanipulation, enabling dual-mechanism (MPP/OFE) ultrafast electron emission with 65 A peak current and $4.98 \times 10^{18} \text{ A m}^{-2} \text{ sr}^{-1} \text{ V}^{-1}$ brightness, offering a tunable platform for high-performance ultrafast electron sources, with quantitative models developed to elucidate the polarization-dependent emission dynamics under dual-mechanism modulation.

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Received: 5 June 2026; Revised: 10 July 2026; Accepted: 13 July 2026

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 **Cite this article:** *Nano Research*, 2026, 19, 94909024 <https://doi.org/10.26599/NR.2026.94909024>

ABSTRACT: Cold-cathode ultrafast electron source is a key component for probing ultrafast dynamics behavior in materials, as well as developing high-frequency and high-power electromagnetic radiation devices. Developing large-current, high-brightness and tunable ultrafast electron sources by leveraging the intrinsic properties of nanomaterials is significant. In this study, we report the in-situ assembly of a double-walled carbon nanotube (DWCNT) cold-cathode based on a tungsten (W) tip via nanotransfer manipulation within a SEM chamber, enabled by electron-beam-induced carbon deposition. The resulting ultrafast electron emission exhibits excellent performance under dual-regime modulation by multiphoton photoemission (MPP) and optical field emission (OFE). Under co-excitation by 800 nm femtosecond laser pulses and a static electric field, the DWCNT cold-cathode demonstrates significantly enhanced emission in both regimes with a maximum peak current of ~65 A and corresponding brightness of $4.98 \times 10^{18} \text{ A m}^{-2} \text{ sr}^{-1} \text{ V}^{-1}$, and its optical excitation threshold reduces by an order of magnitude compared to the conventional metallic W tip cathode at equivalent emission current levels. Comprehensive material characterizations combined with density functional theory (DFT) calculations reveal that the semiconducting nature of the DWCNT emitter, along with its favorable electronic density of states and correspondingly lower effective work function, provides distinct advantages over metallic CNT and W for ultrafast electron emission. Furthermore, quantitative models are developed for both MPP- and OFE-dominated regimes, which elucidate polarization-dependent electron emission behavior and its underlying physical mechanisms. This work presents a promising semiconducting DWCNT cold-cathode for high-performance ultrafast electron sources, and provides a path for investigating ultrafast electron emission dynamics from multiple perspectives.

KEYWORDS: ultrafast electron emission, dual-mechanism emission, cold cathodes, tunable photoemission

1 Introduction

Cold-cathode ultrafast electron sources, which combine nanometre-scale spatial confinement with femtosecond temporal resolution, are pivotal for probing atomic-scale ultrafast dynamics and serving as key enabling technologies for high-frequency and high-power electromagnetic devices, being potential for various applications such as ultra-high spatiotemporal resolution electron microscopy[1-6], compact terahertz sources[7, 8], miniaturized X-ray sources[9-11], and high-sensitivity electromagnetic detectors[12, 13]. Typically, such an ultrafast electron source is triggered by illuminating the cathode with focused femtosecond laser pulses. Over the past decade, the phenomena and mechanisms of ultrafast laser-driven electron emission (*i.e.* photoemission) have been a subject of extensive research, revealing rich nonlinear physics under intense illumination[14-23]. Within this context, two of the most representative ultrafast emission regimes have been identified: as laser intensity increases, the dominant mechanism transitions from multiphoton photoemission (MPP) to optical field emission (OFE)[19, 24]. In the MPP regime, a valence electron absorbs two

or more photons within an ultrafast timescale, gaining sufficient energy to overcome the surface potential barrier. The resulting electron pulses retain the temporal profile of the driving laser, with pulse durations reaching approximately 10 fs[25]. In contrast, during the OFE regime, the laser electric field directly extracts electrons into vacuum within a fraction of an optical cycle[22], yielding electron pulses that are phase-locked to the optical waveform and capable of attosecond durations[20, 22].

For both above regimes, ultrafast electron sources require the assembled cold-cathode emitters capable of delivering femtosecond electron pulses from ultra-narrow spatial confinement under strong geometric field-enhancement. To date, metallic tungsten (W), gold, and silver have served as the most used cold-cathode materials for previous investigations[20-22, 26-28]. Although plasmonic field enhancement has been observed at specific wavelengths[21, 26, 28], these metals still exhibit high excitation thresholds across the MPP-to-OFE transition[15, 24] and face significant fabrication challenges in achieving atomically sharp, high-aspect-ratio emitters leading to only modest geometric field-enhancement factors. Carbon nanotube (CNT) which can be readily synthesized through self-assembly,

represents a promising alternative to conventional metallic emitters[15, 16, 29-31]. It possesses ultrahigh-aspect-ratio[24, 32], atomically sharp emitting edge[30, 33], narrow energy spread[15, 16, 30], broadband optical absorption[15], and excellent mechanical stability[34, 35], making it suitable for ultrafast electron emission. Recent studies have reported an optical field enhancement factor of 26.7 and an energy spread as low as 0.25 eV for CNTs cold-cathodes^[15], while another demonstrated over a 25-fold increase in emission current under two-color laser excitation[29]. Notably, structural parameters, such as wall number[36], cap closure[37], chirality[38, 39], and internal electronic interactions[31], have been shown to govern the intrinsic optical and electronic properties of CNTs[40]. This tunability renders CNTs an ideal platform for exploring ultrafast electron emission mechanisms and applications.

In this work, we utilize electron-beam-induced deposition and *in-situ* curing within a scanning electron microscope (SEM) to precisely transfer and seamlessly weld an individual double-walled carbon nanotube (DWCNT) bundle onto a metallic W tip, resulting in a monolithic, tip-integrated nano-cold-cathode. Under *co*-excitation by 800 nm, 35 fs laser pulses and a static bias field, the DWCNT cold-cathode demonstrates an order-of-magnitude lower excitation threshold than a pristine W tip cold-cathode, along with substantially enhanced emission currents across both the MPP and OFE regimes, achieving a maximum peak current of ~65 A corresponding to a brightness of $4.98 \times 10^{18} \text{ A m}^{-2} \text{ sr}^{-1} \text{ V}^{-1}$. Comprehensive material characterizations combined with density-functional theory (DFT) calculations reveal that the semiconducting nature and tailored electronic density of states (DOS) of the selected DWCNT emitter enable superior optical coupling and reduced surface potential barriers relative to metallic DWCNTs and pure W tips. Polarization-resolved analyses, interpreted through developed quantitative models for MPP and OFE dominated emission, unveil a strong anisotropy in electron yield with respect to laser polarization, highlighting the critical role of dual-mechanism tuned ultrafast laser-excitation in photoemission. These findings establish semiconducting DWCNT cold-cathode as a robust and tunable platform for ultrafast electron sources, offering a pathway toward multidimensional control and device application of ultrafast electronics.

2 Results and discussion

We first investigate the emission current characteristics of the DWCNT cold-cathode under femtosecond laser excitation. A schematic illustration of the electron emission dynamics is shown in **Figure 1a**. In contrast to previous studies where CNT cold-cathodes were implemented as large-area films or bulk arrays[29, 30, 41], in this study, we employed an individual DWCNT bundle structure onto a W tip base to fabricate a "DWCNT-on-W tip" cold-cathode (see the lower left inset in **Figure 1b**). This configuration eliminates interference from multiple emission sites, thereby enabling an investigation into intrinsic ultrafast emission mechanisms. Specifically, an individual DWCNT bundle was extracted from thermal-CVD-grown CNTs film and welded onto the metallic W tip by using a modified nanotransfer technique that integrates *in situ* current annealing with electron-beam-induced amorphous carbon deposition (see Experimental Methods and Supporting Information **Figure S1**). This process finally yielded a freestanding DWCNT emitter with a protruding length of ~10 μm , which ensures mechanically robust adhesion and stable electrical contact, providing a well-defined point emitter structure sample for the subsequent experiments (**Figure 1b**). High-resolution transmission electron microscopy (HRTEM) image (see the upper right inset in **Figure 1b**) further confirms its typical double-walled structure, with an inner diameter of $d = 1.2 \text{ nm}$ and an outer diameter of $D = 2.1 \text{ nm}$. These geometric dimensions result in an aspect ratio of $L/D \approx 5000$. Such a high aspect ratio translates into a strong geometric

field-enhancement factor β , enabling efficient cooperative enhancement of the local optical and DC electrostatic fields at the emitter apex[15].

Figure 1c illustrates the electron emission experimental setup. A linearly polarized Ti:sapphire laser (Central wavelength = 800 nm, Pulwidth = 35 fs, Repetition rate = 1 kHz) is directed through a half-wave plate, Glan prism, polarizer, and focusing mirror to achieve peak intensities exceeding 100 GW cm^{-2} at the DWCNT emitter, well above the thresholds for both MPP and OFE[19]. The laser beam incidence at 60° relative to the DWCNT-on-W tip cold-cathode axis. The polarization angle (θ) is defined as 0° when the electric field vector lies in the plane containing the DWCNT and the incident beam, and 90° when perpendicular to that plane. The DWCNT-on-W tip cold-cathode and a copper anode are enclosed within an ultrahigh-vacuum chamber ($< 8.0 \times 10^{-6} \text{ Pa}$), and a three-axis manipulator sets the gap to 650 μm with micrometer precision. A synchronized DC bias (up to 700 V) is applied to the anode to create a laser-DC *co*-stimulation condition, and the resulting emission current is recorded (see Experimental Methods and Supporting Information **Figure S2**). The representative emission current as a function of incident laser pulse intensity is presented in **Figure 1d**, measured under a 0° polarization angle and a 300 V DC anode-bias voltage. Specifically, the curves for the DWCNT-on-W tip and bare W tip exhibit kinks at laser intensities of 55 GW cm^{-2} and 500 GW cm^{-2} , respectively. Within the Keldysh framework[19, 42, 43], these kinks are defined as $\gamma = 1$. The regions below these kink intensities correspond to the typical MPP regime, where $\gamma > 1$. In the MPP regime, the emission current (I_{MPP}) typically scales with the n th power of the average laser power (P)[44, 45]:

$$I_{\text{MPP}} \propto P^n \#(1)$$

with n denoting the effective number of photons absorbed by the emitter. Keeping the focused spot area constant (see Experimental Methods), we fitted the low-intensity regions of both curves with Eq. (1), yielding exponents of 3.25 (for DWCNT) and 3.16 (for W). Thus, in both cathodes electrons absorb on average three 1.55 eV photons ($\lambda = 800 \text{ nm}$), surpassing the surface work functions (see the ultraviolet photoelectron spectroscopy (UPS) results in **Figure 2** and **Figure S3** (Supporting Information): 4.4 eV for DWCNT, 4.6 eV for W) and escaping into vacuum. Applying the same procedure to the high-intensity region beyond the kink gives effective photon numbers below unity for both cathodes, indicating that OFE now dominates and MPP is strongly suppressed ($\gamma < 1$)[43].

Across both MPP and OFE emission regimes, the fabricated DWCNT-on-W tip cold-cathode exhibits over a ten-fold enhancement in emission current compared to the pure W tip one under identical laser excitation intensities. This translates to an order-of-magnitude reduction in optical excitation threshold for equivalent emission current levels. These results demonstrate that DWCNT emitters significantly outperform conventional refractory metal cathodes in ultrafast electron emission dominated by both MPP and OFE mechanisms. To further elucidate the advantages of DWCNT over pure W, we analyze both geometric features and intrinsic material properties. First, the enhanced electron emission performance of DWCNT in the OFE regime primarily stems from their remarkable geometric field enhancement factor. Compared to the pure W cathode with a tip diameter of approximately 200 nm, the DWCNT apex curvature radius in this study is only about 1 nm, resulting in a significantly higher aspect ratio (~5000) and consequently enhanced geometric field amplification. This substantially reduces the transition intensity from MPP to OFE regime[24]. Second, the intrinsic material properties of DWCNTs, including their electronic band structure and DOS, profoundly influence the effective work function and electron supply during the opto-electric *co*-excitation ultrafast electron emission process.

