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Exploring temporal and rate limits of laser-induced electron emission

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Abstract
To achieve high temporal resolution for ultrafast electron diffraction, Zewail (*Proc. Natl Acad. Sci. USA* **102**, 7069 (2005)) has proposed to use high repetition rate, ultrafast electron sources. Such electron sources emitting one electron per pulse eliminate Coulomb broadening. High repetition rates are necessary to achieve reasonable data acquisition times. We report laser-induced emission from a nanometre-sized tip at one electron per pulse with a 1 kHz repetition rate in the femtosecond regime. This source, combined with 1 MHz repetition rate lasers that are becoming available, will be a primary candidate for next generation ultrafast, high-coherence electron diffraction experiments. We also report that the measured energy bandwidth of our electron source does not support sub-cycle electron emission. This result addresses a current debate on ultrafast nanotip sources. Regardless of the limited bandwidth, this source may be used in conjunction with a recently proposed active dispersion compensation technique (*Proc. Natl Acad. Sci. USA* **104**, 18409 (2007)) to deliver attosecond electron pulses on a target.

(Some figures in this article are in colour only in the electronic version)

Recently, laser-induced electron emission from nanometre-sized tips has been investigated [1–4] for the purpose of generating ultrashort electron pulses. These sources promise to affect a wide variety of technologies. They can add a temporal dimension to scanning tunnelling microscopy [5]. By replacing the usual surface sources with nanometre-sized sources, the transverse coherence can be increased for ultrafast electron diffraction (UED) [6–8] and ultrafast electron microscopy (UEM) [9–11]. Nanometre-sized sources may also be combined with recently proposed dispersion techniques that promise to deliver attosecond electron pulses at a target [12]. Additionally, the high timing resolution of these sources can be used for time-of-flight techniques [13] and has already impacted fundamental physics studies such as the Aharonov–Bohm effect [14, 15]. Finally, pulsed emission fills phase space more than continuous emission with the same average current. The expected increase in quantum degeneracy is promising for further experiments in the emerging field of electron quantum optics [16–18].

Other techniques have been developed [9, 10, 19–23] and proposed [23] to create electron pulses with varying advantages and disadvantages. For example, ultrashort electron pulses have been extracted by focusing an amplified laser pulse on a surface to induce electron emission [9, 10, 19–21]. Although this has been a highly successful source, the pulse duration is broadened by space charge [21, 24], while the spatial extent of the source limits the electron beam coherence.

Fibre amplified laser systems are becoming available that offer 100 kHz to 2 MHz repetition rates combined with 10–100 μJ pulse energy [25, 26]. Such laser systems in conjunction with a nanometre-sized tip offer a promise to overcome the aforementioned problems. The high repetition rate permits a reduction to one electron per pulse, which removes Coulomb broadening [27]. The small source size increases the spatial coherence [28]. While low energy, high repetition rate laser pulses can be used to induce electron

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pulses, higher energy laser pulses are needed to trigger molecular processes or solid-state processes to be studied by UED or other pump–probe techniques. Until now, tip sources have not been tested at one electron per pulse. In this paper we study the emission process from a $\sim$100 nm-sized tip and find that the tip is robust at rates exceeding one electron per pulse and emits in the femtosecond range.

The process responsible for producing the electron pulses is under debate. Initial work claimed that field emission [1] was the primary mechanism for emission. A theoretical model based on field emission predicts sub-cycle electron emission [2]. Experiments [1, 3] showed that the emission process depends on the projection of the laser electric field on the tip direction. This leads to the idea that the electron emission promptly follows the laser electric field oscillations. This idea, combined with the observed high non-linearity of the emission process, suggests that the temporal response would be pushed into the attosecond domain. This leads to the exciting possibility of having macroscopic, attosecond emitters. More recent work puts forth a theoretical model that matches previously presented experimental results with a non-equilibrium model that does not require field emission and predicts much slower emission [29]. We present experimental results that determine the lower bound for the duration of the emission process.

As demanded by Heisenberg's uncertainty principle, an electron pulse with bandwidth ΔE must have a pulse duration of at least $\Delta t \sim \hbar/(2\Delta E)$. A time-of-flight measurement was performed to determine ΔE [13]. We show that this places a lower bound of about 5 fs on the electron pulse duration. Using the relation between energy and time is not without precedent: Lindner *et al* performed a beautiful temporal double-slit experiment which uses this same connection [30]. The lower bound of 5 fs shows that the emission of electrons from the tip does not promptly follow the oscillations of the electric laser field for our experimental configuration.

