

Energy offset between femtosecond and thermal electrons in nanometer field-emitter tips

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We investigate the ultrafast photoelectric effect at sharp tungsten needles and find a considerable energy offset between thermally emitted and photoemitted electrons. Neither changes in temperature nor tuning the work function by the Schottky effect strongly affect the free-space energy of the photoelectrons. We explain these discoveries by a direct optical excitation within the band structure of tungsten, followed by scatter-free transport to the surface and emission into free space. Experiments at different photon energies support that the material and not the photon energy determines the primary properties of the electron beam.

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I. INTRODUCTION

The photoelectric effect [1] is the emission of electrons out of a material by photons of ultraviolet or visible light. The kinetic energy of the electrons and if electrons can be emitted at all, is determined by the difference between the photon energy and the work function of the material. This effect is one of the strongest proofs of the quantum nature of light and is also relevant for many practical applications in physics and chemistry. Examples include photomultipliers, photoemission spectroscopy, and electron beam generation in particle accelerators or ultrafast electron microscopes. To provide beams of the highest current density at optimum photoemission efficiencies, most modern laser-driven electron sources have a nanometer size and apply rather high photon energies, in particular when femtosecond emission shall be achieved [2–4]. At such photon energies, higher than the emitter’s work function, a large range of final electron energies is in reach, and the question arises what physics determines the emission efficiency, the electron energy spectrum, and its bandwidth. Are electrons directly emitted into free space on all the energetically allowed transition paths, or do they scatter from other electrons or phonons while propagating from the inside to the surface of the nanoscale emitter material? In perfect crystals with clean surfaces, as used in angular-resolved photoemission spectroscopy, the emission is direct [5]. However, in high-power emitters, such

as amorphous flat photocathodes [6–8] or nanometer-sized high-brightness emitters in electron microscopes [2–4], the emission is thought to be influenced by the properties of the photons and by scattering effects [6,8–11]. A photon energy close to the work function is then essential for minimizing the spectral and temporal broadening of the emitted femtosecond beam [8]. In photoassisted cold-field emission [3,12–14] or all-optical field emission by single-cycle laser light [15], electrons are expected to tunnel out into free space at energies below the work function. In all cases, the finite energy spread of the photoemission process [8] and the resulting limited coherence in time [16] are detrimental for coherent near-field imaging [17], attosecond electron microscopy [18,19], extreme pulse compression [20], or quantum operations with the free electron [21–23].

II. EXPERIMENT

Here, we investigate the ultrafast photoelectric effect in a single-crystalline tungsten needle tip, one of the most efficient and brightest electron sources for beam formation in time-resolved electron microscopy [2–4]. We perform both thermal emission and femtosecond laser emission simultaneously in the same tip and compare the spectral properties of the two emitted beams. Figure 1 depicts the experiment. We guide femtosecond laser light onto the tungsten needle emitter of an ultrafast electron microscope (JEM F200, JEOL). The tip radius is 500 nm and a thin layer of liquid zirconium oxide (ZrO) with a thickness of less than a few nanometers reduces the work function from approximately 4.8 eV [24] down to ~ 2.7 eV via dipole effects [25–27]. The tip is placed in ultrahigh vacuum (10^{-8} Pa) and an electric resistor circuit (filament current) controls its temperature. An electric field of up to several GV/m [28,29], caused by nanoscale field enhancement,

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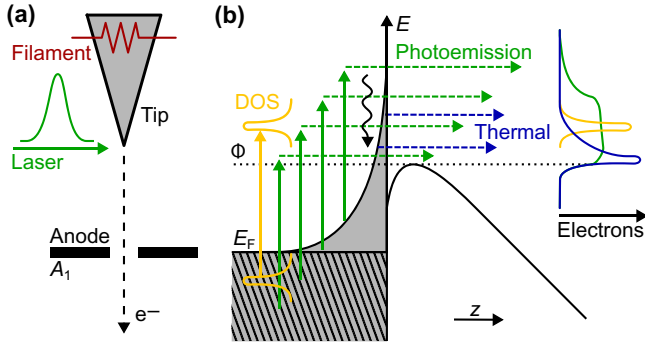