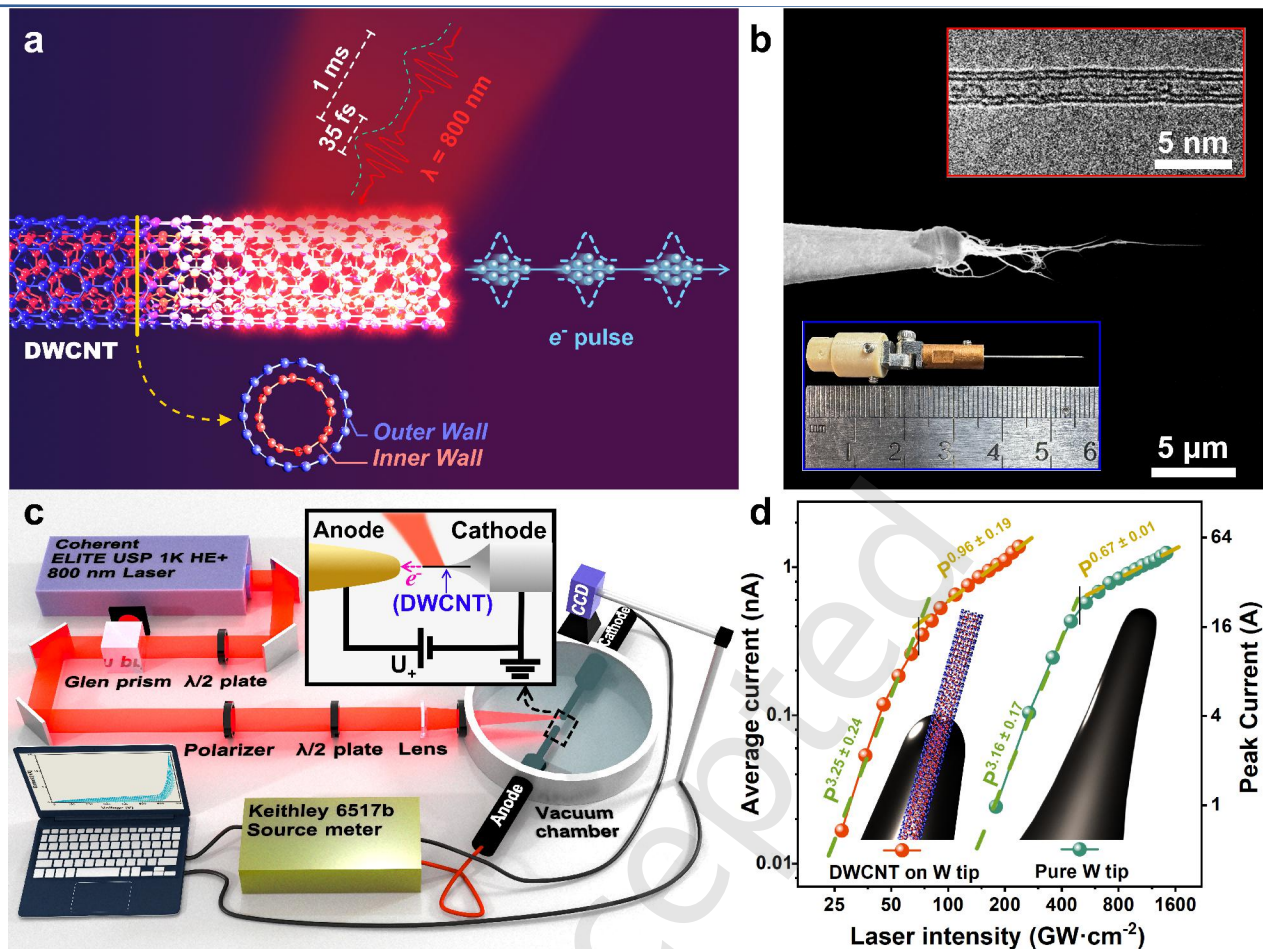


Figure 1 Femtosecond-laser-driven electron emission measurement from the DWCNT-on-W tip cold-cathode. **a**, Schematic of the femtosecond-laser-driven electron emission dynamics of a DWCNT emitter. The carbon atoms colored in red and blue respectively represent the inner and outer walls of the emitter structure. The laser mainly acts on the top end and the nearby stem area of the DWCNT emitter. **b**, SEM image of a typical DWCNT-on-W tip cold-cathode, displaying a long freestanding DWCNT bundle segment ($L \approx 10 \mu\text{m}$) protruding from the bottom W apex. The insets show physical photo of the cold-cathode (lower left) and typical HRTEM image of a DWCNT emitter (upper right), respectively. **c**, Schematic of the electron emission measurement setup. Femtosecond laser pulses are attenuated *via* a half-wave plate and Glen prism, polarization-controlled by a second half-wave plate, focused, and incident at 60° onto the apex of the DWCNT-on-W tip. The inset shows the equivalent circuit diagram for emission current excitation and extraction. **d**, Representative average emission current and peak emission current *versus* incident laser-pulse-intensity curves for DWCNT-on-W tip and pure W tip cold-cathodes.

To determine the electronic structure of the prepared DWCNTs, Raman spectroscopy was performed using a 532 nm laser excitation, as shown in **Figure 2a**. The radial breathing mode (RBM) peak at 187 cm^{-1} is a characteristic signature sensitive to CNT diameter[46]. Based on previous studies and the observed RBM position, the diameter of DWCNT individuals is estimated to be approximately 1.2 nm [47], in excellent agreement with HRTEM measurements (**Figure 1b**). The D peak at 1334 cm^{-1} is barely detectable, indicating high crystallinity, as the D peak originates from vibrational modes of graphitic edge structures and represents disorder[48]. Additionally, the G and G' bands are located at 1576 cm^{-1} and 2651 cm^{-1} , respectively. Peak fitting of the G band enables discrimination between metallic and semiconducting DWCNTs[49]. As shown in **Figure 2b**, the fitted G⁺ peak is centered at 1576 cm^{-1} , while the G⁻ peak appears at 1550 cm^{-1} . The presence of the G⁻ peak, with its typical Lorentzian line shapes rather than the broad and asymmetric Breit-Wigner-Fano profile characteristic of purely metallic counterparts, rules out the possibility of purely metallic characteristics of the prepared DWCNTs. Given that semiconducting CNTs are direct bandgap semiconductors,[50, 51] the Tauc plot method[52] was employed to determine the bandgap, by plotting $(\text{hf}(\text{R}))^2$ *versus* photon energy ($h\nu$) (**Figure 2c**). Extrapolation of this curve yields a bandgap of 0.6 eV for the DWCNTs in this study, directly proves that the prepared and selected DWCNT emitters possess significant intrinsic

semiconducting characteristics. Additionally, the I - V curve obtained during the *in situ* preparation of the ultrafast electron source (**Figure S1**) can also provide supporting evidence. When we performed *in situ* current-induced welding and extraction of an individual DWCNT bundle using a nanomanipulator, it can be deduced from the I - V characteristic curve prior to its electrical breakdown that in the low-voltage region, carrier transport is suppressed due to the existence of an internal energy gap. As the bias voltage increases, the entire I - V curve presents strong nonlinear semiconducting transport characteristics. This is completely different from the classic linear Ohm's law behavior exhibited by metallic CNTs. Therefore, one can essentially conclude that the prepared DWCNT emitter is of a semiconducting nature.

To further characterize the electronic energy levels of the DWCNTs, an UPS measurement was performed. **Figure 2d** and **2e** show the UPS survey spectrum and high-resolution spectra of the secondary electron cutoff and Fermi edge, respectively. The cutoff binding energy is determined to be 17.0 eV , while the Fermi edge is located at 0.2 eV . Combined analysis of UPS and optical absorption data enables construction of the DWCNT energy level diagram relative to vacuum level, as schematically illustrated in **Figure 2f**. Using He I radiation (photon energy = 21.2 eV) in UPS, the conduction band minimum (CBM), Fermi level (E_F), and valence band maximum (VBM) are determined to be at -4.2 eV ($17.0 - 21.2 = -4.2 \text{ eV}$), -4.4 eV ($-4.2 - 0.2 = -4.4 \text{ eV}$, *i.e.*, work function as 4.4

eV), and -4.8 eV (-4.2 - 0.6 = -4.8 eV), respectively, all referenced to the vacuum level. For comparison, UPS characterization of a pure metallic W tip was also performed, and the corresponding energy

level diagram is provided (Figure S3). The Fermi level of W is determined as -4.6 eV (work function as 4.6 eV).

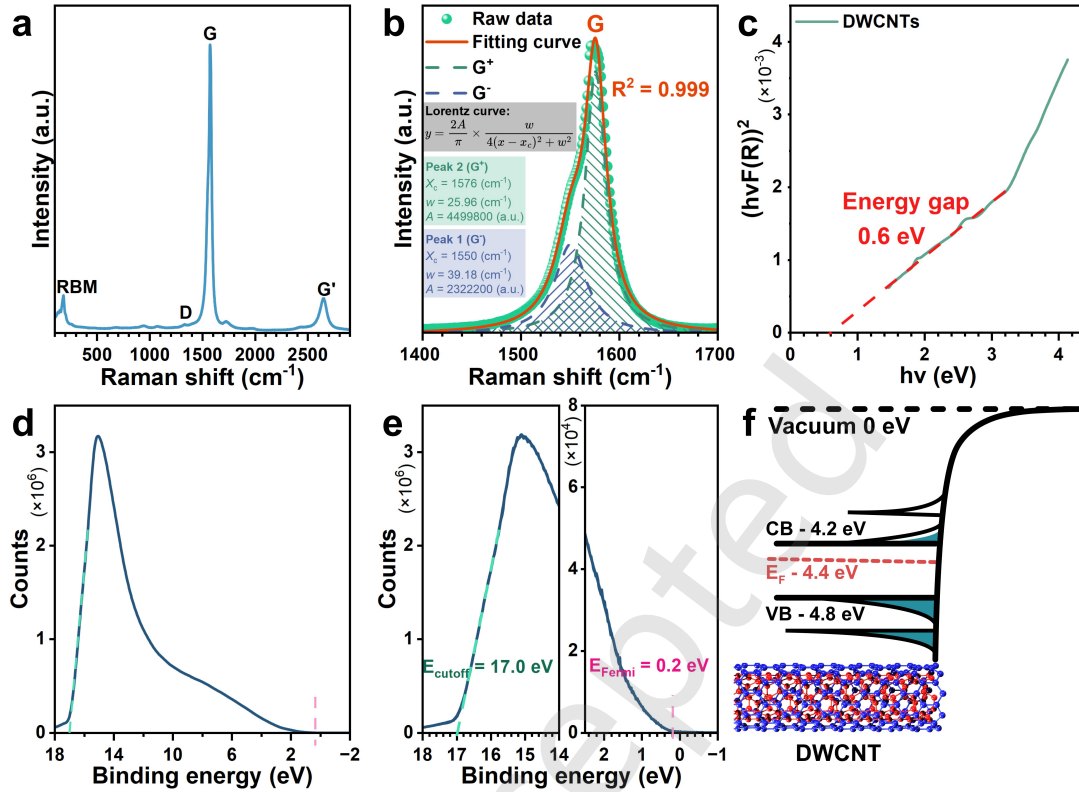


Figure 2 Material characterization of the prepared DWCNTs. **a**, Raman spectrum. **b**, Raman spectrum of high-resolution G-band region in **a**, with deconvolution into G^+ and G^- components. **c**, Tauc plot $(h\nu F(R))^2$ - $h\nu$, revealing a direct band gap of 0.6 eV for DWCNTs. **d**, UPS survey spectrum. **e**, Fine UPS spectrum of the secondary-electron cutoff and Fermi-edge regions in **d**. **f**, Schematic energy-level diagram of DWCNTs derived from **c** and **e**.

