Multiphoton Photoemission from a Copper Cathode Illuminated by Ultrashort Laser Pulses in an rf Photoinjector

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(Received 24 November 2009; published 22 February 2010)

In this Letter we report on the use of ultrashort infrared laser pulses to generate a copious amount of electrons by a copper cathode in an rf photoinjector. The charge yield verifies the generalized Fowler-Dubridge theory for multiphoton photoemission. The emission is verified to be prompt using a two pulse autocorrelation technique. The thermal emittance associated with the excess kinetic energy from the emission process is comparable with the one measured using frequency tripled uv laser pulses. In the high field of the rf gun, up to 50 pC of charge can be extracted from the cathode using a 80 fs long, 2 μJ, 800 nm pulse focused to a 140 μm rms spot size. Taking into account the efficiency of harmonic conversion, illuminating a cathode directly with ir laser pulses can be the most efficient way to employ the available laser power.

DOI: 10.1103/PhysRevLett.104.084801 PACS numbers: 29.25.Bx, 41.75.Ht, 82.50.Pt

Photoemission is one of the fundamental processes in the physics of the generation of charged particle beams. In rf photoinjector sources, widely used for free-electron lasers and advanced accelerator applications, a cathode in a high field rf cavity is illuminated with photons with energy higher than the cathode work-function. Under these conditions, when a bound electron in the cathode material absorbs a single photon, it can be emitted by the surface, resulting in a linear relationship (with proportionality constant called quantum efficiency or QE) between the number of emitted electrons and the incident number of photons [1,2].

It has long been known that for higher illumination intensities, charge will be emitted even for sub-work-function energy photons. The condition in this case is that n photons have to be absorbed at the same time in order to promote an electron to an unbound state with enough kinetic energy to escape the material. The signature of the n-photon photoemission is the scaling of the emitted charge as the nth power of the laser intensity [3]. On a log-log scale, the charge yields (number of electrons per number of photons) for single and multiphoton emission are lines with different slopes. Even though the n-photon process is statistically more rare than the single photon one, for large enough illuminating intensity, it will be advantageous to use multiphoton photoemission to extract the maximum amount of charge from a cathode.

A limit to reaching this crossover intensity could be set by the damage threshold of the surface. Using relatively low energy but ultrashort laser pulses allows one to stay below the damage threshold (a few 100 GW/cm^2 for sub-ps pulses), yet access very high intensities, and hence achieve relatively large charged particle yields. In doing this, one must be careful to avoid space charge effects from the emitted electrons near the surface. The accumulation of a surface charge layer in the region close to the cathode eventually leads to the creation of a virtual cathode and cuts off further emission [4]. This can be avoided by providing a large extracting field on the cathode to quickly accelerate away the emitted charge.

In the low field regime, many papers have explored the subject of multiphoton photoemission from metal cathodes using short laser pulses [5–8]. The focus of this early work was on the significant role played by thermal decoupling of the electron and the lattice after the short pulse excitation. The total charge yields found in these early experiments were not impressive as space charge and virtual cathode effects in the low accelerating field environment set a tight upper limit to the extractable surface charge density. Further, up until a few years ago, virtually all rf photoinjectors were, at least in the design stage, conceived to run with a relatively long laser pulse ~5–10 ps on the cathode in order to limit the space charge forces and generate high brightness 1 nC class beams [9,10].

Given these premises, it is not surprising that the common paradigm to generate high brightness electron beams with photocathodes has been to frequency upconvert the available drive laser power to the ultraviolet (uv) to overcome the work function of the material and emit sizable beam charge. Depending on the beam applications, metals (having relatively low QE, typically between 10^{-5} and 10^{-4}) have often been preferred to semiconductors in rf photoinjectors for their fast response, durability, and vacuum compatibility [11].

Recently, due to the favorable brightness scaling, ultrashort low charge beams have proved to be useful for a variety of applications like driving free-electron laser [12] or as structural dynamics probes in ultrafast electron diffraction [13,14]. In this context we, at the UCLA Pegasus laboratory, have demonstrated a new regime of operation of the rf photoinjector [15,16], where a pancakelike beam is generated and then evolves under the action of its own space charge forces into a nearly ideal uniformly filled ellipsoidal beam distribution [17]. This beam generation method calls for ultrashort laser pulses and so it naturally goes along with exploiting the advantage of multiphoton photoemission at high laser intensities.

In this Letter we study the properties of an electron beam generated by multiphoton photoemission from a very short infrared (ir) laser pulse on a Cu cathode in a rf photoinjector. We have recorded up to 50 pC of beam charge when using a 80 fs long 2 μJ of 800 nm pulse. For multiphoton photoemission, it is improper to define a QE as the number of electrons is not linearly dependent on the number of photons. If we would have up-converted the photons to uv instead of sending the ir directly onto the cathode, assuming a 10% conversion efficiency, the QE required to obtain the same amount of charge would be $>0.1\%$, an order of magnitude higher than any metal cathode ever tested in a working rf photoinjector. The photoemission has been measured to be prompt and the thermal emittance levels are comparable to the values measured for the uv case [17].