FIG. 1. Thermal and photoelectric emission at a nanometer tungsten tip. (a) Sketch of the experiment. Short laser pulses (green) with an energy of 2.4 eV emit photoelectrons. At the same time, a filament current (red) heats the tip. The strong electric field of the anode acceleration voltage A_1 allows for the emission of thermal electrons. Both beams travel downward in the electron microscope as an almost collimated beam (dashed line). (b) Energy diagram of several possible emission paths. Thermal electrons originate from a small tail of the Fermi-Dirac distribution (gray) above the work function Φ . The spectrum is, therefore, narrow and centered around Φ (inset, blue). In contrast, photoelectrons driven by the laser photons (green arrows) can be excited from a wide range of initial states above and below the Fermi level E_F . The final spectrum (inset, green) can, therefore, have a much broader energy distribution. If photoelectrons scatter from defects or other electrons in the material before release (black curved arrow), the final photoelectron spectrum would become thermal-like. Here, we report a third possibility, the emission from a well-defined donor into narrow acceptor states (yellow).

accelerates emitted electrons from the tip at an effective work function that is further reduced by the Schottky effect. Cold-field emission is negligible. In addition to continuous electron emission, we also use frequency-doubled femtosecond laser pulses (green) from an Yb:YAG laser system (Carbide, Light Conversion) with a wavelength of 515 nm, a pulse duration of ~ 250 fs, a bandwidth of ~ 20 meV, and a photon energy of $E_{ph} = 2.4$ eV to extract electrons via the photoelectric effect. The microscope's acceleration system subsequently speeds up both kinds of electrons to a kinetic energy of 80 keV. Afterward, a magnetic electron energy spectrometer (CEOS) is used to measure final electron energies.

Figure 1(b) depicts the energy levels that are relevant for our two emission mechanisms. The gray area shows the available electron energies in the material. In thermal emission, electrons can only be emitted into free space if they have energies higher than the effective work function Φ , defined by the ZrO layer and the Schottky effect (dashed line). Without laser excitation, an elevated emitter temperature and thereby a broadened Fermi-Dirac distribution are needed to reach energies above Φ and emit thermal electrons (blue). The energy spectrum (inset on

the right-hand side) is, therefore, centered at Φ . Slightly above Φ , there is the tail of the Fermi-Dirac distribution; slightly below Φ , there are small contributions from tunneling effects. In contrast, laser photons at $E_{ph} = 2.4$ eV (green) can emit from a large range of bound states into a large range of free states and the final spectrum can, therefore, be very broad [8–11]. In principle, any final energy between the work function and the work function plus the photon energy is reachable. The spectrum, therefore, would range from Φ to $\Phi + E_{ph}$. Evidence for this effect is the measured improvement of electron coherence when the photon energy approaches the work function in flat gold emitter geometries [6,8]. Some researchers also argue that electrons scatter with other electrons or crystal defects while propagating through the surface to the outside of the emitter material [9–11]. In this case, the photoelectron spectrum narrows to a hot electron gas with a center close to Φ . Evidence for such an indirect emission mechanism in amorphous metal emitters originates from comparisons of measured emission currents with theory [9–11]. Here, we discover a third mechanism, a well-defined emission from donor into acceptor states (yellow).