After obtaining the basic energy level structure of DWCNTs, we constructed three distinct chiral DWCNT models in Materials Studio software: (16,16)@(9,9), (28,0)@(16,0), and (20,10)@(12,6). These three specific configurations were strategically selected based on a dual-constraint strategy combining experimental geometric dimensions with electronic structure diversity. On one hand, to strictly replicate the actual morphology of the experimental samples, the selected DWCNT models exhibit theoretical inner and outer diameters of approximately 1.2 nm and 2.1 nm, respectively, which precisely match the statistical diameter distribution derived from the HRTEM image (Figure 1b) and Raman RBM peaks (Figure 2a). On the other hand, to comprehensively systematically investigate how the intrinsic electronic properties affect the dual-mechanism tuned ultrafast electron emission, these configurations were chosen to represent three distinct topological archetypes: a fully metallic symmetric system, a classic zigzag semiconducting system, and a more generalized chiral semiconducting system. Additionally, a pure metallic W crystal with a diameter of 2.1 nm was also constructed as a control structure for comparison. DFT calculations were subsequently performed to compute the electronic DOS for all structures, as shown in Figure 3a. Herein, the (16,16)@(9,9) DWCNT exhibits metallic behavior, characterized by a finite DOS at the E_F , whereas both the (28,0)@(16,0) and (20,10)@(12,6) DWCNTs display distinct band gaps of approximately 0.6 eV at E_F , confirming their semiconducting nature. This calculated value is in excellent quantitative agreement with the 0.6 eV statistical optical bandgap experimentally measured through the Tauc plot results (Figure 2c). Furthermore, the electronic DOS of the metallic W crystal is significantly higher than that of all DWCNT structures. Under the excitation of an external electric field, when the potential difference between the anode and cathode is positive, the Fermi

level of the emitter is elevated by an amount (ΔW) equal to the local potential (U) multiplied by the absolute value of the electron charge (neglecting the Schottky barrier between the emitter and cathode support). Consequently, the induced charge density in the cold-cathode emitter is given by:

$$\rho(U) = -2e \int_{E_F}^{E_F+U} \text{DOS}(E) dE \quad (2)$$

where e is the elementary charge (1.6×10^{-19} C), $\text{DOS}(E)$ is the electronic DOS obtained from Figure 3a, and E_F denotes the equilibrium Fermi level. Based on this formulation, geometric models of different chiral DWCNTs and the pure W crystal were constructed using the Electrostatics module in COMSOL Multiphysics software. All models were set to a uniform length of 10 μm , and a uniform external electric field of 1 $\text{V } \mu\text{m}^{-1}$ was applied. This field strength corresponds to the same order of magnitude as that used in actual measurements (0-700 V bias voltage with fixed vacuum gap of 650 μm). The charge densities for each emitter were solved self-consistently as functions of the local potential (U) using Eq. (2). Under the applied electric field, the spatial variation of the work function compared to the emission apex was extracted along both the radial center and the outer wall (edge) of each DWCNT and W structure, as illustrated in Figure 3b. It can be observed that under the external field, the reduction in work function from the mid-length region to the apex is significantly more pronounced for semiconducting DWCNTs than for the metallic (16,16)@(9,9) DWCNT or the pure W. This behavior is governed by the material-dependent electronic DOS. These results indicate that under the combined excitation of femtosecond laser and electrostatic field employed in this study, the effective work function of semiconducting DWCNTs (applicable to both different chiral

structures) is significantly reduced, enhancing the electron emission current through the following two mechanisms: For regions dominated by the MPP mechanism, the reduced work function at both the middle section and apex of semiconducting DWCNTs effectively lowers the threshold energy required for electrons to absorb photon energy and overcome the surface potential barrier, thereby providing more high-energy electrons. On the other hand, for regions dominated by the OFE mechanism, the significant reduction in work function at the DWCNT apex greatly increases the probability of electron tunneling. Under the influence of these two emission mechanisms, the calculated energy level diagrams for different DWCNT and pure W structures are presented in **Figure 3c**. The results demonstrate that even without considering the half-cycle electric field of the femtosecond laser pulse, the surface work function of semiconducting DWCNT decreases from 4.4 eV (dark state) to approximately 3.2 (~3.19 eV for (28,0)@(16,0) DWCNT and ~3.22 eV for (20,10)@(12,6) DWCNT), which is lower than that of the metallic (16,16)@(9,9) DWCNT (~3.46 eV) and

significantly below that of the 2.1 nm diameter pure W crystal (~4.26 eV) with identical geometry. For the bare W sample with a tip diameter of 200 nm used for experimental comparison in this study (measured initial work function of 4.6 eV), the surface effective work function during the same excitation process was estimated using the identical methodology. As shown in **Figure S4** (Supporting Information), owing to the larger curvature radius of the W tip and its higher bulk electron density, which enables stronger electrostatic screening, the computed work function reduction at the outer edge is only 0.015 eV, whereas the change at the radial center is negligible. Consequently, the overall effective work function decreases only marginally from 4.60 eV to 4.59 eV under identical excitation conditions, resulting in negligible contribution to electron emission enhancement. These results demonstrate that semiconducting DWCNTs are inherently more suitable as photo-coupled excitation cold-cathode materials for ultrafast electron sources.

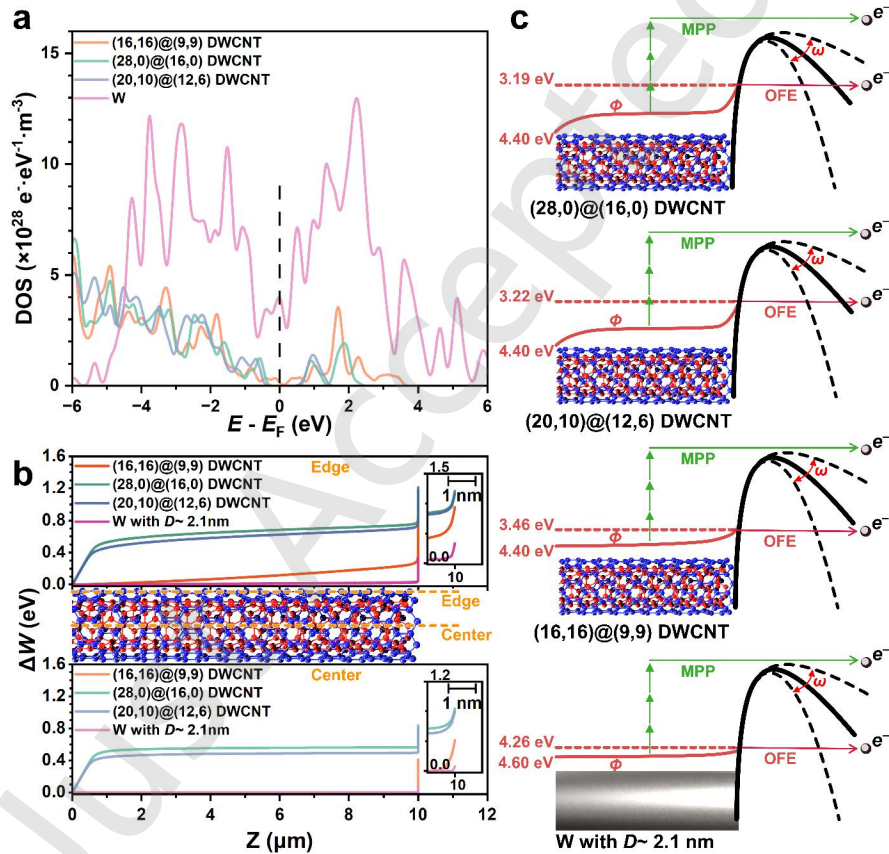


Figure 3 Calculation of effective work functions for different DWCNT and W emitters. **a**, Electronic DOS distributions for various chiral DWCNT structures and pure W crystal obtained through DFT calculations. **b**, Variations in surface work function as a function of distance from the electron emission apex for different structures with an outer diameter of 2.1 nm and length of 10 μm , under a uniform external electric field of 1 $\text{V} \mu\text{m}^{-1}$. **c**, Schematic illustration of electron emission energy levels for different structures after calculation, showing distinct effective work functions under identical excitation conditions.

To elucidate the ultrafast laser-driven electron emission characteristics of the DWCNT cold-cathode under dual-mechanism modulation of MPP and OFE, the emission current measurements were performed as a function of femtosecond laser power and optical polarization. **Figure 4a** and **4b** present the electron emission current *versus* bias voltage (I - V) characteristics of the DWCNT cold-cathode under varying optical powers at polarization angles of 0° and 90° , respectively. In the anode-bias voltage range of 0–600 V, the emission current increases approximately linearly with applied voltage and gradually saturates under both polarization conditions, indicating that the electron emission is predominantly governed by field-assisted photoemission. However, for the bias voltages

exceeding 600 V, the emission current exhibits exponential, nonlinear growth with increasing voltage, that is a signature characteristic of conventional field emission indicating a transition to a photo-electronic synergistic field emission regime. Notably, the cathode emission current is consistently enhanced with increasing optical power across all polarization angles. In the photo-electronic synergistic field emission regime (bias voltage > 600 V), the emission current under 0° polarization exceeds that under 90° polarization at identical laser powers. This anisotropy arises from the alignment between the electric field vector of the incident laser pulse (at 0° polarization) and the direction of the applied

electrostatic field, which enhances the total local field at the cathode apex.

Since the DC component of the total emission current arises from the applied electrostatic bias and makes not contribution to the ultrafast pulsed emission, our analysis focuses on the roles of MPP and OFE in governing ultrafast pulsed electron emission under photoemission-dominated conditions (bias voltage < 600 V). To this end, the emission current (I) as a function of incident optical power (P) was measured at bias voltages of 300, 400, and 500 V under both 0° and 90° polarization conditions, as shown in **Figure 4c** and **Figure 4d**. The calculated maximum peak emission current in these measurements generally reaches over 65 A, corresponding to a current density of over $1.88 \times 10^{18} \text{ A m}^{-2}$ and a peak brightness exceeding $4.98 \times 10^{18} \text{ A m}^{-2} \text{ sr}^{-1} \text{ V}^{-1}$ (see the Supporting Information

for calculation details). Applying the same fitting procedure used in **Figure 1d**, a distinct kink in the emission current dependence is observed at an optical power of 18 mW, which corresponds to a peak laser pulse intensity of $\sim 65.5 \text{ GW cm}^{-2}$ and an optical field amplitude of $\sim 0.703 \text{ V nm}^{-1}$ for both polarization states in the DWCNT cold-cathode. Below this threshold (MPP regime), the photoemission current follows a power-law dependence on optical power with exponents of approximately 3.2 to 3.3, implying effective absorption of 3.2 photons (0° polarization) and 3.3 photons (90° polarization) by the DWCNT emitter. Above an optical power P of 20 mW, the I - P relationship transitions to a linear dependence (order ≈ 1.0) under 0° polarization and a near-linear dependence (order ≈ 0.9) under 90° polarization, consistent with theoretical predictions for the OFE regime.

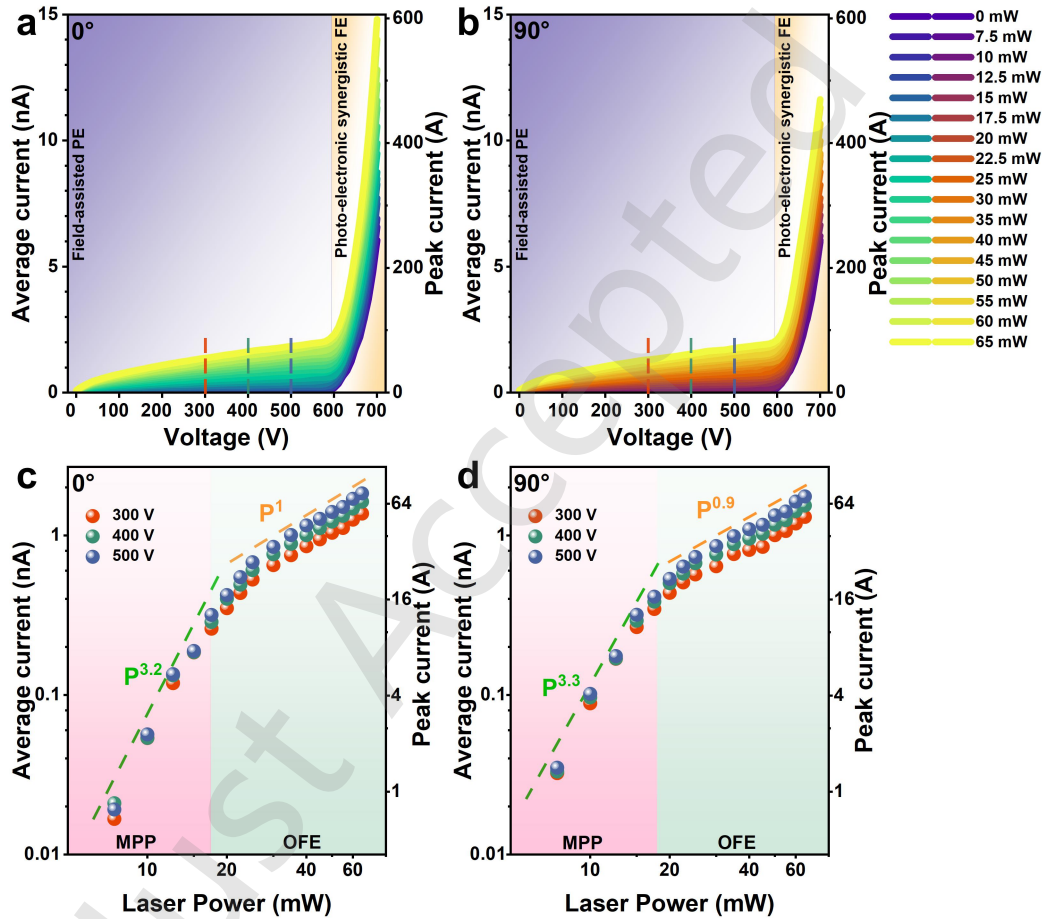


Figure 4 Femtosecond laser plus DC bias co-excited electron emission characteristics of the DWCNT-on-W tip cold-cathode. a, b, Emission current-bias voltage (I - V) characteristics under 0° and 90° polarized laser illumination at varying optical powers. c, d, Emission current-optical power (I - P) characteristics under applied bias voltages of 300 V, 400 V, and 500 V for 0° and 90° polarized lasers.

