Ultrafast time resolved photo-electric emission

Thomas Juffmann, Brannon B. Klopf, Gunnar E. Skulason, Catherine Keañofer, Fan Xiao, Seth M. Foreman, and Mark A. Kasevich

1Physics Department, Stanford University, Stanford, California 94305, USA
2Department of Physics and Astronomy, University of San Francisco, 2130 Fulton St., San Francisco, CA 94117, USA

The emission times of laser-triggered electrons from a sharp tungsten tip are directly characterized under ultrafast, near-infrared laser excitation at intermediate Keldysh parameters $2.1 < \gamma < 4.8$. Prompt emission is observed at the highest field strengths. At lower field strengths, emission delays as large as 10 fs are observed. Emission time is inferred from the energy gain of photoelectrons directly emitted into a synchronously driven radio-frequency (RF) cavity. 1 fs timing resolution is achieved in a configuration capable of measuring timing shifts greater than 10 ps. The technique can also be used to measure the RF phase inside the cavity with a precision below 70 fs upon the energy resolved detection of a single electron.

Ultrafast laser excitation of metallic nanostructures has recently been exploited to generate triggered ultrafast electron sources [1–4]. These sources are finding increasing application in ultrafast laser microscopy and electron diffraction applications [5, 6] as well as in the characterization of ultrafast electronic [7] or optical [8] signals. They are further discussed as promising electron sources for laser-based electron accelerators [9, 10]. While the basic physical processes underlying ultrafast photo-excitation mechanisms have been understood for decades [11], a detailed understanding of these mechanisms and their interplay in technologically relevant systems requires clarification. Since below barrier emission (possibly thermally mediated) can in principle be substantially delayed with respect to the prompt processes, the timing resolution and capabilities of these sources is uncertain.

In this Letter, we directly measure the emission time for photo-excited electrons from sharp metal tips, in a technologically relevant regime of intermediate laser excitation strength (Keldysh parameter $2.1 < \gamma < 4.8$, < 0.1 electrons per laser pulse) and ~ 10 fs pulse duration for near-infrared (NIR) laser excitation. While recent experiments show that prompt electron emission processes contribute to the electron signal [11, 12, 13], below barrier field emission could still lead to a significant amount of delayed emission on a timescale of 10 fs to 1 ps [14, 15]. A recent 2-photon pump-probe experiment provides evidence for the process being prompt on the 100 fs time scale [3]. Attosecond extreme ultra-violet NIR streaking experiments [16, 17], while achieving excellent timing resolution, do not rule out delayed emission processes on these slower time scales.

Here, we directly characterize the electron emission time by placing the electron source in a microwave cavity, which shifts the electron energy depending on its emission time with respect to the phase of the cavity electric field. Synchronization of the cavity field phase with respect to the ultrafast laser pulse allows for fs resolution of the emission time with a timing dynamic range limited by the period of the microwave field ($\sim 100$ ps). This technique avoids interference effects between a pump and probe pulse as well as probe induced quiver motion and electron recollisions.

The setup is sketched in Fig. 1a: A titanium-sapphire laser provides 10 fs short laser pulses at a repetition rate of about 150 MHz. A half-wave plate and a linear polarizer are used to set the intensity of the laser pulses and to set the polarization parallel to the tip axis. A 25 mm focal length curved mirror focuses the laser onto a tungsten tip [W(111), tip radius $r \sim 400$ nm] with a beam waist of 6 $\mu$m ($1/e^2$ radius). The tungsten tip is placed inside a cylindrically symmetric re-entrant cavity. Fig. 1b shows the simulated longitudinal electric field of the TM$_{020}$ mode, which is homogeneous along the axis except for the local field enhancement close to the nanopip. The transverse field components of the TM$_{020}$ mode are zero on the symmetry axis. The cavity features an entry and an exit hole for the laser beam as well as an exit aperture for the electron pulses. The resonance of the cavity is at 9.08 GHz and we measure a Q-factor of up to 2500. Fig. 1c shows charged particle tracing simulations at a bias voltage $U_{DC} = 2.1$ kV. The electrons gain about 95% of their kinetic energy within the first 50 ps. The sensitivity of this cavity enhanced RF photo-electron streaking technique is maximal if the electrons exit the cavity after interaction times close to half-integer multiples of the RF period, which can be controlled by tuning $U_{DC}$.