The possibility of sub-cycle emission for this type of source is not necessarily eliminated. Sub-cycle emission is predicted for an 8 fs laser pulse [2]. The emission process for our 45 fs laser pulse may be different than that of an 8 fs laser pulse if electron–electron interactions in the tip occur at a timescale of tens of femtoseconds [31]. Additionally, different tip materials (different work functions and thermal conductivities), cold tips (less thermal effects) or single-atom tips (fewer interacting particles) could still provide sub-cycle emission. Future experimental investigations of laser-induced electron emission from such tips are needed.

Our experimental setup has been described in detail previously [3]. A femtosecond laser beam is tightly focused on a nanometre-sized tip. The 100 MHz femtosecond oscillator is replaced by a 1 kHz amplified laser (Spectra Physics Spitfire) to test the tip at its damage threshold while limiting the electron detector exposure. The tip is mounted on a three-dimensional translational stage to position it within the laser focus. The emission rate is measured for different tip locations in the focus. The data shown in figure 1 (inset) show a 20 μm peak of maximum emission centred at the laser focus. As the tip is moved past the laser focus, the laser light hits the
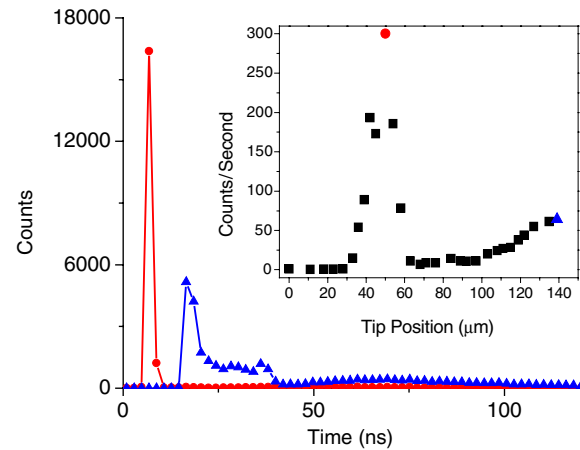

Figure 1. Position dependence on emission characteristics. The position dependence of the tip with respect to the laser focus has a peak of maximum emission when the apex is in the focus (inset). The emission rate increases again when the tip's shank is in the focus. The temporal spectra for different tip locations within the focus show that different processes are present at these locations. When the apex is in the focus (circles), the emission is temporally sharp. When the laser hits the tip shank (triangles), the emission is temporally broad. All of the data that follows are taken with the apex in the focus.

tip shank, causing a second increase in the emission rate. The temporal spectrum taken when the laser hits the apex of the tip has a distinct signature as compared to when the laser hits the shank. The temporal spectrum of the electron emission when the apex is in the focus is sharp (figure 1, circles). At this tip location, the polarization dependence shows maximum emission when the polarization is parallel to the tip. These features coincide with the field emission [2] and non-equilibrium models [29]. When the tip is positioned so that the focus hits the shank, the temporal spectrum shows a much broader peak. The polarization dependence shows a maximum when the polarization is perpendicular to the tip. These are features associated with thermal processes [32]. For all the measurements that follow, we took care to hit the tip at its apex rather than at the shank.

To characterize the fast emission process when the apex is in the focus, we try to determine a lower bound for the emission time. For an emission process with an energy bandwidth ΔE, Heisenberg's uncertainty principle demands a lower bound, Δt, of $\Delta t \sim \hbar/(2\Delta E)$. If, for our source, the electron emission would follow the electric field, the electron pulse duration would be about $\Delta t_{\text{FWHM}} < 0.66$ fs $= (\lambda/4)/c$. Through Heisenberg's uncertainty principle, this corresponds to $\Delta E_{\text{FWHM}} = 2.7$ eV.