To further quantify the electron emission behavior of the DWCNT-on-W tip cold-cathode under the modulation of different mechanism regimes, the polarization angle of the incident laser was systematically varied from 0° to 360° in 10° increments, and the polarization-dependent emission current characteristics were measured at incident optical powers of 10 mW and 45 mW, respectively. Herein, at P of 10 mW (corresponding to peak pulse intensity of 36.4 GW cm^{-2} and optical field amplitude of 0.524 V nm^{-1}), MPP dominates the electron emission process. In contrast, at P of 45 mW (corresponding to peak pulse intensity of 164 GW cm^{-2} and optical field amplitude of 1.112 V nm^{-1}), electron emission is predominantly governed by OFE. The measured emission currents as a function of polarization angle at different applied bias voltages of 300 V, 400 V, and 500 V are plotted in polar coordinates in

Figure S5a and **S5b** (Supporting Information), revealing two distinctly different angular dependencies for MPP and OFE regimes.

In the MPP-dominated regime, the polarization-resolved emission current (**Figure S5a**, Supporting Information) exhibits pronounced anisotropy: the current is maximized when the laser polarization (*i.e.*, the electric field vector) is perpendicular to the plane formed by the DWCNT axis and the incident laser beam direction, with a maximum near 90° while a minimum near 0°. According to Eq. (1), if MPP were the sole mechanism and given that photon absorption occurs uniformly along the DWCNT axis, the emission current would be expected to exhibit negligible polarization dependence, resulting in an isotropic angular distribution. Indeed, the fitted effective photon numbers at 0° and 90° polarization are nearly identical (3.2 and 3.3, respectively; see **Figure 4c** and **Figure 4d**), confirming the weak polarization dependence of the MPP process

itself. However, the experimental results clearly exhibit anisotropic behavior, indicating the presence of additional physical mechanisms influencing emission in the MPP-dominated regime. One likely contributor is polarization-dependent photo-thermal energy loss within the DWCNT emitter during laser excitation.

To quantitatively assess the influence of photo-thermal energy loss on MPP and its role in modulating the polarization-dependent electron emission from the DWCNT-on-W tip emitter, we extracted a representative current–polarization curve measured at a bias voltage of 500 V (**Figure 5a**). By incorporating photo-thermal energy loss as a limiting factor, Eq. (1) was extended resulting in the following modified expression (see Supporting Information for derivation):

$$I_{\text{simulate}} = k(P_{\text{total}} - P_{\text{qth}})^n \quad (3)$$

where I_{simulate} is the simulated emission current that accounts for photo-thermal energy loss. The coefficient k is a proportionality constant that depends on factors including the effective emission area, multiphoton absorbing quantum efficiency, and laser pulse duration; it was empirically determined to be $2.61 \times 10^{-13} \text{ A W}^{-n}$. The exponent n , representing the effective number of photons absorbed per emission event, ranges from 3.2 (0° polarization) to 3.3 (90° polarization), as determined from previous experimental fits. P_{total} is the instantaneous total optical power of the incident femtosecond pulse, and P_{qth} denotes the polarization-dependent photo-thermal loss power within the DWCNT emitter (see Supporting Information), expressed as:

$$P_{\text{qth}} = \frac{1}{T} \iint_{V,t} \mathbf{F} \cdot \mathbf{J} dV dt \quad (4)$$

where T is the optical cycle period, $\mathbf{F}$ is the transient electric field of the incident laser, $\mathbf{J}$ is the photo-induced current density inside the DWCNT emitter, V is the nanotube volume, and t is time. Herein, P_{qth} is intrinsically a linear absorbed power, which is governed by the complex permittivity of the DWCNT emitter and is entirely converted into lattice internal energy *via* electron-phonon scattering. In contrast, the MPP current arises from a coherent quantum behavior where electrons instantaneously and simultaneously absorb multiple photons within an ultrashort timescale to overcome the effective work function and escape into the vacuum. This falls under the category of a nonlinear optical absorption process under strong-field excitation. In standard microscopic electromagnetic field simulations, this nonlinear quantum transition process cannot be encompassed within the aforementioned linear macroscopic

calculations based on the complex permittivity. Based on this physical decoupling, Eq. (3) actually constructs a localized effective nonlinear optical field modulation model after subtracting the intrinsic thermal losses. Only after deducting the specific portion of optical power that is linearly absorbed by the system and dissipated as thermal energy, can the remaining localized effective electromagnetic energy serve as the driving source to excite electrons within the DWCNT for nonlinear multiphoton coherent transitions into the vacuum. Using COMSOL Multiphysics software, we constructed a model of the DWCNT structure and simulated the spatial distribution of photo-thermal losses under varying polarization states (see Supporting Information **Figure S6**). By integrating the spatially resolved thermal losses, the total P_{qth} for each polarization angle was obtained. The calculated polarization dependence of P_{qth} over the 0° to 360° range is shown in **Figure 5b**. Notably, the photo-thermal loss is significantly higher at 0° polarization than at 90° , attributable to the quasi-one-dimensional structure of DWCNT and their intrinsic anisotropic optical dielectric function. This trend exhibits fourfold symmetry, repeating periodically across all quadrants. These values were then substituted into Eq. (3) to compute the simulated MPP emission currents (I_{simulate}) across all polarization angles (see Supporting Information Table S1), which were plotted in polar coordinates (red curve, **Figure 5a**). The simulated curve shows agreement with the experimentally measured MPP emission current profile at 500 V, capturing the characteristic anisotropy induced by photo-thermal effects. This agreement confirms that, under MPP dominance, the polarization dependence of ultrafast laser-driven electron emission from the DWCNT cold-cathode is predominantly governed by photo-thermal energy loss. Under the modulation of polarization, an increase in P_{qth} indicates enhanced linear optical dissipation, which consequently reduces the net optical power effectively coupled into the nonlinear MPP channel.

In contrast, in the OFE-dominated regime (**Figure S5b** in Supporting Information and **Figure 5c**), a polarization dependence opposite to that of the MPP mechanism is clearly observed: the emission current peaks at 0° polarization and reaches a minimum at 90° . To quantitatively analyze the electron emission behavior in the OFE-dominated regime, we derive an analytical expression for the current density of the DWCNT emitter by extending the quasi-static Fowler-Nordheim (F-N) model to include the time-varying electric field component from femtosecond laser excitation (see Supporting Information for derivation)[13, 20, 53, 54]:

$$J(x,y,t) = \frac{C_1(\beta_{\text{optical}}(x,y) \cdot F(t) + \beta_{\text{electric}} F)^2}{\phi} \cdot e^{-\frac{C_2 \cdot v(f) \cdot \phi^{3/2}}{\beta_{\text{optical}}(x,y) \cdot F(t) + \beta_{\text{electric}} F}} \quad (5)$$

where $F(t)$ is the time-dependent electric field component of the femtosecond laser field, F is the bias static electric field, $\beta_{\text{optical}}(x,y)$ represents the local optical field enhancement factor, β_{electric} is the electrostatic field enhancement factor, $v(f)$ is the barrier correction factor[55] (see Supporting Information), and ϕ is the effective work function of the DWCNT surface. The constants are defined as $C_1 = 1.54 \times 10^{-6} \text{ A eV V}^{-2}$; $C_2 = 6.83 \times 10^9 \text{ V eV}^{-3/2} \text{ m}^{-1}$. The spatial distribution of $\beta_{\text{optical}}(x,y)$ across the DWCNT emission apex in the XOY plane was obtained *via* finite-difference time-domain (FDTD) simulations (see Supporting Information **Figure S7**). Under ultrafast optical excitation, the electric field component of the femtosecond laser varies dynamically in time. During the optical half-cycle, the field can be approximated as a sinusoidal waveform, with the laser electric field magnitude oscillating periodically (see Supporting Information). In contrast, the electrostatic field enhancement factor β_{electric} , arising from the high aspect ratio geometry of the DWCNT emitter, is determined as

a constant value (~ 2790) by fitting the field emission I - V characteristics and their corresponding F-N plots under dark conditions (laser off), as shown in **Figure S8** and detailed in the Supporting Information.

Using these parameters, the OFE current density $J(x,y,t)$ at any given position and instant on the DWCNT emission apex can be calculated. Notably, during numerical computation, the Keldysh parameter γ is evaluated to preclude contributions from non-tunneling mechanisms (*e.g.*, MPP), ensuring that only tunneling-dominated OFE current is included (see Supporting Information). Subsequently, by integrating $J(x,y,t)$ over the duration of a single femtosecond laser pulse ($\sim 35 \text{ fs}$) at each spatial point, one can obtain the spatially resolved average current density $\bar{J}(x,y)$ across the emission apex under different polarization angles (see Supporting Information **Figure S9**). This enables the reconstruction of the temporal profile of the OFE current $I_{\text{OFE}}(t)$ within one laser pulse cycle (**Figure 5d**). The computed $I_{\text{OFE}}(t)$ exhibits a clear

trend: its peak amplitude monotonically decreases as the polarization angle increases from 0° to 90° , with maximum emission at 0° and minimum at 90° . This trend exhibits fourfold symmetry across all quadrants. The polar plot of the polarization-dependent average current $\bar{I}$, derived from $I_{\text{OFE}}(t)$, matches the experimental measurements in polar coordinates (red curve in **Figure 5c**). Moreover, both the theoretically predicted and experimentally measured $I_{\text{OFE}} \sim \cos^6(\theta)$ dependence were

observed[15], as illustrated in the yellow inset of **Figure 5c**. These results not only validate the accuracy of the proposed OFE current model for DWCNT emitters but also demonstrate that ultrafast laser-driven electron emission can be effectively modulated by the polarization state of the driving optical field in the OFE-dominated regime.

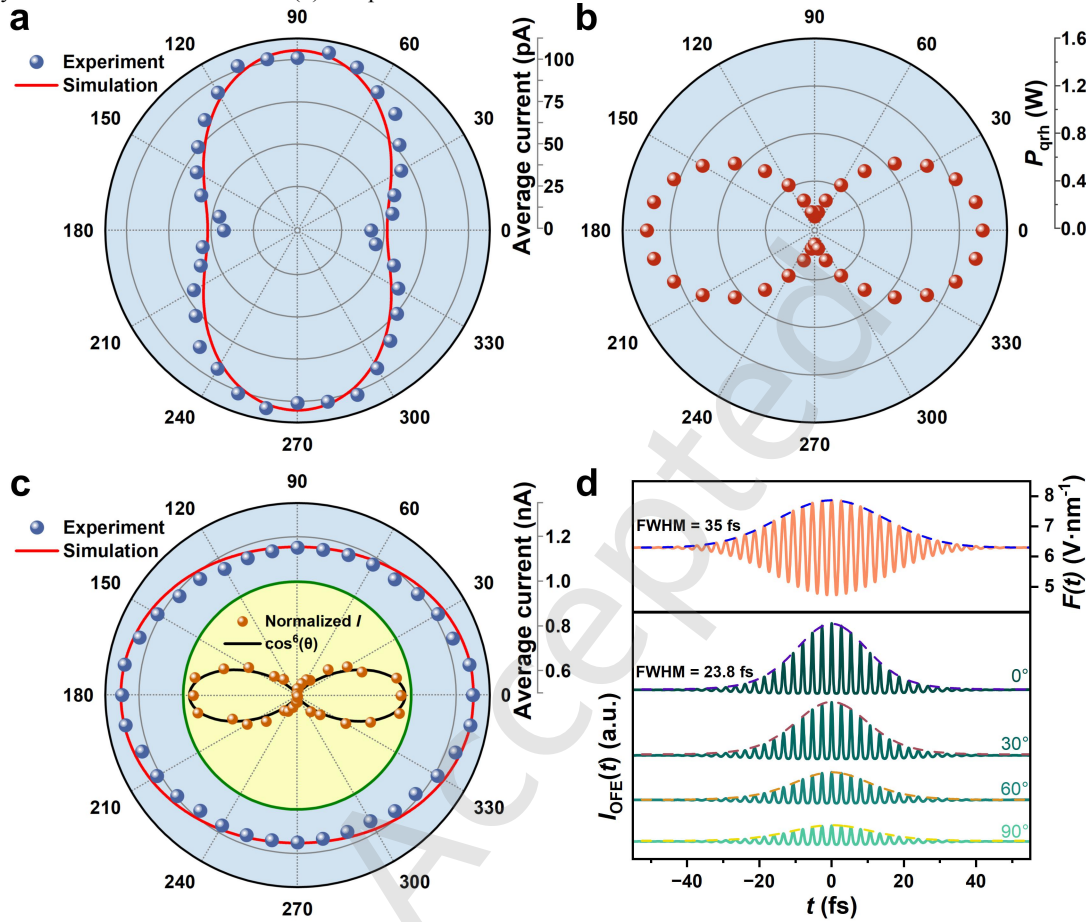


Figure 5 Quantitative analysis of electron emission current characteristics from the DWCNT-on-W tip cold-cathode under the modulation of MPP and OFE dual mechanisms. a, Typical polar plots of emission current *versus* laser polarization angle in the MPP regime, along with simulated MPP current curves accounting for photo-thermal energy losses. b, Polarization dependence of the calculated photo-thermal losses in the DWCNT structure. c, Measured emission current *versus* polarization angle curves in the OFE regime and the simulated emission currents using an extended quasi-static F-N emission model including the time-varying electric field component from femtosecond laser excitation. The inset (yellow) shows the normalized emission current (I) as a function of the incident laser polarization angle, revealing a $\cos^6(\theta)$ dependence. d, Theoretically calculated temporal profiles of the time-varying electric field component under femtosecond laser illumination and the corresponding OFE current response within a single femtosecond excitation pulse (FWHM~35 fs).