In order to synchronize the RF fields inside the cavity with the laser pulses, we derive the RF signal directly from a fast photodiode (Hamamatsu G4176), which detects a pick-off beam of the laser (see Fig. 1a). The photodiode signal is split into a low frequency component at the 4th harmonic of the laser repetition rate and a high frequency component at the 60th harmonic of the laser repetition rate. The low frequency component is used as a clock for a direct digital synthesizer (DDS, Novatech 409B), which outputs a signal at the repetition rate of the laser with a digitally controllable phase. This phase...
shifted signal is single-sideband (SSB) mixed with the high frequency component to create a signal at the 61st harmonic, which is at the cavity resonance. It is amplified, filtered and fed into the cavity using an RF antenna.

After the laser triggered emission of electrons, the electrons are accelerated by the DC bias field and the RF field, both of which are enhanced close to the tip. Typically the tip is set to $U_{\text{tip}} = -35$ V with respect to ground and the cavity is floated at $U_{\text{DC}} + U_{\text{tip}}$. The interaction with the RF field leads to a dependence of the final kinetic energy of the electrons on the phase of the RF field at the moment the electrons leave the tip. After exitting the cavity the electrons are collimated using two lens electrodes and are decelerated towards a retarding field energy analyzer. It consists of five grids and yields a resolution of about 1.5 eV (FWHM). After the energy analyzer the electrons are accelerated and detected using a micro-channel plate (MCP) in chevron configuration. The amplified signal is accelerated onto a phosphor screen, which is imaged onto a CCD camera.

Fig. 2a shows the measured electron energy gain $E(\Phi)$ as a function of the RF phase $\Phi$. At 0.8 W of RF input power the final electron energy shows a peak-to-peak modulation of about 164 V at $U_{\text{DC}} = 2.1$ kV. At the zero crossing of this modulation curve, the slope is about 4.6 V/ps. Qualitatively similar results were obtained with an RF input power of 8 W, where the modulation was 275 V corresponding to a slope of 7.8 V/ps.

Repeated measurements of $E(\Phi = \Phi_0)$ at the zero crossing of the energy modulation curve at $U_{\text{DC}} = 2.1$ kV are shown in Fig. 2b. Correcting for a slow drift $< 4$ fs/min yields a standard deviation of 22 fs for a single measurement. To test for the resolution of cavity enhanced RF photoelectron streaking, the dataset is divided into two interleaved datasets (alternating blue and green data points) and Allan deviations for the differential quantity $\Delta E = E_{\text{blue}}(\Phi_0) - E_{\text{green}}(\Phi_0)$ are calculated and shown in Fig. 2c. It agrees well with the standard deviation of the sample mean and shows that a resolution on the order of 1 fs can be reached with sufficient averaging. For these measurements the RF chain before the final amplifier was simplified by removing the direct digital synthesizer and the single sideband mixer and by directly working with the 61st harmonic of the photodiode signal, leading to an increased long term stability of the system.

Fig. 2c highlights the scheme’s capability to detect fs level timing shifts. Every data point shows the average of 15 measurements of

$$\Delta E_{\Delta \Phi} = \frac{1}{2} \left[ \left( E(\Phi_0 + \Delta \Phi) - E(\Phi_0 + \Delta \Phi + 180^\circ) \right) \right. \left. - \left( E(\Phi_0) - E(\Phi_0 + 180^\circ) \right) \right].$$

$\Delta \Phi$ is controlled using the DDS with 14-bit resolution corresponding to 0.022$^\circ$ (384 $\mu$rad) or a timing shift of 6.722 fs. Measurements at both the positive and negative zero crossing further suppress possible errors from drifts in $E(\Phi_0)$, due to, for example, slow changes in the tip work function. The energy depends on phase linearly with a root mean square residual error of 3.9 fs.

In order to study electron emission delays, we look for a laser power dependent energy shift

$$\Delta E_{\Delta P} = \frac{1}{2} \left[ \left( E(\Phi_0, P) - E(\Phi_0 + 180^\circ, P) \right) \right. \left. - \left( E(\Phi_0, P_0) - E(\Phi_0 + 180^\circ, P_0) \right) \right],$$

which is proportional to a delay in photoemission ($P_0 = 250$ mW). Laser intensity dependent changes in the initial kinetic electron energy (e.g. due to the ponderomotive potential of the pump pulse or the temperature dependence of the electron energy distribution inside the
solid) do not contribute directly to $\Delta E_{\Delta P}$ in this differential measurement scheme involving measurements at $\Phi_0$ and at $\Phi_0 \pm 180^\circ$. Instead, they only contribute indirectly by influencing the kinematics of the free electrons, especially the interaction time with the RF field. According to charged particle tracking simulations, the contribution of the kinematics to $\Delta E_{\Delta P}$ amounts to $52\text{ meV}$ for electron energies initially differing by one photon energy.