In our experiment, we emit thermal and femtosecond electrons at the same time and compare their spectrum and emission characteristics in a single experiment. We first set the temperature to just emit a finite but low current of thermal electrons, less than 10 pA or $\sim 10^8$ electrons per second. Then, we adjust the femtosecond laser power, at a repetition rate of 2 MHz, to produce ~ 100 electrons per pulse at the tip, to achieve a similar average current as in the continuous beam. We do not use multiphoton absorption [6,30] or all-optical field emission [15], and the number of emitted electrons scales linearly with the laser pulse energy. After the beam-selecting condenser-lens aperture of the electron microscope, we arrive at ~ 0.5 electrons per pulse, a good setting for ultrafast experiments [4]. An event-based, laser-triggered single-electron detector with nanosecond time resolution (Timepix3, ASI), together with a real-time clustering algorithm [31], allows us to separate the continuously arriving thermal electrons from the photoemitted electrons, which arrive at a well-defined time after the laser hits the tungsten tip. The time window is 5 ns and only about 0.1% of the femtosecond electrons are misclassified as thermal electrons.

III. RESULTS

Figure 2(a) shows the measured electron energy spectrum of the combined beam (black), the thermal electrons (blue), and the femtosecond photoelectrons (green). As expected, the thermal electrons (blue) undergo a sharp rise at the work function, followed by a longer tail due to the Fermi-Dirac distribution of the hot emitter material. The spectral width is 625 meV, typical for an electron beam

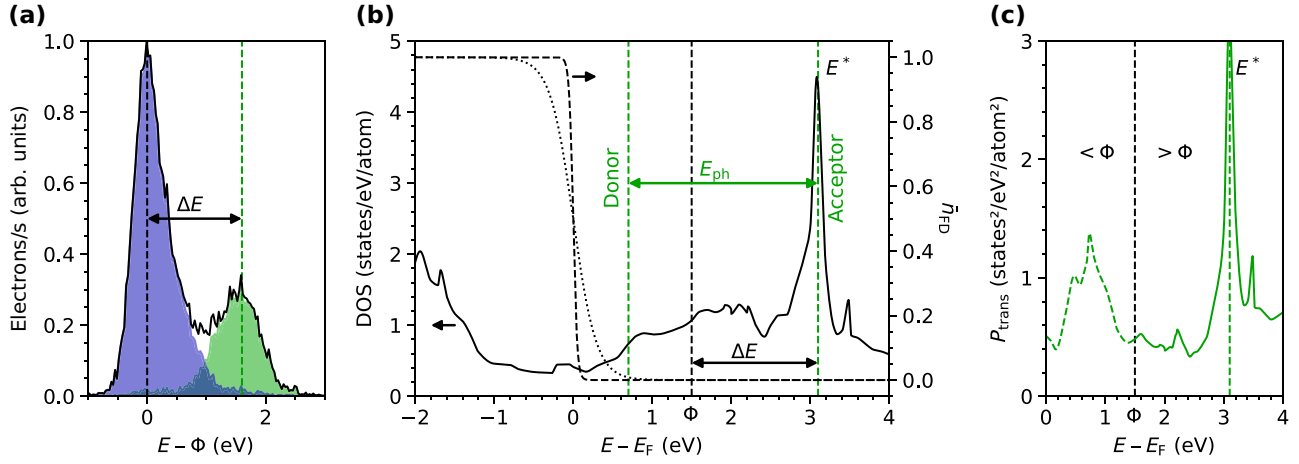


FIG. 2. Electron spectra and density of states in tungsten. (a) Different times of arrival allow for sorting thermally emitted electrons (blue) and photoemitted ones (green). We see a considerable difference in energy ΔE between the two spectra. (b) Density of state of tungsten (solid black, taken from Ref. [32]). We see a pseudogap around the Fermi energy and a distinct peak at an energy of $E^* = 3.1$ eV. In equilibrium, these states are occupied according to the Fermi-Dirac distribution $\bar{n}_{\text{FD}}(E)$ drawn for room temperature (dashed black) and a typical tip temperature of 1700 K (dotted black). The thermal electrons are emitted from slightly above the work function Φ , while the photoelectrons are excited from the donor into the acceptor band and then emitted at a fixed energy $\Phi + \Delta E$. (c) Transition probability $P_{\text{trans}} = \text{DOS}(E) \cdot \text{DOS}(E - 2.4 \text{ eV})$ to excite electrons to an energy E . Above Φ , there is only the distinct peak at the acceptor band situated at E^* .