The DWCNT cold-cathode developed in this work exhibits excellent polarization-sensitive emission current characteristics across both the MPP and OFE regimes. The emission current can be flexibly modulated through the interplay of these two distinct mechanisms. The combined theoretical analysis and model development establish a robust framework for active control of ultrafast laser-driven electron emission dynamics in DWCNTs under diverse excitation conditions. By harnessing this dual-mechanism tunability, the DWCNT-based cold-cathode may be engineered into high-performance devices capable of ultrafast electron polarization modulation, enabling advances in technologies, including ultrafast electron microscopy, terahertz sources, synchrotron radiation sources, and free-electron lasers, thereby providing more precise and efficient electron source solutions for fundamental scientific research.

3 Conclusions

In summary, we have successfully fabricated a semiconducting DWCNT integrated onto a W tip *via in-situ* nano-manipulation

within a SEM, realizing a monolithic cold-cathode with exceptional potential as a high-performance ultrafast electron source. Under identical femtosecond laser illumination and static electric field *co*-excitation conditions, the emission current from this semiconducting DWCNT cold-cathode is enhanced by more than an order of magnitude relative to conventional metallic W tip cathodes, achieving a maximum peak emission current of ~65 A and brightness of $4.98 \times 10^{18} \text{ A m}^{-2} \text{ sr}^{-1} \text{ V}^{-1}$. Comprehensive material characterizations, coupled with DFT calculations, reveal distinct advantages in both geometric field enhancement and intrinsic electronic properties. The DWCNT cold-cathode exhibits a pronounced polarization-dependent emission response in both the MPP and OFE regimes. To elucidate this dual-mechanism-modulated electron emission behavior, quantitative models incorporating both photo-thermal loss in MPP and time-resolved field enhancement in OFE have been developed. These insights into the underlying physical mechanisms guide the pathway for the development of large-current, high-brightness and tunable cold-

cathode ultrafast electron sources and vacuum nano-optoelectronic devices.

4 Methods

Synthesis of DWCNT Films: DWCNT films were synthesized by thermal chemical vapor deposition (CVD) under high vacuum ($< 10^{-4}$ Pa) using ethanol as the carbon source. The catalyst was composed of ammonium iron (III) citrate (3 wt%) and MgO (97 wt%)[56]. The reaction was carried out at 875 °C under a pressure of ~70 mbar with ethanol vapor for 30 min. After growth, the as-synthesized product was immersed in concentrated HCl (~37 wt%) to remove residual metal catalysts, followed by thermal annealing in air at 400 °C for 30 min to remove amorphous carbon. A second HCl treatment was applied to ensure complete removal of residual catalyst. To selectively remove the more reactive single-walled CNT, the sample was annealed in air at 500 °C for 2 h. The final semiconductor-enriched DWCNTs were collected by filtration, thoroughly rinsed, and dried to form a free-standing film.

Transfer and Fabrication of the DWCNT-on-W Tip Cold-Cathode: Within a SEM chamber, a W tip probe mounted on a nanomanipulator was precisely positioned to approach a DWCNT film. A bundle of DWCNTs was mechanically extracted via controlled lateral dragging. From this bundle, DWCNTs with high aspect ratios and structurally pristine morphologies were selectively identified. A second W tip probe was then brought into physical contact with the chosen DWCNT to form a mechanical junction, as documented in Fig. S1 (see Supporting Information). Upon contact, an electrical pathway was established between the DWCNT and the W tip. By applying a voltage and driving a high current through the junction *in situ*, substantial Joule heating occurred due to the initially high contact resistance, inducing localized thermal welding that firmly anchored the DWCNT to the W tip. To further enhance mechanical robustness and electrical continuity, electron beam-induced amorphous carbon deposition was performed at the DWCNT-W interface for 1 h under SEM guidance, reinforcing the structural integrity of the junction. Finally, the first W tip probe was carefully retracted, allowing the DWCNT to remain attached to the second W tip probe, resulting in a free-standing, DWCNT-on-W tip cold-cathode.

Material Characterizations: The morphology and structure of the materials were analyzed by scanning electron microscopy (SEM, Zeiss SUPRA-60) at an accelerating voltage of 10 kV and high-resolution transmission electron microscopy (HRTEM, FEI Titan G² 60–300) operated at 300 kV. Ultraviolet photoelectron spectroscopy (UPS) measurements were conducted on a Thermo Fisher Scientific ESCALAB 250Xi spectrometer using He I radiation (photon energy: 21.2 eV) to probe the electronic structure of the materials. Raman spectroscopy was performed using a confocal Raman system (Renishaw inVia Reflex) coupled with a dark-field optical microscope (Leica, 50× objective, numerical aperture = 0.8), employing a 532 nm laser (power: 80 mW, continuous wave) for excitation, with an integration time of 10 s per acquisition. UV–Vis absorption spectra were recorded using a Hitachi U-4100 spectrophotometer with a wavelength step of 1 nm and an integration time of 1 s per data point.

Ultrafast Laser-Driven Electron Emission Measurements: The experimental setup is schematically illustrated in Fig. 1c. A femtosecond laser amplifier system (Coherent, ELITE USP 1K HE+) served as the excitation source, generating linearly polarized pulses with a pulse width of 35 fs, a center wavelength of 800 nm, and a repetition rate of 1 kHz. The laser polarization was initially aligned parallel to the plane of the optical table. The incident laser intensity was precisely controlled using a combination of a Glan Thompson polarizer and a half-wave plate ($\lambda/2$), allowing for both coarse and fine tuning of the pulse energy while maintaining consistent illumination conditions across all polarization and intensity

configurations. Before reaching the cold-cathode, the polarization state of the laser beam was further purified using a high-extinction-ratio polarizer, and the linear polarization angle was precisely tuned from 0° to 360° using a second half-wave plate. The laser beam was focused onto the apex of the DWCNT emitter using a focusing objective, with an incidence angle of 60° relative to the emitter axis. To ensure uniform optical excitation, the focused spot size was maintained at a constant diameter of 1 mm across all measurements, fully encompassing the DWCNT emitter apex. This was confirmed by imaging using a CMOS CCD camera (Basler, acA1920-40um) mounted outside the vacuum chamber (see Fig. S2a, Supporting Information). The absolute laser fluence incident on the emitter surface was calibrated using a thermopile-based optical energy meter (Ophir, PE9-CO; model PE9-RS). For accurate energy measurement, the laser beam was directed perpendicularly onto the sensor surface at the same spatial position previously occupied by the cold-cathode, thereby minimizing errors due to polarization-dependent detector responsivity. Laser power stability was maintained with a precision better than $\pm 2\%$ under all experimental conditions.

The electron emission measurements were performed in an ultrahigh-vacuum chamber with a base pressure of 8.0×10^{-6} Pa. The DWCNT-on-W tip cold-cathode (inset in Fig. 1b) and the copper anode were independently mounted on precision three-axis micrometer translation stages, enabling accurate spatial alignment. After focusing the laser beam onto the apex of the DWCNT emitter, the cold-cathode position was fixed. The anode was then adjusted to achieve a fixed vacuum gap of 650 μm between the cold-cathode tip and the W anode, as confirmed by a CMOS CCD image (Fig. S2b, Supporting Information). All electron emission measurements were conducted at this fixed vacuum gap distance. During laser excitation, a static bias voltage was applied to the copper anode through a picoammeter (Keithley, Model 6517B), which simultaneously measured the resulting emission current from the cold-cathode. The electric field in the vacuum gap was calculated as the ratio of the applied bias voltage to the gap distance, yielding the average macroscopic field.

DFT Simulation: All calculations were performed within the framework of DFT, using the generalized gradient approximation (GGA) in the form of the Perdew–Burke–Ernzerhof (PBE) functional for the exchange–correlation potential[57]. Core electrons were described using effective core potentials (ECPs), and valence electrons were treated with the double-numerical plus polarization (DNP) basis set. All calculations were carried out using the DMOL3 package[58, 59]. The K-point mesh of $1 \times 1 \times 2$ was used for the DWCNTs, and $4 \times 4 \times 4$ for the W tip. The convergence tolerance in the energy is 10^{-5} Ha, and the maximum allowed force and displacement are 0.002 Ha $\text{\AA}^{-1}$ and 0.005 $\text{\AA}$, respectively. For the DWCNT models, the supercell lattice constants in the directions perpendicular to the tube axis were set to $40 \text{\AA} \times 40 \text{\AA}$ to ensure that there was no interaction.

Simulation of Work Function Variation under Electrostatic Field: The work function variation of a material under an electrostatic field was simulated using the electrostatics module in COMSOL Multiphysics Software. A representative DWCNT with an outer diameter of 2.1 nm and a total length of 10 μm was modeled as a conductive cylindrical nanostructure. A grounded boundary condition (0 V) was applied to the bottom end of the DWCNT emitter, while a potential of 20 V was imposed on a reference plane located 20 μm above the substrate. To establish a uniform background electric field of $1 \text{ V } \mu\text{m}^{-1}$ in the absence of the DWCNT, a cylindrical symmetry boundary with a radius of 10 μm , coaxial with the DWCNT was defined, enforcing zero normal electric field component (*i.e.*, insulating boundary condition).

FDTD Simulation: Optical absorption and local optical-induced electric field enhancement around the DWCNT structure were

simulated using the FDTD method in Lumerical FDTD Solutions. The complex refractive index of the DWCNT was adopted from the experimental data reported by Timothy et al.[60]. A plane wave source at 800 nm was incident at a fixed angle of 60° relative to the DWCNT axial direction, with the polarization angle varied from 0° to 90° in 10° increments. The boundary condition used in the simulation area is the Bloch boundary condition, with a mesh size of 0.5 nm. To compute the localized optical field enhancement factor ($\beta_{\text{optical}}(x, y)$), a total-field/scattered-field (TFSF) source at 800 nm was used under identical polarization configurations. Perfectly matched layers (PMLs) were applied at all boundaries to absorb outgoing waves, with a refined mesh size of 0.1 nm in the vicinity of the nanotube. The resulting electric field distributions, field enhancement factors, and absorption profiles were recorded using frequency-domain field and power monitors.

Photo-Thermal Energy Loss Simulation: The photo-thermal loss in DWCNT structure was simulated using COMSOL Multiphysics. A DWCNT structure with an outer diameter of 2.1 nm and length of 10 μm was illuminated by a light source with intensity matching experimental conditions. The incident light was directed at an angle of 60° relative to the DWCNT axis, with propagation vector lying in the YOZ plane. The optical properties of the DWCNT were consistent with those employed in the FDTD simulations. A scattering domain of $1.6 \times 1.6 \times 12 \mu\text{m}^3$ was defined around the DWCNT to ensure sufficient separation between the nanostructure and the domain boundaries. A 200 nm-thick PML was implemented to absorb scattered light, and scattering boundary conditions (SBCs) were applied at the outer boundaries to further suppress numerical reflections. The total photo-thermal loss was obtained by spatial integration of the volumetric loss density over the DWCNT volume for each polarization state.

Supplementary Material: Supplementary material is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94909024>.

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Acknowledgements

This work was financially supported by the National Key Research and Development Program of China (Grant no. 2025YFA1213400), the National Natural Science Foundation of China (Grant no. 62471498), Department of Science and Technology of Guangdong Province (Grant no. 2025TQ09X991), the Guangdong Basic and Applied Basic Research Foundation (Grant no. 2025A1515010637), the Science and Technology Planning Project of Guangdong Province (2023B1212060025), and the Fundamental Research Funds for the Central Universities, Sun Yat-sen University.