To account for this effect, electron energy spectra are taken at various laser powers (see Fig. 3b). While 2-photon induced emission is dominant at low laser power, enhanced 3-photon induced electron emission is observed for higher laser power. This can also be seen in Fig. 3b), which shows the emitted electron current as a function of laser power. A polynomial fit (black line) to the data reveals the contributions of $n$-photon induced emission. Coulomb interactions can be neglected in this regime since on average 0.1 electrons per laser pulse are emitted from the tip at highest laser power.

We measure $\Delta E_{\Delta P}$ as a function of laser power between 50 and 250 mW, corresponding to a Keldysh parameter of $4.8 > \gamma > 2.1$, assuming a local laser field enhancement of 4 near the surface of the tip [18]. The measured energy shift is shown in Fig. 3a, where every data point represents the average of 50-100 individual energy measurements. For $P > 60\text{ mW}$ the energy shift is less than $20\text{ meV}$, which excludes a delay of the center of the electron pulse of more than 5 fs.

To explain the peak at lower laser power, both the kinematics of the free electrons and delays in photoemission have to be taken into account: Assuming a linear dependence of the kinematic energy shift on the initial kinetic energy, as well as a linear shift of the energy spectra as a function of laser power, this kinematic model would correspond to a delay $>5\text{ fs}$. Delayed emission is observed at low laser power.
is approximately two times the photon energy, as indicated by the red line and the gray shaded area in Fig. 3:

$$\varphi_{\text{eff}} = \varphi_0 - \sqrt{\frac{e U_{\text{DC}}}{4 \pi \epsilon_0 k_e k}} \sim 3.27 \text{ eV},$$

where $\varphi_0 = 4.5 \text{ eV}$ is the work function of tungsten and $k \sim 5$ is a parameter dependent on the tip geometry, which influences the local field enhancement of the DC field [19]. Recent calculations show that in this regime tunneling delays of a few fs are plausible [13], especially when the effective binding energy of an electron is close to a multiple of the photon energy [20]. Further detailed studies will allow to study these transmission resonances by quantifying the respective contributions of kinematic energy shifts and photoemission delays.

The sensitivity of this cavity enhanced RF photoelectron streaking setup can also be used to measure the RF phase within the cavity. With a streaking slope of 7.8 V/ps and an intrinsic width of the electrons’ energy distribution of about 0.5 eV one can measure the phase of the RF field within 64 fs, or 0.5 mrad at 9.08 GHz, upon the measurement of the kinetic energy of a single electron. Such precision in measuring the RF phase in situ, i.e. within the microwave cavity itself [21], will directly benefit techniques such as temporal focusing [22, 26] and aberration-free lensing [27] which compress or maintain femtosecond electron pulse durations at samples distant from the source.

Cavity enhanced RF electron streaking enables a direct measurement of the timing of laser triggered electron emission. While RF fields represent a minimally invasive probe avoiding quiver motion of the electron, recollision processes or interference of the pump and probe pulse, they still allow for a resolution at or below 1 fs and a dynamic range extending to the ps regime. We use this technique to longitudinally streak laser triggered photoemission from a tungsten nanotip and find significant photoemission delays at lower laser powers.

TJ would like to thank Dominik Leuenberger and Philipp Haslinger for helpful discussions. This research is funded by the Gordon and Betty Moore Foundation, and by work supported under the Stanford Graduate Fellowship.