The energy bandwidth of our source is obtained by fitting to time-of-flight spectra (an example spectrum is available in the inset of figure 2). Note that the experimental electron pulse lies on a background. This background is constant throughout the spectrum and the polarization dependence of the background is consistent with thermal emission. This suggests that there are no significant sub-cycle emission processes being masked by our dominant peak. The temporal pulse width is plotted as a function of electron energy in
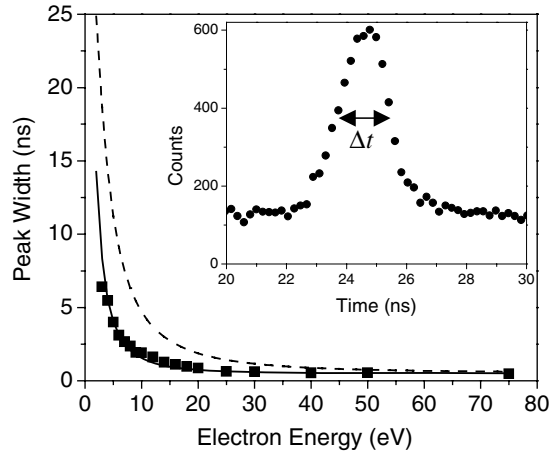
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Figure 2. Determining the lower bound on the emission process. From temporal spectra, the widths of the electron pulses after traversing a time-of-flight energy analyser are determined. The inset shows an example spectrum for 10 eV. Experimental findings (squares) from [13] are plotted along with curves from a simulation that assumes an energy distribution with a 0.8 eV width (solid curve). Simulation results with a width of 2.7 eV (corresponding to a sub-cycle emission process) are also plotted (dashed curve). The latter curve does not match our experimental data, ruling out sub-cycle emission.

Figure 3. Power dependence on electron emission rate. The triangles represent data taken with the laser oscillator, presented in [3] with a −300 V dc voltage on the tip. The circles represent new data taken with the amplified laser beam with a −220 V dc voltage on the tip. Different tips were used for each of the data sets. Both sets are fitted with a P^3 curve (solid lines). The two data sets would overlap with a straight line if one data set were given a horizontal shift of a factor of 10. This shift is attributed to different laser foci. These data show that a four-order-of-magnitude increase in emission per laser pulse can be achieved with similar emission process characteristics.

figure 2. Details for this energy analyser are found in [13]. The experimental data are fitted with curves from a simulation that calculates the arrival time for electrons travelling through the drift tube. The temporal pulse width at the detector (the *y*-axis in figure 2) is found by determining the arrival time for each electron in a Gaussian energy distribution. The centre of the distribution is the electron energy (the *x*-axis in figure 2), while the only fitting parameter is the width of the energy distribution.

The dashed curve in figure 2 is a simulation curve with an energy bandwidth of 2.7 eV. The experimental data indicate that the electrons are emitted with a lesser energy bandwidth, and thus they are not emitted in a sub-cycle process. The experimental data fit well with an energy bandwidth of $\Delta E_{\text{FWHM}} = 0.8$ eV, which places a lower bound of about $\Delta t_{\text{FWHM}} = 2.3$ fs on our source. This is the lowest possible estimate, because a Gaussian distribution of emission energies was assumed. For an emission process following the sinusoidal electric field of the laser beam with an intensity-cubed dependence [1, 3], Heisenberg's uncertainty principle predicts a lowest duration time of about 5 fs.

Previous work done with our source showed the emission duration to be less than 100 fs, but this was tested for electron count rates less than 10^{-3} electrons per pulse [3]. At the full laser intensity, electron currents of about 0.1 nA (>1 electron per pulse) were observed with a Faraday cup and electrometer (Keithley 610C) combination. However, the electron pulse duration could not be measured with this slow detector. In this work, we performed a pump–probe experiment at higher laser intensities. Before presenting our pump–probe test for the pulse duration, the robustness and emission characteristics of the tip were to be tested at the higher laser intensities required to reach one electron per pulse. A Spectra Physics Spitfire
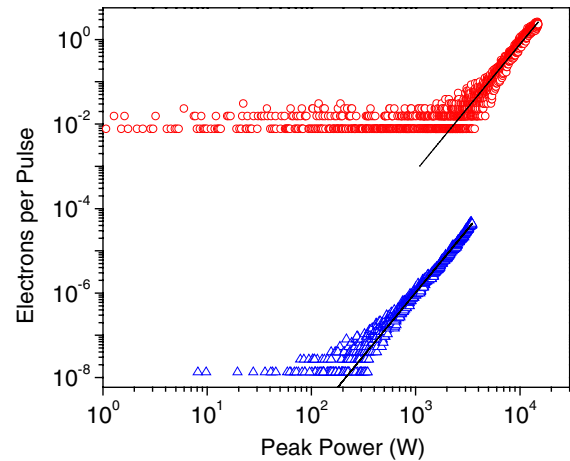
Ti:sapphire laser with a repetition rate of 1 kHz was used. This lower repetition rate allows emission of one electron per laser pulse without causing detector damage. A rate of one electron per pulse was observed. The solid angle acceptance of our detector captures nearly 10% of the emitted electrons. Thus, the source was emitting about ten electrons per pulse. This shows that the tip is robust for emission rates exceeding one electron per pulse.