from a Schottky field-emitter source. However, the photoelectrons from laser emission (green) show a narrow peak at an elevated energy of $\Delta E = 2$ eV above the work function Φ (green dashed line). The bandwidth of the femtosecond electrons is 890 meV, but it reduces to less than 460 meV when we avoid space charge effects (see below). We do not observe directly emitted photoelectrons with a broad spectrum between Φ and $\Phi + E_{\text{ph}}$ [see Fig. 1(b)], and also no indirectly emitted photoelectrons with a lowered energy toward Φ . Therefore, neither a laser-assisted field emission process [12,13] nor a direct photoemission [6,8] or even an indirect emission with scattering processes [8–11] can explain our measurements.

To interpret the measured results, we consider the band structure of our emitter material. Figure 2(b) (black line) shows the density of states of tungsten, obtained from first-principles electronic structure calculations [32]. We see a broad distribution of states with a pseudogap [33] around the Fermi energy E_F . Such a general structure is typical for a transition metal with an s-band, p-band, and intermediate d-band electrons, but tungsten also has a sharp peak at an energy of $E^* = 3.1$ eV above the Fermi energy. At finite temperatures, all the states are filled according to Fermi-Dirac statistics. The dashed and dotted curves in Fig. 2(b) show examples for temperatures of 300 and 1700 K, respectively. In thermal emission, some electrons reach energies above Φ (dashed vertical line) and are emitted into free space. The energy spectrum of thermal electrons is, therefore, located just above Φ . In laser emission, the optical penetration depth is ~ 15 nm [34] and the ZrO layer is sufficiently thin [27] to not absorb

a significant amount of laser light. Inside the tungsten needle, photon absorption can transfer electrons from almost any initially occupied state of the bulk material (here, called the donor states) into almost any unoccupied higher-energy state (here, called the acceptor states), provided that the energy difference is E_{ph} . We here neglect spin effects and potential selection rules. The probability P_{trans} of such a transition can be estimated by multiplying the density of states at the acceptor energy E by the density of states at the donor energy $E - E_{\text{ph}}$. All energies E are here given with respect to the Fermi energy E_F . The result is depicted in Fig. 2(c); the green curve denotes the estimated likelihood that electrons arrive at a final energy E above E_F . We see that the transition probability is highest for final electron energies at tungsten's special peak. There is also a high transition probability at ~ 0.8 eV (dashed green line), but final states below the effective work function (black dashed line) cannot be emitted into free space.

Based on these considerations, we argue that the absorption of a laser photon in tungsten first creates a rather narrow density of states around E^* . The donor states need to be thermally populated for this transition, but the laser is strong and only very few thermally excited states are needed to produce the measured tens to hundreds of electrons per laser pulse. In turn, the excited electrons are directly, without internal scattering and reduction of energy, emitted into free space [35]. The measured energy difference in the experiment is then simply $\Delta E = E^* - \Phi$. Based on this interpretation, the energy bandwidth of the photoelectrons (prior to space charge effects) is determined by the width of the acceptor band (~ 140 meV) and not

by the difference between photon energy and work function, as previously assumed [8]. The needle's ZrO layer does not cause substantial thermalization effects that could change the energetic properties and bandwidth of the femtosecond beam because the photoexcited electrons have a kinetic energy of ~ 2.7 eV and a typical velocity of $\sim 10^6$ m/s. They pass through the thin layer of ZrO in a few-femtosecond time, shorter than needed for substantial scattering [8–11]. We conclude from these considerations that the ZrO layer only reduces the effective work function via its dipole moment [27] and does not affect the effective band structure of the emitter material in a substantial way. Therefore, we argue that the emission from nanometer tungsten needles into free space is direct: statistical scattering effects are weak and the band structure of tungsten is decisive for the observed energy offsets and the surprisingly small spectral width of the measured photoelectrons.