Declaration of competing interest

The authors declare no competing interests.

Author contribution statement

S.D. and Y.S. conceived the idea and initiated the present study. S.D., Y.S., S.X., and Z.S. proposed the experimental scheme. S.X. and Z.S. carried out the experiments. W.W., Z.S., X.Z., R.Z., Y.W. and Z.L. assisted in the theories and experiments. S.D., Y.S., W.W., S.X. and Z.S. discussed and interpreted the results. S.T., Y.Z. and H.C. assisted in the data analysis. S.X., Z.S., Y.S., W.W. and S.D. co-wrote the manuscript.

Use of AI statement

None.

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Just Accepted

Dual-mechanism tuned double-walled carbon nanotube cold-cathode for ultrafast electron emission

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Supporting information to <https://doi.org/10.26599/NR.2026.94909024>

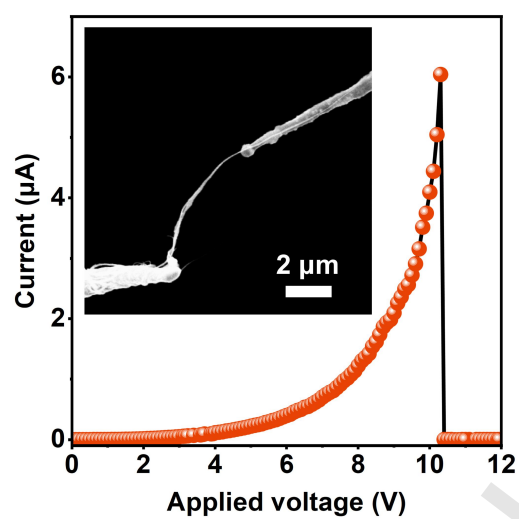


Fig. S1. Experimentally recorded current–voltage (I – V) characteristics of *in-situ* current heating and DWCNT bundle melting in a SEM chamber. The inset shows the SEM image illustrating the material transfer process during the fabrication of the DWCNT-on-W tip cold-cathode.

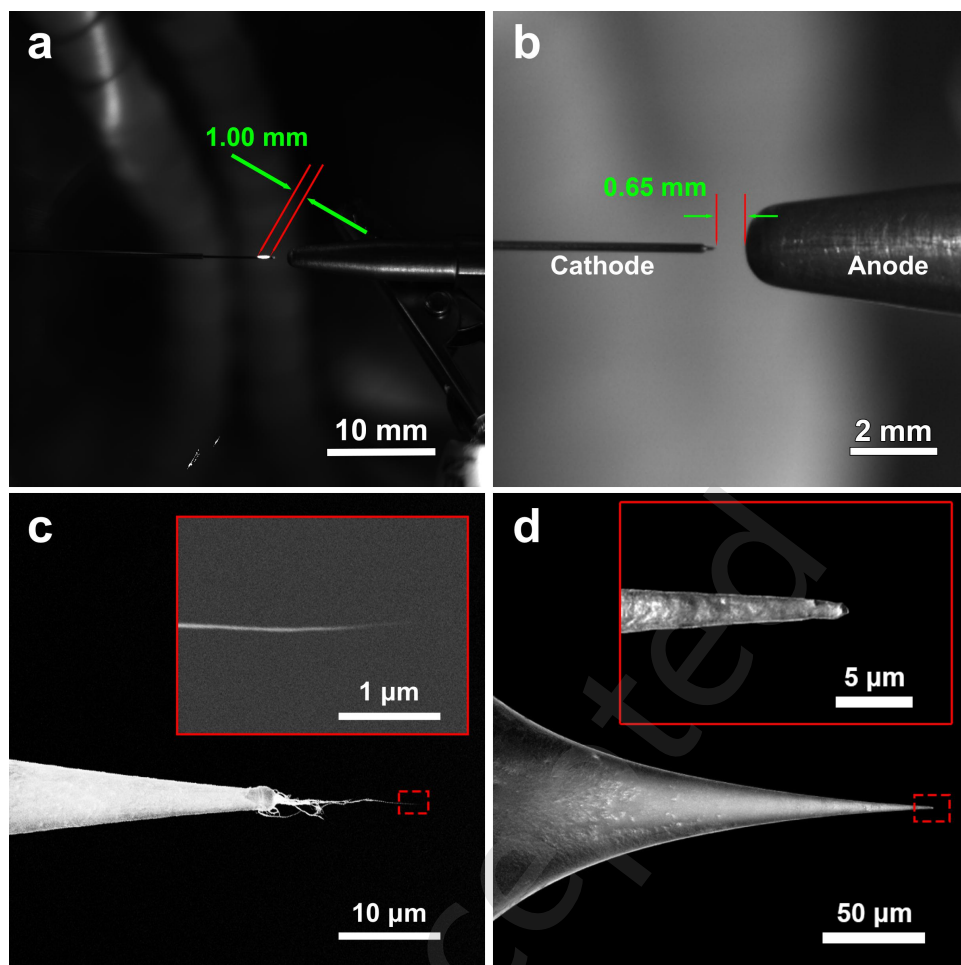


Fig. S2. Measurement of electron emission characteristics from a DWCNT-on-W tip cold-cathode under femtosecond laser excitation. a, CCD image of the laser spot focused on the cold-cathode. b, CCD image illustrating the vacuum gap distance between the anode and the cold-cathode. c, d, SEM images of the DWCNT-on-W tip and a pure W tip for comparison, respectively. The insets show high-magnification SEM images of their apex regions. The radius of curvature of the pure W tip sample used for comparison is approximately 100 nm.

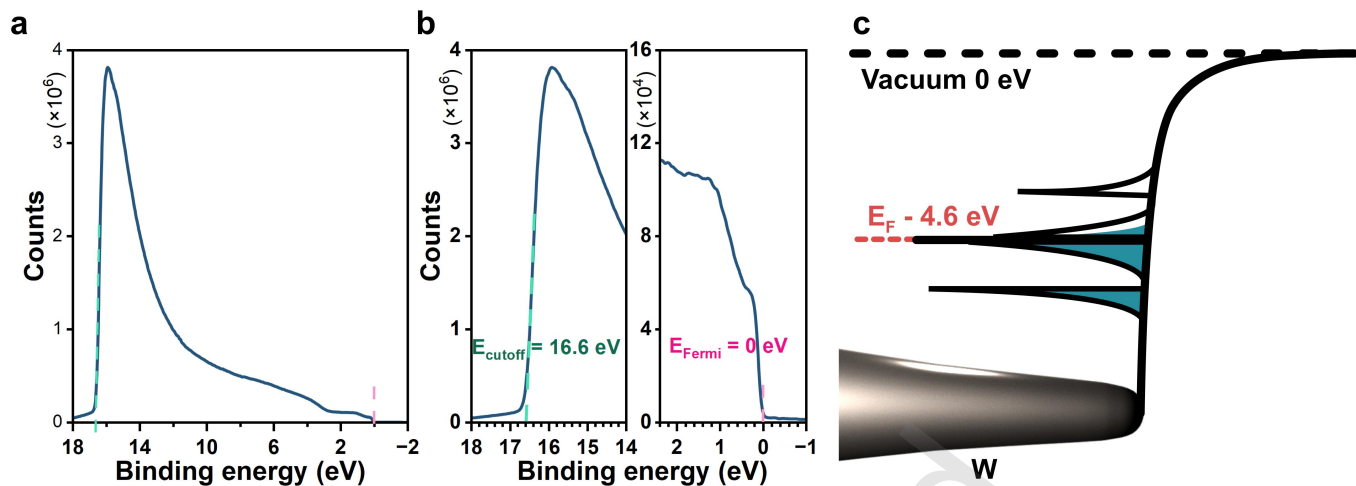


Fig. S3. UPS characterization and electronic band structure of the pure W tip. **a**, Full UPS spectrum of the W sample. **b**, Fine UPS spectra in the regions of the secondary electron cutoff and Fermi edge. **c**, Schematic band structure of W reconstructed from UPS data. The UPS measurements were performed using a He I radiation source (photon energy: 21.2 eV). The binding energy of the Fermi level is determined as 4.6 eV below the vacuum level (*i.e.*, at -4.6 eV relative to the vacuum level, calculated as 16.6 eV - 21.2 eV), yielding a work function of 4.6 eV for the pure W tip.

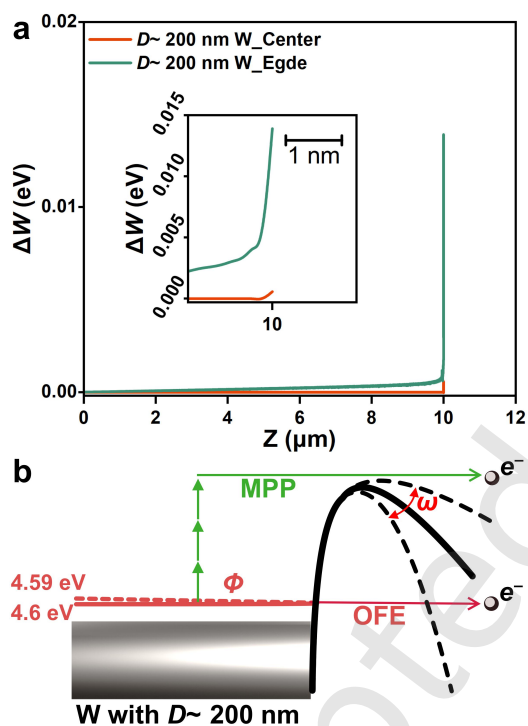


Fig. S4. Calculation of the effective work function for the pure W tip used as a reference in experimental comparisons. **a**, Spatial variation of the work function along a simulated W cylinder structure with diameter of 200 nm and length of 10 μm , modeling the geometry of a pure W tip emitter, as a function of distance from the electron emission apex under a uniform external electric field of $1 \text{ V } \mu\text{m}^{-1}$. **b**, Schematic energy band diagram derived from the simulation, illustrating the effective work function of the pure W tip.

Calculation of the Cold-Cathode Brightness:

The maximum reduced brightness (B_{r_max}) of the DWCNT cold-cathode is defined as the maximum electron flux emitted per unit area per unit solid angle. As expressed in Eq. (S1), J_{max} denotes the maximum emission current density, it equals maximum peak current divided by the emission area ($\pi D^2/4$), where D is the outer diameter of a DWCNT emitter. The parameter h represents the tangential energy (in eV), as detailed in Eq. (S2). The term t_y refers to the mirror-image force correction factor, approximated by Eq. (S3), where with the Nordheim parameter given by $y = 3.79 \times 10^{-5} \times \beta_{optical_max} \cdot F_{max}^{0.5} / \Phi$.^[S1] For this calculation, $\beta_{optical_max}$ was set to 2.92, corresponding to the maximum value in our FDTD simulation (see the Construction of the OFE-Dominated Emission Current section in the Supporting Information file). Furthermore, F_{max} represents the electric field component of laser pulse corresponding to the peak current, while Φ was taken as 4.4 eV based on the UPS characterization.

$$B_{r_max} = 1.44 \frac{J_{max}}{\pi h} \#(S1)$$

$$h = 9.76 \times 10^{-11} \frac{\beta_{optical_max} \cdot F_{max}}{\Phi^{\frac{1}{2}} t_y} \#(S2)$$

$$t_y = 1 + 0.1107 y^{1.33} \#(S3)$$

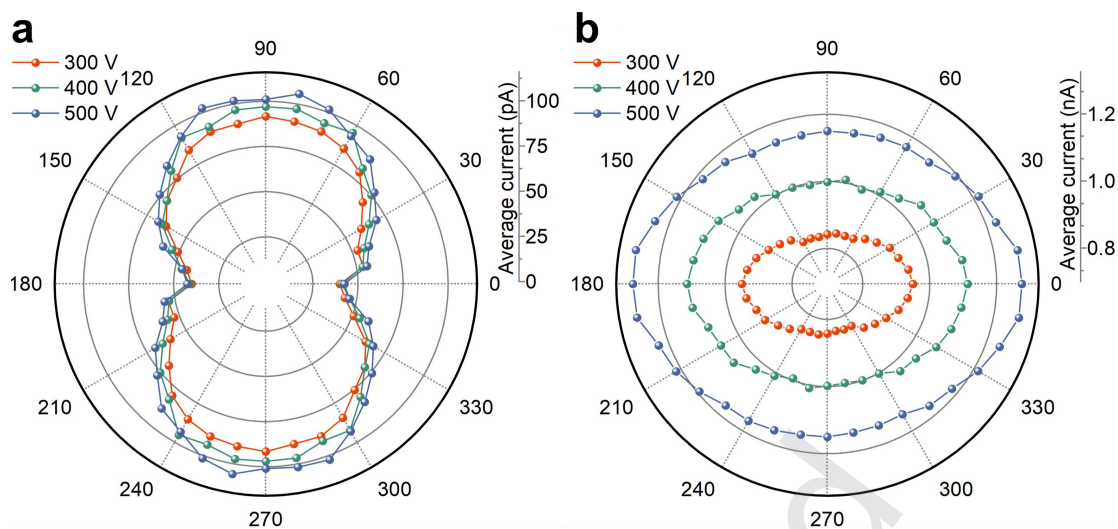


Fig. S5. Polarization-dependent emission current characteristics of the DWCNT-on-W tip cold-cathode. a, Polar plots of the average emission current at a lower incident laser power of 10 mW for the MPP regime, illustrating the intrinsic polarization anisotropy of the cold-cathode. **b,** Polar plots of the emission current at a higher incident laser power of 45 mW, characteristic of the OFE dominated regime.