Figure 3 shows the power dependence of the emission from the amplified pulse and compares this to the emission from the oscillator pulse published in [3]. Both laser pulses provide an electron yield with a power-cubed dependence. Additionally, the emission from the amplified beam is maximized when the laser beam polarization is parallel to the tip, identical to the oscillator emission. These are indications that the emission processes are similar. We note that the laser power necessary for ten electrons per pulse emission is very close to the damage threshold of the tip. Very small increases in laser power from this setting would reform the tip and cause order of magnitude variations in electron emission rate and change the power dependence.

The two data sets in figure 3 do not connect with one straight line. This may be explained by the difference in laser beam size in the two experiments. The diameter of the oscillator beam is a few millimetres while that of the amplifier is nearly a centimetre. Thus, the amplifier beam is expected to have a smaller focal waist (the same focusing mirror was used) and, for identical peak powers, a larger intensity. For an estimated ratio of two between the waist sizes, the data sets would nearly fall on a single line when plotted as a function of intensity.
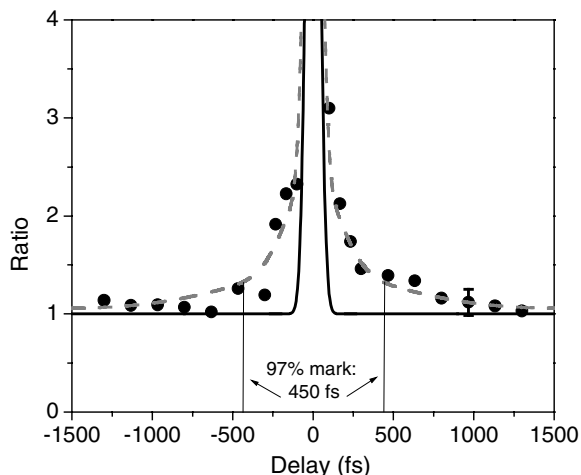
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Figure 4. Upper bound on emission duration with amplified beam. A pump and a controlled delay probe pulse are used to determine an upper bound on the emission time. If the delay is sufficient such that emission from the later pulse is not affected by the earlier pulse, then the emission time is less than the delay. To determine if the two pulses affect each other, a ratio of the emission from both pulses together to the sum of the emission from each pulse individually is measured. When this ratio is one, the emission time is less than the delay. A simulation (solid curve) predicts what the ratio would look like if the emission process were instantaneous with the laser field, assuming an I^3 dependence. The experimental data (circles), with a guide to the eye (dashed grey curve), show that the emission duration is in the femtosecond regime.

To determine the duration of the emission process, we use a pump and delayed probe pulse (for details on the method, see [3]). If the emission caused by the delayed pulse is not affected by the first pulse, the process is faster than the delay. In this case, the count rate when both pulses are present should equal the sum of the count rates caused by each pulse individually [3].

The count rate for both the pump and probe together, C_b, is integrated for 60 s. This is determined as a function of delay between the two pulses. The electron count rate is also measured for the pump and the probe each individually for the same time period, $C_{1,2}$. The ratio $C_b/(C_1 + C_2)$ is shown in figure 4 to reach one near 450 fs. Thus, at emission rates of one electron per pulse, electron pulse durations in the high femtosecond range are possible. We note that the delay that achieves a ratio of one varies from tip to tip. Ratios of one have been reached at 200 fs, but also at picoseconds. This may be due to the tip manufacturing method used [33]. Electron microscope images reveal tip radii variations of about a factor of two.

Figure 4 also compares the experimental ratios with a calculated ratio, where the electron emission is assumed to be instantaneous with the laser field. An I^3 dependence is used to determine the count rates. The laser chirp measured with a frequency resolved optical gating technique is taken into account. This best-case-scenario ratio reaches one at 150 fs—about two to ten times faster than experimental findings. The discrepancy could be due to laser heating or could indicate that the limits in emission rate and electron pulse duration are reached for the present source configuration.

In summary, this nanometre-sized source is ultrafast at emission rates of one electron per laser pulse and is a candidate for use in UED and UEM technology [34]. Additionally, the source could be combined with temporal lens techniques [12] to deliver attosecond electron pulses on a target. This electron source has already been used for fundamental physics studies in demonstrating the absence of forces for the Aharonov–Bohm effect [14], and it is anticipated that other fundamental electron experiments will be possible [15].

Acknowledgment

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