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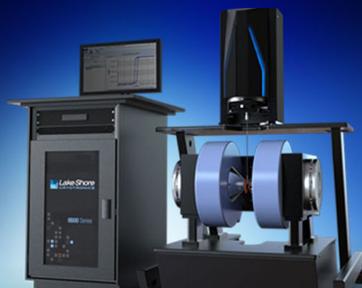
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Coulomb interactions in sharp tip pulsed photo field emitters

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Photofield emitters show great potential for many single electron pulsed applications. However, for the brightest pulses $>10^{11}$ A/(m² srV), our simulations show that Poisson statistics and stochastic Coulomb interactions limit the brightness and increase the energy spread even with an average of a single electron per pulse. For the systems, we study we find that the energy spread is probably the limiting factor for most applications. © 2016 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). [<http://dx.doi.org/10.1063/1.4963783>]

In this study, we will use the term photofield emitter to mean a sharp tip in a strong electric field (>0.1 V/nm), with a laser incident on the tip. So, we consider both field emission of photo excited electrons, and directly photo emitted electrons entering into a strong field.

People have suggested using photofield emitters in ultra-fast electron microscopy for many years, see for example, King *et al.*¹ The groups of Hommelhoff^{2,3} and Ropers^{4,5} are highly also active in this area. Further theoretical studies include the recent work of Zhang and Lau⁶ or Jensen.⁷ Recently, Mohammed *et al.*⁸ have adapted an SEM by laser illuminating the already present Schottky tip, and Feist *et al.*⁹ have a similar set up in a TEM with a tungsten field emission cathode (apex radius of curvature of about 120 nm). Photofield emitters could also be useful in electron interferometers or other quantum experiments. Given the wide range of uses and recent interest, we will investigate photofield emitters in more detail using simulations and theory.

We restrict ourselves to low charge (0.1 – 20 electrons per pulse) ultra-fast pulse applications like stroboscopy and electron interferometry where beam quality, coherence, emittance, and brightness are important. For a bright photofield emitter, we want an emission area in the nm range, ultra-short pulses <100 fs, and as many electrons as possible within the desired emittance.

Here, we will show that Coulomb interactions, which occur after the electrons are emitted into a beam, will reduce its quality. The logical conclusion is to work with bunches containing a maximum of one electron per pulse. Unfortunately, in this case, we expect that Poisson statistics will start to limit the reduced brightness B_r when the average number of electrons per pulse $\langle N_e \rangle$ is less than one. To avoid any confusion, we will ignore the physics of emission and emit the electrons into the vacuum with 0 energy spread and 0 emittance, so all the emittance and energy spread come from Coulomb interactions occurring in the journey from tip apex to screen.

To get some feeling for the magnitude of the Coulomb interactions, we can calculate the effect for a pulse of 2 electrons emitted on axis with a 10 fs spacing. Let us assume

that near the surface of the tip there is a uniform field of 1 V/nm, (this is typical see, for example, Ref. 10). The first electron emitted with no initial velocity travels 8.8 nm in the 10 fs before the next electron is emitted. The potential energy of the one electron in the field of the other is 0.16 eV, which is about equal to the intrinsic energy spread caused by the Fermi-Dirac distribution in the metal.¹⁰ In this case, Coulomb interactions will cause the final energy spread to be about twice the intrinsic value. This is a conservative estimate since the electrons will likely be closer together depending on the temporal shape of the pulse.

Coulomb effects may already limit the emission well before other factors such as thermal effects or laser damage (ablation) start to cause problems. For an average of 1 electron in our 10 fs pulse as described, we would expect a laser intensity >10 GW/cm² (assuming a quantum efficiency of $<0.1\%$). According to Hommelhoff *et al.*,² these kind of intensities are close to the damage threshold for W tips.

Donders¹¹ has investigated Coulomb interactions in continuous photofield emitters, Qiang *et al.*¹² have looked at 30 ps pulses from nano-tips, and Siwick *et al.*¹³ investigated Coulomb interactions in drift space for ultra-fast electron diffraction. Finally, there is our own work,^{14–16} which investigated electron-electron interactions in Schottky, cold-field, and DC photo field emitters.

None of the above has investigated the ultra-short pulses with up to 20 electrons per pulse, suitable for stroboscopy. Therefore, it is high time we understood the consequences Coulomb interactions and Poisson statistics have on photofield emitted electron pulses.

We begin by describing the manner in which we simulated the photofield emitter. Then, we examine the emittance and brightness of the photofield emitter as a function of $\langle N_e \rangle$ and find that even with pulse compression there is a maximum Coulomb limited B_r . We finally look at the energy spread and again find that even with corrective time dependent electron optics a stochastic energy spread remains.

The bulk of the calculation are performed using a home built Poisson solver based on the work of Kang *et al.*,¹⁷ using the geometry in Figure 1. Ray tracing and Coulomb interactions were performed with GPT (General Particle Tracer) from pulsar physics,¹⁸ for more information, see the [supplementary material](#). The emission area on the tip is limited to

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1/4 of the tip's radius, where most of the current comes from, see Ref. 15 for more discussion. We start the electrons with a uniform spherical distribution at 10 nm from the front of the tip, as if they had come from the centre of the spherical part of the tip. The speed of the electrons is $\sqrt{2Ue/m}$ where U is the potential at their launch point. Since the electrons appear to come from a point, they have zero emittance, and we assign them zero energy spread. The position and velocity are measured in the field free region $z = 1.2$ mm. Temporally, we used a 10 fs top hat which is a best case scenario (see the [supplementary material](#) for more information). We measure the pulse length τ_{20-80} from when 20% of the electrons arrive until 80% arrive. Therefore, the 10 fs top hat becomes $\tau_{20-80} = 6$ fs. The number of electrons in a pulse is decided by Poisson statistics using the random number generator `poissrnd` in Matlab.