Construction of the MPP-Dominated Emission Current Model Considering Photo-thermal Energy Loss:

For the simulation of electron emission current from the DWCNT cold-cathode under the MPP regime, photo-thermal energy loss within the nanotube body is considered. The following empirical model is established:

$$I_{\text{simulate}} = k(P_{\text{total}} - P_{\text{qth}})^n \quad \#(S4)$$

where I_{simulate} denotes the simulated MPP current, and k is a positive proportionality coefficient representing the conversion efficiency from absorbed optical power to photoemission current. This coefficient depends on several factors, including the effective emission area of the DWCNT illuminated by the laser, the quantum efficiency of multiphoton absorption, and the effective interaction time of the laser pulse. The exponent n represents the effective number of absorbed photons, as determined from prior experimental results; it varies slightly with polarization angle, ranging from 3.2 (at 0° polarization) to 3.3 (at 90° polarization). P_{total} is the instantaneous total optical power incident on the DWCNT under femtosecond laser excitation. In this study, it is defined as the product of the illuminated area S and the peak laser intensity I_s . For normal incidence, S corresponds to the projected cross-sectional area; however, for 60° oblique incidence, the effective area is reduced by a factor of $\sin(60^\circ)$. For example, at an average incident laser power of 10 mW (corresponding to a peak intensity $I_s = 36.4 \text{ GW cm}^{-2}$) irradiating a DWCNT of length $L = 10 \text{ }\mu\text{m}$ and outer diameter $D = 2.1 \text{ nm}$, the instantaneous total power is calculated as: $P_{\text{total}} = \sin(60^\circ) I_s S = \frac{\sqrt{3}}{2} \times 3.64 \times 10^{10} \text{ W}\cdot\text{cm}^{-2} \times 10 \text{ }\mu\text{m} \times 2.1 \text{ nm} = 6.62 \text{ W}$. P_{qth} denotes the photo-thermal power loss in the DWCNT under different polarization states, obtained *via* finite-element simulations using COMSOL Multiphysics, incorporating realistic optical excitation conditions. Specifically, P_{qth} is computed as the time-averaged volumetric integral of the dot product of the electric field vector $\mathbf{E}$ and the induced current density $\mathbf{J}$:

$$P_{\text{qth}} = \frac{1}{T} \iint_{V,t} \mathbf{E} \cdot \mathbf{J} dV dt \quad \#(S5)$$

where T is the optical cycle, $\mathbf{E}$ is the transient electric field component of the incident laser, $\mathbf{J}$ is the photo-induced current density within the DWCNT structure, V is the nanotube volume, and t is time. In the simulations, the electric field amplitude is set to match the experimental condition ($F_0 = 0.524 \text{ V nm}^{-1}$). The resulting spatial distribution of P_{qth} over one optical cycle is obtained, and its cross-sectional profile in the XOZ plane is presented in Fig. S6.

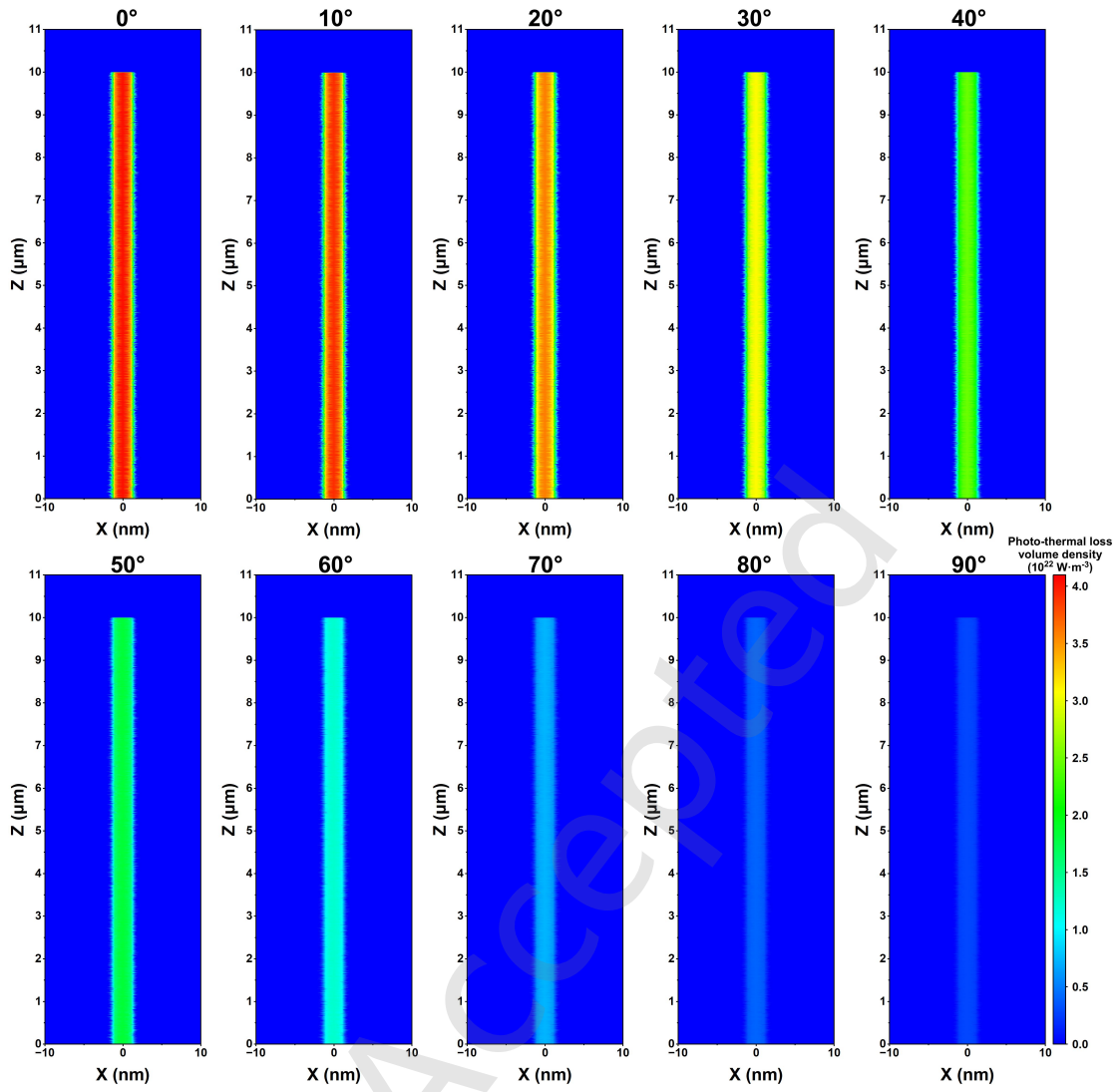


Fig. S6. Simulated spatial distribution of photo-thermal loss (P_{qrh}) in DWCNT structure under femtosecond laser illumination as a function of polarization angle (from 0° to 90° , in 10° increments).

After obtaining the spatial distribution of the photo-thermal loss power in the DWCNT structure for different optical polarization directions, the next step involves integrating the values over the entire nanotube volume to determine the total photo-thermal loss power (P_{qrh}) within the emission structure. Empirically, the scaling coefficient k is set to $2.61 \times 10^{-13} \text{ A W}^{-n}$, and the P_{qrh} values derived from COMSOL Multiphysics simulations are substituted into Eq. (S4). This yields the simulated ultrafast MPP electron emission current from the DWCNT emitter as a function of polarization angle. Table S1 lists the calculated P_{qrh} , the residual optical power (*i.e.*, the fraction of incident power effectively available for MPP emission, defined as $P_{\text{total}} - P_{\text{qrh}}$), and the corresponding simulated MPP current values for polarization angles ranging from 0° to 90° . The results for the remaining three quadrants (90° – 360°) exhibit high symmetry with those in the 0° – 90° range due to the cylindrical symmetry of the DWCNT structure. These simulated MPP current values are subsequently plotted in a polar coordinate diagram as a function of polarization angle and show agreement with the experimentally measured current–polarization dependence (Fig. 5a).

Table S1. Simulated values of photo-thermal loss, residual optical power, and MPP current at different polarization angles for the DWCNT emitter under MPP-dominated regime, obtained from modeling and simulation.

Polarization	Photo-thermal loss power	Residual optical power	Simulated MPP current
(°)	(P_{qth} , W)	($P_{\text{total}} - P_{\text{qth}}$, W)	(I_{simulate} , pA)
0	1.396	5.224	51.9
10	1.357	5.263	53.1
20	1.244	5.376	56.8
30	1.072	5.548	62.9
40	0.861	5.759	70.8
50	0.636	5.984	80.1
60	0.424	6.196	89.5
70	0.252	6.368	97.7
80	0.140	6.480	103.3
90	0.101	6.519	105.3

Construction of the OFE-Dominated Emission Current Model Considering Quasi-Static F-N Framework Including the Time-Varying Electric Field Component from Femtosecond Laser Excitation:

In this study, the extended quasi-static F-N model for OFE-dominated electron emission is derived from the classical F-N theory of field electron emission, incorporating the time-dependent characteristics of the femtosecond laser electric field. The resulting expression for the emission current density is given by:

$$J(x,y,t) = \frac{C_1(\beta_{\text{optical}}(x,y) \cdot F(t) + \beta_{\text{electric}} F)^2}{\phi} \cdot e^{-\frac{C_2 \cdot v(f) \cdot \phi^{3/2}}{\beta_{\text{optical}}(x,y) \cdot F(t) + \beta_{\text{electric}} F}} \quad \#(S6)$$

where $F(t)$ is the time-dependent electric field component of the ultrafast excitation laser, F is the applied static bias field, and ϕ is the effective work function of the DWCNT. The constants are defined as $C_1 = 1.54 \times 10^{-6} \text{ A eV V}^{-2}$; $C_2 = 6.83 \times 10^9 \text{ V eV}^{-1.5} \text{ m}^{-1}$. Under photo-electric *co*-excitation, the total field enhancement factor β is expressed as a combination of the optical field localization enhancement at the DWCNT emitter (denoted by β_{optical}) and the electrostatic field enhancement arising from geometric effects (denoted by β_{electric}). The barrier correction factor $v(f)$ is given by:

$$v(f) = 1 - f + \frac{f \ln(f)}{6} \quad \#(S7)$$

where:

$$\begin{aligned} f &= \frac{\beta_{\text{optical}}(x,y) \cdot F(t) + \beta_{\text{electric}} F}{F_\phi} \\ &= 1.44 \times 10^{-9} \cdot \frac{\beta_{\text{optical}}(x,y) \cdot F(t) + \beta_{\text{electric}} F}{\phi^2} \text{ eV}^2 \cdot \text{V}^{-1} \cdot \text{m} \quad \#(S8) \end{aligned}$$

Herein, F_ϕ denotes the electric field strength required to fully suppress the Schottky–Nordheim potential barrier, the height of which equals the local work function ϕ . The optical field localization enhancement induced by the femtosecond laser at the DWCNT emitter is modeled using FDTD simulations. As shown in Fig. S7, the optical field enhancement factor (β_{optical}) at the emission apex of the DWCNT emitter exhibits a pronounced dependence on the laser polarization angle. Moreover, because the electric field component of the femtosecond laser varies rapidly in time, its temporal evolution particularly during the negative half-cycle, can be approximated by a sinusoidal function. Consequently, within a single optical cycle, the time-dependent optical electric field is expressed as:

$$F(t) = \sqrt{2} F_0 \cdot \exp - \left(\frac{2\sqrt{\ln 2} \cdot t}{35 \cdot 10^{-15}} \right)^2 \cdot \cos \left(\frac{2\pi t c}{\lambda} \right) \quad \#(S9)$$

where F_0 is the root-mean-square electric field amplitude of the incident laser, c is the speed of light in vacuum, and λ is the wavelength of the incident light (approximately 800 nm). Accordingly, the time-dependent electric field of the 35 fs laser pulse is modeled as a sinusoidal function modulated by a Gaussian temporal envelope.