---

1. P. Hommelhoff, C. Kealhofer, and M. A. Kasevich, Physical Review Letters 97, 247402 (2006).
2. P. Hommelhoff, Y. Sortais, A. Aghajani-Talesh, and M. A. Kasevich, Physical Review Letters 96, 077401 (2006).
3. B. Barwick, C. Corder, J. Strohaber, N. Chandler-Smith, C. Uiterwaal, and H. Batelaan, New Journal of Physics 9, 142 (2007).
4. C. Ropers, D. Solli, C. Schulz, C. Lienau, and T. Elsaesser, Physical Review Letters 98, 043907 (2007).
5. J. Hoffrogge, J. P. Stein, M. Krüger, M. Förster, J. Hammer, D. Ehberger, P. Baum, and P. Hommelhoff, Journal of Applied Physics 115, 094506 (2014).
6. M. Gulde, S. Schweda, G. Storeck, M. Maiti, H. K. Yu, A. M. Wodtke, S. Schäfer, and C. Ropers, Science 345, 200 (2014).
7. C. Kealhofer, B. B. Klopf, G. E. Skulason, T. Juffmann, S. M. Foreman, and M. A. Kasevich, Optics Letters 40, 260 (2015).
8. L. Wimmer, G. Herink, D. R. Solli, S. V. Yalunin, K. E. Echternach, and C. Ropers, Nature Physics 10, 432 (2014).
9. J. Breuer and P. Hommelhoff, Physical Review Letters 111, 134803 (2013).
10. E. A. Peralta, K. Soong, R. J. England, E. R. Colby, Z. Wu, B. Montazieri, C. McGuinness, J. McNeur, K. J. Leedle, D. Walz, E. B. Sozer, B. Cowan, B. Schwartz, G. Travish, and R. L. Byer, Nature 503, 91 (2013).
11. R. Yen, J. Liu, and N. Bloembergen, Optics Communications 35, 277 (1980).
12. M. Krüger, M. Schenk, M. Förster, and P. Hommelhoff, Journal of Physics B: Atomic, Molecular and Optical Physics 45, 074006 (2012).
13. R. Bormann, M. Gulde, A. Weismann, S. Yalunin, and C. Ropers, Physical Review Letters 105, 147601 (2010).
14. H. Yanagisawa, M. Hengsberger, D. Leuenberger, M. Kliöcker, C. Hafner, T. Greber, and J. Osterwalder, Physical Review Letters 107, 087601 (2011).
15. C. Kealhofer, S. M. Foreman, S. Gerlich, and M. A. Kasevich, Physical Review B 86, 035405 (2012).
16. A. L. Cavalieri, N. Müller, T. Upahue, V. S. Yakovlev, A. Baltuška, B. Horvath, B. Schmidt, L. Blümel, R. Holzwarth, S. Hendel, M. Drescher, U. Kleineberg, P. M. Echenique, R. Kienberger, F. Krausz, and U. Heinzmann, Nature 449, 1029 (2007).
17. S. Neppi, R. Ernstorfer, E. Bothschafter, A. Cavalieri, D. Menzel, J. Barth, F. Krausz, R. Kienberger, and P. Feulner, Physical Review Letters 109, 087401 (2012).
18. M. Schenk, M. Krüger, and P. Hommelhoff, Physical Review Letters 105, 257601 (2010), PRL.
19. V. T. Binh, N. Garcia, and S. T. Purcell, Advances in Imaging and Electron Physics, edited by W. H. P., Vol. Volume 95 (Elsevier, 1996) pp. 63–153.
20. M. Pant and L. K. Ang, Physical Review B 88, 195434 (2013).
21. G. J. H. Brussaard, A. Lassise, P. L. E. M. Pasmans, P. H. A. Mutsaers, M. J. van der Wiel, and O. J. Luiten, Applied Physics Letters 103, (2013).
22. T. van Oudheusden, P. Pasmans, S. van der Geer, M. de Loos, M. van der Wiel, and O. Luiten, Physical Review Letters 105, 264801 (2010).
23. A. Gliner, A. Apolonski, F. Krausz, and P. Baum, New Journal of Physics 14, 073055 (2012).
24. E. Fill, L. Veisz, A. Apolonski, and F. Krausz, New Journal of Physics 8, 272 (2006).
25. R. P. Chatelain, V. R. Morrison, C. Godbout, and B. J. Siwick, Applied Physics Letters 101, 081901 (2012).
26. G. H. Kassier, N. Erasmus, K. Haupt, I. Boschhof, R. Siegmund, S. M. M. Coelho, and H. Schooer, Applied Physics B 109, 249 (2012).
27. P. L. E. M. Pasmans, G. B. van den Ham, S. F. P. Dal Conte, S. B. van der Geer, and O. J. Luiten, Ultra-microscopy Frontiers of Electron Microscopy in Materials Science, 127, 19 (2013).