According to the generalized Fowler-Dubridge theory for multiphoton photoemission, the emitted current density when photons of energy $h\nu$ illuminate the surface can be written [18] as a sum of partial currents J_n where

$$J_n(h\nu) = a_n A\left(\frac{e}{h\nu}\right)^n (1 - R_\nu)^n I^n T_e^2 F\left(\frac{nh\nu - e\Phi}{k_b T_e}\right) \quad (1)$$

in which n is the order of the n-photon process, a_n is a constant representing the probability of the multiphoton process to happen, T_e the electronic temperature, $A = 120\ \mathrm{A/cm^2\,K^2}$ is the Richardson constant, e is the electron charge, k_B and h are the Boltzmann and the Planck constant, and R_ν is the reflectivity coefficient at the frequency ν and Φ is the cathode work function modified by the Schottky effect. The Fowler function $F(x) = \int_0^\infty \ln(1 + e^{-(y+x)})dy$ strongly suppresses the terms for which its argument is negative. Since the a_n, representing the likelihood of a n-photon process, are quickly decreasing function of n, the dominant term in the sum is given by the lowest n such that $nh\nu > e\Phi$.

For laser intensities larger than a critical value $I_{\mathrm{cr}} \geq \sqrt{\frac{a_1(1-R_{3\nu})}{3a_3(1-R_\nu)^3}}\frac{h\nu}{e_0}$ for which $J_3(h\nu) \geq J_1(3h\nu)$, the 3-photon process yields more electrons than the single photon emission process. The a_n are material dependent and difficult to calculate theoretically. Using order of magnitude estimates for $a_1 \sim 10^{-14}\ \mathrm{cm^2\,A^{-1}}$ and $a_3 \sim 10^{-34}\ \mathrm{cm^6\,A^{-3}}$ [19,20] and assuming similar reflectivities at ir and uv wavelengths, $I_{\mathrm{cr}} \approx 10\ \mathrm{GW/cm^2}$. Generally, solid state laser media have gain bandwidth centered in the near ir region (and so below the work function of most metal cathodes) so that the higher energy photons are usually obtained by frequency multiplying the ir laser pulses. Since the harmonic conversion (usually taking place in non linear crystals) is lossy, the intensity threshold over which employing a n-photon emission process starts to be convenient is practically even lower than I_{cr}.

The experiments took place at the UCLA Pegasus photoinjector laboratory. The photoinjector driver laser is a Ti:Sa laser system generating ultrashort laser pulses centered at 800 nm. In previous experiments described elsewhere [17], uv was obtained by frequency tripling the laser using BBO crystals. For the measurements described in this Letter, we bypassed the harmonic generation crystals and illuminated the cathode directly with the ir photons. A half-wave plate and a polarizer are used to vary the energy of the pulse illuminating the cathode. The laser pulse length can be adjusted between 80 and 400 fs by moving the retroreflector in the final grating compressor, and is monitored with a polarization gating scanning autocorrelator. The laser transverse spot is controlled by overfilling an adjustable iris aperture which is then imaged to the cathode using a $f = 1.5$ m lens in a $2f$-$2f$ configuration. Prior to the final vacuum window, a beam splitter is used to sample a portion of the beam for measuring the energy per pulse and the beam transverse shape at the cathode. The gradient in the 1.6 cell rf gun can be varied between 55 MV/m and 90 MV/m by changing the input rf power. The cathode was prepared using off-axis single-point diamond turning, followed by a protective $\mathrm{MgF_2}$ antireflective coating to reduce oxidation and increase lifetime. Normal incidence reflectivity at 800 nm was measured to be <15%. No difference in the QE or the thermal emittance was observed due to the cathode coating. The laser incidence angle on the cathode is less than 3° from the normal so that polarization effects are negligible. After exiting the gun, the beam is transported to the end of a 4 m long beam line where the charge is measured using a Faraday cup.

Experimentally, we obtain the order n of the emission process by measuring the emitted charge as a function of input laser intensity. In Fig. 1 we report the measurements of emitted charge as a function of laser pulse energy for different spot sizes.