On the other hand, the local electric field enhancement factor arising from the high-aspect-ratio geometry of the DWCNT-on-W tip emitter (β_{electric}) is assumed to remain constant across different polarization conditions. In this study, we assume that the external morphology and geometric aspect ratio of the DWCNT-on-W tip emitter are preserved before and after the photo-electric *co*-excited ultrafast electron emission process. Consequently, the value of β_{electric} can be extracted by fitting the I - V characteristics and their corresponding F-N plots measured under dark-field conditions (laser off), as shown in Fig. S8.

According to classical F-N theory, the field enhancement factor is $\beta_{\text{electric}} = -C_2 \phi^{1.5}/m$, where m is the slope of the F-N plot, $C_2 = 6.83 \times 10^9 \text{ V eV}^{-3/2} \text{ m}^{-1}$, and ϕ is the surface work function of the cold-cathode emitter, experimentally determined to be 4.4 eV. From the fitting, we obtained a local electric field enhancement factor $\beta_{\text{electric}} \approx 2790$, which is in close agreement with the typical geometric aspect ratio ~ 5000 expected for the DWCNT emitter.

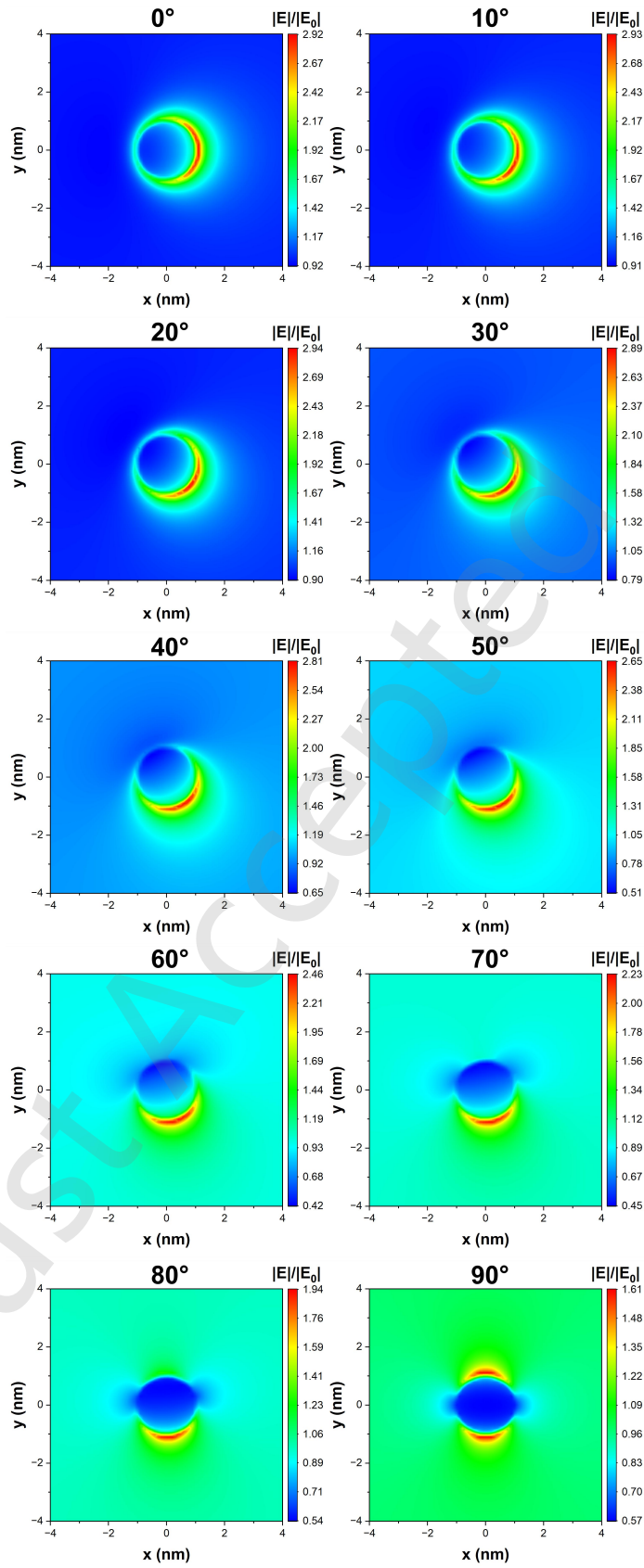


Fig. S7. Spatial distribution of the optical field enhancement factor $\beta_{optical}(x,y)$ at the emission apex of a DWCNT emitter in the XOY plane, computed from FDTD simulations under varying laser polarization angles.

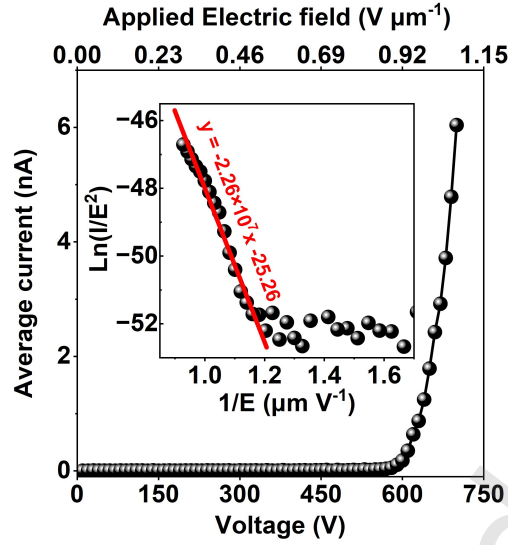


Fig. S8. I - V characteristics of the DWCNT-on-W tip cold-cathode under dark-field conditions (laser off). The inset shows the corresponding F-N plots with a linear fit, confirming field emission behavior.

Next, by substituting the obtained parameters into Eq. (S7), Eq. (S8), and Eq. (S9), and subsequently into Eq. (S6), we calculated the OFE current density $J(x,y,t)$ at a specific location on the emission apex of the DWCNT emitter at a given time. Notably, it is essential to pre-evaluate and exclude contributions from non-OFE mechanisms based on the Keldysh framework. Specifically, when the Keldysh parameter $\gamma > 1$, electron emission is dominated by photon-driven processes, such as single-photon photoemission, multiphoton photoemission, and above-threshold photoemission, which lie outside the regime of applicability of the present OFE model. In contrast, the model is valid only in the tunneling-dominated regime where $\gamma < 1$. Therefore, in describing the OFE physical process, regions or conditions for which $\gamma > 1$ must be excluded. Accordingly, the spatially resolved optical field enhancement factor $\beta_{\text{optical}}(x,y)$ and the near-field electromagnetic distribution are incorporated into the Keldysh framework, yielding:

$$\gamma(x,y,t) = \frac{\omega \sqrt{2m_e \Phi}}{e \beta_{\text{optical}}(x,y) \cdot F(t)} \#(S10)$$

where e is the elementary charge ($\sim 1.60 \times 10^{-19}$ C), m_e is the electron rest mass ($\sim 9.11 \times 10^{-31}$ kg), ω is the angular frequency of the incident laser, and ϕ is the effective work function of the cathode. By numerically evaluating Eq. (S10), any spatial point on the emission apex at time t for which $\gamma(x,y,t) > 1$ is excluded from contributing to the OFE current. Furthermore, the resulting time-dependent current density $J(x,y,t)$ is integrated over the duration of the femtosecond laser pulse to obtain the average current density per laser shot:

$$\bar{J}(x,y) = 1000 \int_0^T J(x,y,t) dt \#(S11)$$

where the factor 1000 corresponds to the laser repetition rate (1 kHz), and T represents the effective pulse duration. This integration yields the spatial distribution of the average emission current density $\bar{J}(x,y)$ across the emission apex of the DWCNT emitter under different polarization conditions, as shown in Fig. S9.

Furthermore, by integrating the current density obtained from Eq. (S11) over the emission area, one can derive the time-dependent OFE current $I_{\text{OFE}}(t)$, which characterizes the temporal evolution of the emission current within a single optical cycle:

$$I_{\text{OFE}}(t) = \iint_0^S \bar{J}(x,y) dx dy \#(S12)$$

As shown in Fig. 5(d), within the duration of a single femtosecond laser pulse (~ 35 fs), the calculated $I_{\text{OFE}}(t)$ exhibits a decreasing trend with increasing polarization angle from 0° to 90° . The maximum OFE current occurs at 0° polarization, while the minimum is observed at 90° polarization. The results for the remaining three quadrants (90° – 360°) exhibit high symmetry with those in the 0° – 90° range, consistent with the structural and optical symmetry of the DWCNT emitter.

Finally, by integrating $I_{\text{OFE}}(t)$ over time and incorporating the laser pulse duration and repetition rate (~ 1 kHz), the average OFE current $\bar{I}$ from the DWCNT emitter under the OFE regime was calculated. The theoretical $\bar{I}$ values as a function of polarization angle show excellent agreement with the experimental measurements (see the red curve in Fig. 5c). This strong consistency validates the accuracy of the developed OFE current model for the DWCNT cold-cathode.

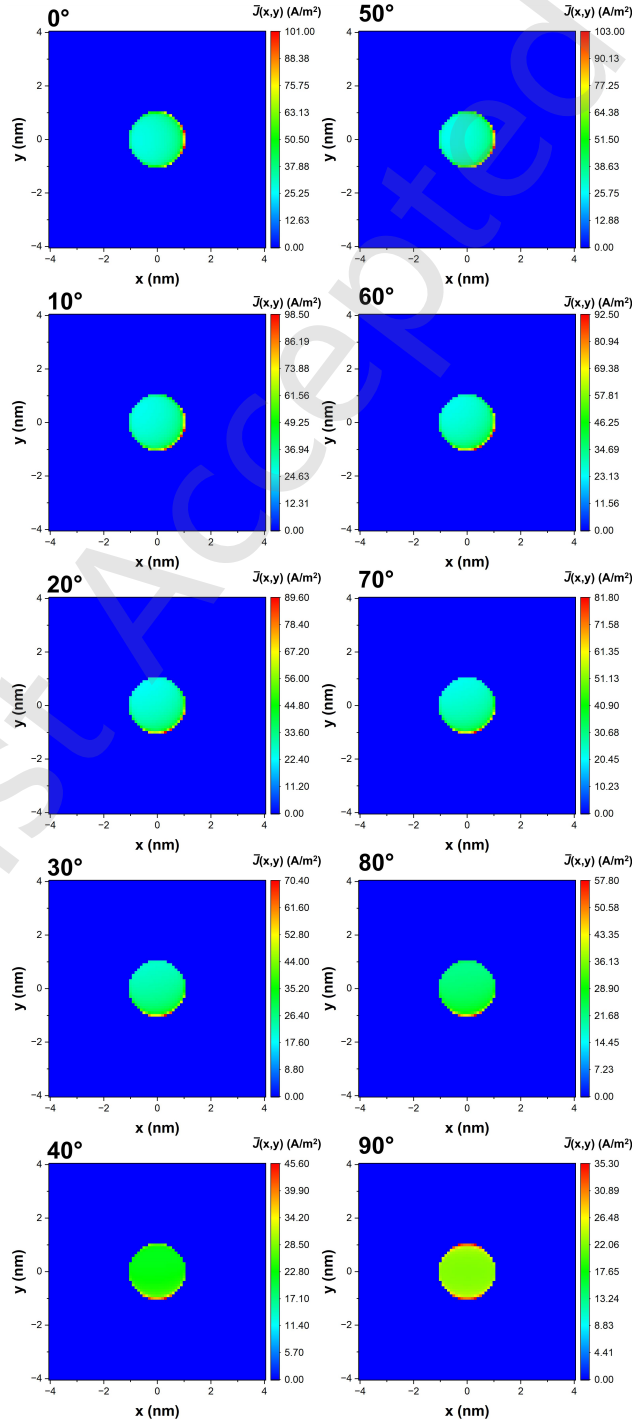


Fig. S9. Simulated spatial distribution of the average current density at the emission apex of the DWCNT-on-W tip emitter in the XOY plane.

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Reference:

[S1] Shen, Y.;Ding, Y.;Chen, Y.;Song, Z.;Cui, T.;Cheng, A.;Tang, S.;Cao, Z.;Ming, F.;Zhang, Y.;Chen, H.;Xu, N.; Deng, S. Nano-cold-cathode electron source based on plasmon-mediated emission. *ACS Photonics* **2024**, *11*, 3878-3889.

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