To further verify these hypotheses, we measure the photoelectron spectra as a function of the acceleration voltage A_1 and the temperature of the tip. Due to the vast changes in the thermal emission current with both parameters, eventually saturating our detector electronics, it is not possible to make independent measurements. Therefore, we first set the voltage A_1 to a desired value and then adjust the filament current and thereby the emitter temperature to return to roughly the same current of the thermal electrons in the measurable range (i.e., 1.5×10^4 electrons per second). Finally, we adjust the laser power P_{Laser} to obtain roughly the same number of photoelectrons (i.e., 1.5×10^5 electrons per second at the detector or ~ 100 electrons per pulse at the tip), as well. In this regime of a high emission peak current, the width of the photoelectron spectrum is dominated by space charge effects [4,28,36,37].

Figure 3 depicts the results. We first measure the electron energy spectrum as a function of the acceleration

voltage A_1 . This changes the work function by the Schottky effect. After each change, we wait for ~ 15 min for the experiment to stabilize. Our electron energy spectrometer is not stable in absolute scales over such extended measurement times and we, therefore, use the thermal electrons (black dashed line) emitted at $\Phi(A_1)$ as a reference and plot the photoelectron spectra as a function of $E - \Phi(A_1)$. Contributions from tunneling electrons below $\Phi(A_1)$ may slightly shift the reference peak but this effect is approximately constant and also small [26,29]. Figure 3(a) shows the results. Here, we see an increasing offset between the thermal electrons and photoelectrons with an increasing A_1 (from blue to green). This result fits with our earlier assumptions: when we increase the extraction voltage, we reduce the work function, and laser-excited electrons initially created at a fixed energy of E^* inside of tungsten are emitted at an increasing energetic distance to the thermal electrons at Φ . Figure 3(b) shows the measured shift of the photoemission electron energy with the acceleration voltage. Here, we approximately see the expected square-root dependence of the Schottky effect (dashed curve). The characteristic value of the shift is 1–2 eV, which is typical for modern electron emitter geometries with nanometer tips [29]. However, the position of the origin of the relevant parameters appears to be shifted in absolute voltage and absolute energy (dashed lines). These offsets originate from the peculiarities of our electron emission geometry. Namely, in the electron gun of our microscope, there is an additional suppressor anode at a voltage of -0.3 kV placed about 0.25 mm behind the tip, intended to pull stray electrons from the beam. A physical electric field of zero at the tip, therefore, corresponds to an extraction voltage A_1 of 1–2 kV (dashed vertical line). In the energy axis, there is an offset of ~ 0.4 eV (dashed horizontal line). When the field at the tip is zero (suppressor ≈ -0.3 kV and $A_1 \approx 1.6$ kV), there is no Schottky effect. Therefore, the effective work