In order to understand the results, we note using Eq. (1) that it is the current density (and not simply the charge)

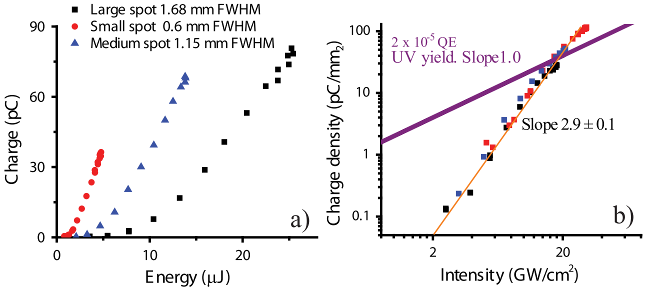

FIG. 1 (color online). (a) Charge yield for different spot sizes at the cathode as a function of laser energy. (b) Emitted charge density vs laser intensity. The curve for uv photoemission with the measured QE of 2×10^{-5} is also reported.

which is proportional to the third power of the intensity. The final current density of the electron beam depends on the beam dynamics with the final bunch length dominated by the space charge expansion taking place during the beam "blowout." At emission, the current density is simply the charge density divided by the laser pulse length. By plotting the charge density vs the laser intensity, we obtain the curve in the log-log scale in Fig. 1(b) where all the measurement points fall back on the same line. A linear fit gives a power law $J = CI^x$ with an exponent $x = 2.9 \pm 0.1$ which within the errors fits well with the simple three photon photoemission model. The constant for the power law is $C = 30\ \mathrm{A/cm^2}$ per $(\mathrm{GW/cm^2})^3$ which corresponds to $a_3 = 9 \times 10^{-35}\ \mathrm{cm^6/A^3}$ in Eq. (1).

To obtain a more useful scaling law, we observe that the emitted charge Q is proportional to the temporal and area integral of the current density J. Since the time and the area of emission are the same as the laser pulse length and spot size, we have

$$Q = J\tau A = CI^3\tau A = C\frac{E^3}{\tau^2 A^2} \qquad (2)$$

where E is the pulse energy, τ its length, and A its transverse area. For example, if an application requires a 100 pC beam in a 300 μm rms spot size at the cathode ($A = 0.56\ \mathrm{mm}^2$), we would need 16 μJ (6.5 μJ) of ir energy with a 200 fs (50 fs) long laser pulse.

In Fig. 2(a), we show the charge yield curves for different peak field on the cathode to illustrate the effect of the rf field. There are two components of this effect. The main one which explains the larger yield at higher field values is the lowering of the effective work function. In a high field environment, Φ in Eq. (1) has to be replaced by $\Phi - \sqrt{e\beta E_l/4\pi\epsilon_0}$ where $E_l = E_0 \sin\theta$ is the field at injection phase θ. Using the data, we can fit a field enhancement factor β of 1.5 ± 0.2. The other important effect derives from the fact that a larger accelerating field at the cathode allows the emission of a higher charge density σ before being limited by the virtual cathode formation effects $\sigma/\epsilon_0 < E_l$. In the measurements, the deviation from the cubic power law at high laser intensities for the $E_0 = 55$ MV/m curve is due to the onset of these saturation effects. The maximum charge emission efficiency was obtained by raising the gradient in the rf gun in order to obtain an injection field $E_l > 50$ MV/m. In these conditions, we have recorded up to 50 pC of beam charge using a 80 fs 2 μJ laser pulse focused down to 140 μm rms spot size. Assuming a 10% frequency tripling efficiency and a 5×10^{-5} QE for typical Cu cathode, in order to generate the same amount of charge, 50 μJ of ir would have been required. Using multiphoton photoemission in this case reduces the photoinjector driver laser power requirements by more than 1 order of magnitude.

Another important effect that we investigated experimentally is the dependence of the charge yield on the laser pulse length. An important issue in earlier papers on multiphoton photoemission is the role taken by the nonthermal electron distribution generated by the ultrashort laser pulse excitation [18,21]. For certain materials (for example tungsten) it was verified that the charge yield follows a different power law due to the fast heating of the electrons, and that the emission occurs on time scales longer than the laser pulse. To rule out any effect of the thermal decoupling of the electrons and the lattice, we performed an autocorrelation measurement by recording the amount of charge as a function of the time delay between two ir pulses on the cathode [see Fig. 2(b)]. The good agreement with the optical autocorrelation indicates that the emission is prompt with no delay introduced by the 3-photon absorption. We also measured the charge yield for different pulse widths [Fig. 3(a)]. The variation of the charge as a function of pulse length at constant laser energy [Fig. 3(b)] shows the predicted [see Eq. (2)] inverse square dependence confirming that a simple multiphoton photoemission process is occurring.

One could argue that the multiphoton photoemission would be more sensitive to energy fluctuations (jitter) and spatial inhomogeneity of the laser pulse. On the other hand, in practice, the uv laser pulse used to overcome the cathode work function in conventional rf photoinjectors is created by nonlinear processes starting from an ir pulse and its characteristics (energy stability and spatial inhomogeneity) depend from the cubic power of the initial ir laser pulse intensity also in this case.

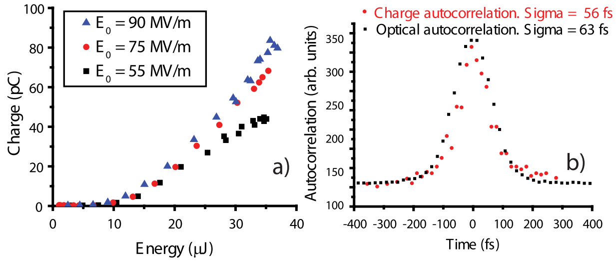

FIG. 2 (color online). (a) Multiphoton photoemission charge yield for three different accelerating gradients. The laser spot size is 900 μm rms. The measurements are taken at a constant injection phase $\theta = 30°$. (b) Charge (black) and optical (red or gray) autocorrelation of the ir pulse.

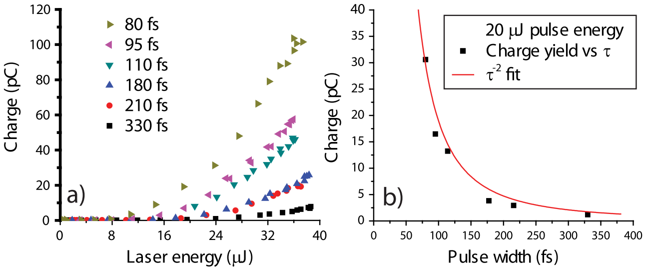

FIG. 3 (color online). (a) Yield vs different laser pulse length for 700 μm spot size at the cathode. (b) Charge as a function of pulse length for constant 20 μJ laser energy.

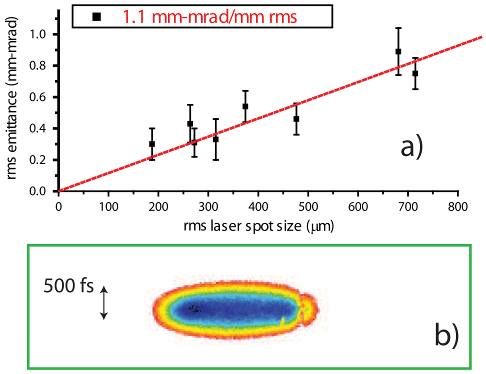

FIG. 4 (color online). (a) Thermal emittance of the ir photoelectrons. (b) Uniformly filled ellipsoidal beam obtained using an ir laser pulse on the cathode.

A final concern is related to the beam thermal emittance. The beam emittance has been measured as a function of the laser spot size using the rf deflector and the quadrupoles by a slice emittance measurement technique. The measurement results are reported in Fig. 4. The obtained value for the slope is 1.1 mm mrad/mm rms, in agreement with the value measured for uv. This is not surprising since the excess kinetic energy of the single uv photon or the 3-photon emission process is the same. We note that the thermal emittance value is a factor of 2 larger than the theoretical predictions. There is an ongoing debate in the literature on this inconsistency [22].

We have also verified that the space charge driven expansion dynamics, leading to the formation of the uniformly filled ellipsoidal beam, is not affected by the emission process. In Fig. 4(b), we show the projection of the beam distribution onto the x-z space obtained using a quadrupole and the rf deflector (as in [17]) revealing an elliptical boundary with very sharp transition indicating a very short beam at the cathode. Another indication that the emission is prompt was obtained by reducing the charge to a few pC and observing an electron bunch length on the rf deflector shorter than 200 fs.

In conclusion, we have shown very significant charge yield using modest levels of laser energy from metal cathodes through multiphoton photoemission. Because of the tremendous progress in ultrashort laser systems and the new developments in photoinjector beam dynamics, it becomes feasible to drive a photoinjector with sub-work-function laser photons. For this operating mode, the favorable scaling with laser intensity is even more important than the measured emission efficiency, which is influenced by many factors including the surface quality. The explanation of the larger yields measured compared to early papers on this subject are to be sought in the very high electric field on the cathode and the ultrashort pulse length which enables us to reach high intensities without incurring surface damage. These results have the potential to revolutionize the field of electron beam generation in rf photoinjectors. In particular these findings are poised to greatly simplify the requirements (and the cost) of the photoinjector driver laser system which typically starts from ir lower energy photons and uses non linear optics to generate photons of enough energy to overcome the material work function. The conversion processes are typically lossy and introduce undesirable complexity in the system. High average power photoinjectors that are currently limited by the achievable laser power could consider the use of multiphoton photoemission from metal photocathodes. Multiphoton emission from semiconductor cathodes should also be reconsidered in this short pulse, high extraction field regime.

This work was supported by Office of Naval Research Grant No. N000140711174 and Department of Energy Grant No. DE-FG02-92ER40693.

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