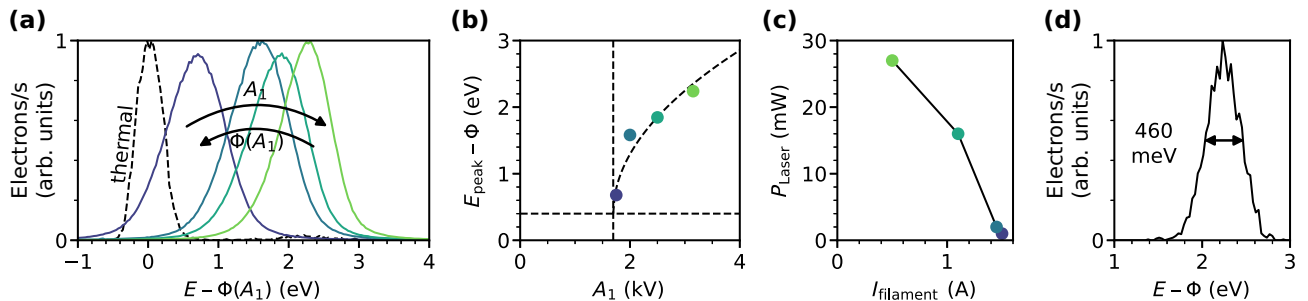


FIG. 3. Dependence on acceleration voltage A_1 and tip temperature. (a) Emission spectra with A_1 increasing from blue to green (1.75 kV, 2 kV, 2.5 kV, and 3.15 kV) and decreasing filament current (1.5 A, 1.45 A, 1.1 A, and 0.5 A). Potential long-term drifts of the energy spectrometer are calibrated by setting the thermal electrons (dashed line) to Φ . (b) Dependence of the measured energy offset on the acceleration voltage. We observe higher differences for higher acceleration voltages with a square-root dependency. (c) Measured laser power that is needed to obtain the same number of photoelectrons as a function of filament temperature. With increasing filament current [see Fig. 1(a)], the emission efficiency increases by more than one order of magnitude. (d) Measured spectrum of the photoelectron peak for lower laser power and less space charge effects. The width is 460 meV (full width at half maximum).

function Φ approaches the literature value of ZrO-covered tungsten of ~ 2.7 eV [25], leading to the observed value $\Delta E = E^* - 2.7$ eV ≈ 0.4 eV. The ability to explain the measured energetic properties of our laser-emitted photoelectrons [Fig. 3(a)] with the physics of the Schottky effect [Fig. 3(b)] supports the decisive role of a constant internal electron energy of $E^* = 3.1$ eV within tungsten, which is independent of temperature or acceleration field. In other words, our photoelectrons are always emitted into free space at the same energy with no substantial dependency on temperature or the Schottky effect.

We next investigate the role of temperature. Figure 3(c) shows the measured efficiency of the laser-driven emission process at different emitter heating currents I_{filament} , that is, how much laser power is needed to emit the same number of electrons. We observe that a higher needle temperature (i.e., a larger filament current) requires much less laser power than a cold emitter tip. The photon-to-electron conversion efficiency decreases by a factor of 30 when the heating current is reduced from 1.5 to 0.5 A. At our laser photon energy of 2.4 eV, we argue that the special state in tungsten at E^* can only be reached from already thermally excited electrons at the donor band above the Fermi energy [compare with Fig. 2(a)]. At higher temperatures, more electrons are available for the laser emission process and less total optical pulse energy is, therefore, required for creating the same average number of electrons. However, the spectral offset and the bandwidth stay approximately the same.

Important for many applications in electron microscopy is the bandwidth of the photoelectrons because it determines the spectral resolution of electron energy loss spectroscopy and the ability of the electron matter waves to interfere in time [16]. Figure 3(d) shows a low-current measurement of the photoelectrons in which we avoid space charge effects [4,8,36,37]. We see a bandwidth of 460 meV, which is probably limited by an inhomogeneous Schottky effect or the resolution of our energy spectrometer. Although the laser wavelength in our experiment has not been adjusted to the work function of the emitter material [6,8], we create a rather monochromatic beam that is useful for spectroscopy or interference experiments. If we avoid inhomogeneous Schottky effects from round surfaces or other imperfections in the experiment, we may expect a bandwidth of ~ 140 meV (see Fig. 2).

Figure 4 shows further support for our suggested theory. Figure 4(a) depicts a measured emission spectrum at a very high tip temperature (filament current 1.6 A). As expected, we see that thermal electrons (blue) can now, in part, reach the acceptor band even without laser light. A new thermal peak at ~ 1.5 eV appears at the energy of the optically emitted electrons (green). Figure 4(b) shows a comparison between thermal electrons (blue) and laser-emitted electrons at a photon energy of 3.6 eV (the third harmonic of our laser pulses). As expected, the photoelectrons appear at

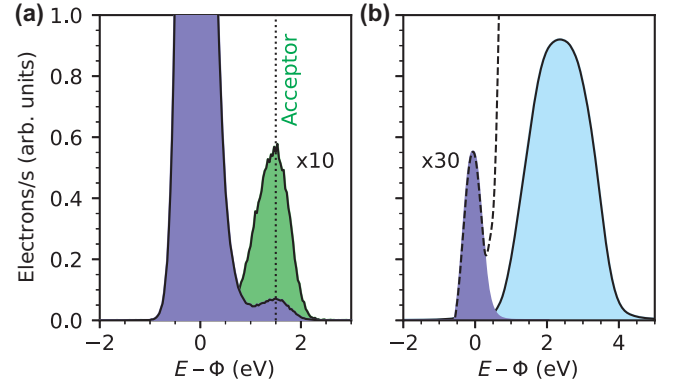


FIG. 4. Thermal emission at very high temperature and photoemission at 3.6 eV. (a) Measured emission spectra of thermal (blue) and photoelectrons (green) at elevated tip temperature (filament current 1.6 A). The thermal electrons show a hump at the same energy as the peak energy of the photoelectrons (dotted line). (b) Measured emission spectrum (dashed line) of thermal electrons (blue) and photoelectrons (light blue) when using a photon energy of 3.6 eV. As expected, we see two distinct peaks.

similar energy levels as with emission at 2.4 eV (compare Fig. 3), and we again observe a clear energy separation between the thermal electrons (blue) and photoemitted electrons (light blue).

IV. DISCUSSION

In summary, femtosecond photoelectric emission from a tungsten needle tip creates free-space electrons at specific energies far above the work function of the emitter material. Photoelectric emission from a sharp needle tip is, therefore, neither an indirect emission process with scattering events [8–11] nor a direct transition from bound states into free space [12,13]. Rather, the absorption of a photon first creates a preferred excited state within the material that is then directly and without scattering emitted into free space. Therefore, the spectrum and the spectral width of the emitted electron beam are dominated by the band structure of the emitter material, while the properties of the laser light or the temperature of the source only determine the number of emitted electrons. Moreover, the surface effects from amorphous interfaces or thin liquid coatings are not substantial for the bandwidth and coherence of the beam. This is because the reported energy offset implies a large electron velocity that allows it to pass such imperfections in a short enough time to avoid scattering phenomena. Therefore, the properties and band structure of the basic emitter material are relevant for the resulting electron pulse characteristics such as central energy, energy bandwidth, temporal coherence, or potential entanglement.

Besides these general insights, the reported results have also practical implications for ultrafast electron microscopy [2,4,38] and investigations of ultrafast dynamics of matter with atomic resolution in space and time.

Even in the complete absence of space charge effects [8], short single-electron pulses so far have a time-energy product that is usually much larger than allowed by the uncertainty principle [39]. Compression to a few femtoseconds [4] or attoseconds duration [18,19] therefore implies a substantial broadening of the spectral width to several electronvolts, which is detrimental for spectroscopy or high-resolution near-field microscopy [40]. However, the mechanisms reported here indicate that the selection of a nanometer-sized emitter material with a narrow transition that matches the bandwidth of the driving laser light (but not necessarily its center wavelength) can help to create ultrashort electron pulses at an uncertainty-limited product of time and energy. The 140-meV band of tungsten (full width at half maximum) indicates that laser pulses with a duration of ~ 15 fs (full width at half maximum) can be applied. The resulting near-monochromatic but still ultrashort electron pulses will be beneficial for ultrafast electron energy loss spectroscopy [41], chiral electron microscopy [42], nonclassical electron beams [43], or free-electron qubit operations [21,23] at the highest coherence in space and time.

Note added. Recent data on cold field emitter tips [44] align well to our reported findings.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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