A 50 nm tip radius is typical and likely to be robust compared to for example, an atomically sharp tip. In Ref. 19, we found that a 50 nm field emitter tip suffered similarly with Coulomb interactions compared to a 10 nm tip and less than larger 100 nm tips. However, as shown by Verduin,^{15,20} Coulomb effects depend on geometry and tip radius. According to Figure 7 of Ref. 20, reducing the tip length from 0.5 mm to 0.05 mm would increase the Coulomb limited B_r by around 4 times due to the similarly 4 times larger extraction voltage needed to achieve the same field at the tip. In reality, a field emission tip with such a short length is exotic and would require a change in the traditional method of tip manufacturing. We choose a length of about 1 mm because energy spread effects occur close to the tip and emittance growth becomes too dependent on the particular design of the gun beyond the first mm.

We determine the emittance from the trace results by finding the area in a plot of radial position versus radial velocity that contains 50% of the electrons. In Figure 2, we see the emittance growth for a 50 nm tip with an initial $\tau_{20-80} = 6$ fs electron pulse. The emittance does not drop below 10^{-11} mrad since we are limited by aberrations and numerical errors (see [supplementary material](#) for more discussion on numerical errors).

Despite the aberrations the emittance grows even when $\langle N_e \rangle$ is less than one, clearly the Poisson statistics take effect at below the (on average) single electron per pulse level. The emittance values that were initially 0 are comparable to

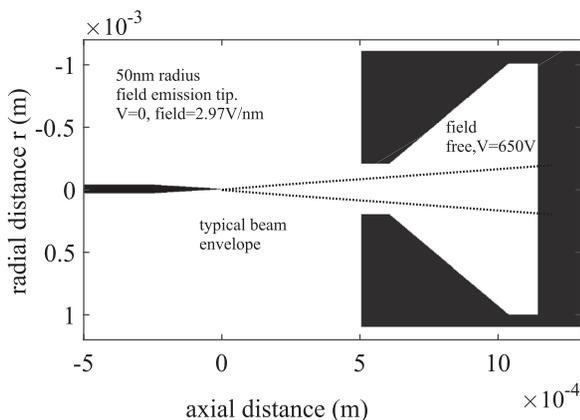


FIG. 1. Photofield gun, black indicates metal.

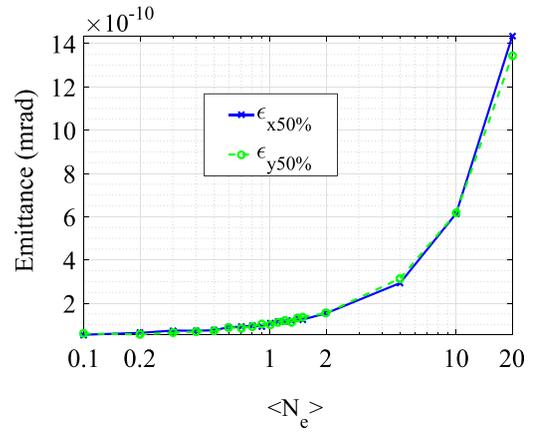


FIG. 2. Emittance (x and y) growth due to Coulomb interactions for the photofield emitter in Figure 1.

intrinsic values as can be found from a back of the envelope calculation. From Ref. 10, for a pure photo emitter (i.e., no field), we can write $r_{tip}^2 \Delta E_x = \epsilon^2 V$, where V is the beam voltage and ΔE_x is the excess photon energy typically $\Delta E_x = 0.1$ eV making $\epsilon = 10^{-9}$ m rad. A method for adding a Coulomb contribution to an intrinsic distribution is discussed by Bronsgeest in Ref. 16, often simple addition in quadrature will suffice.

Now let us examine how the B_r of the field emitter changes with $\langle N_e \rangle$. We calculate B_r as $\langle N_e \rangle q / (\tau_{20-80} \epsilon^2 V)$. In Figure 3, the dotted (red) line uses τ_{20-80} as measured at the screen. During the 1.2 mm trip τ_{20-80} for the pulse with $\langle N_e \rangle = 20$ extends to nearly 1 ps this in addition to the lateral broadening also reduces B_r . Here, the B_r as determined purely by Coulomb interactions reaches a maximum at $\langle N_e \rangle = 0.4$. The solid (blue) line is the theoretical B_r limit assuming perfect pulse compression back to the original $\tau_{20-80} = 6$ fs. In this case, only lateral broadening reduces B_r and the maximum is between 0.4 and 2 electrons per pulse. This result also matches with our other work on Coulomb interactions with continuous current field emitters.²¹

Note that the maximum $B_r = 2.4 \times 10^{11}$ A/(m² sr V) is less than the Pauli/quantum limit of 1×10^{12} A/(m² sr V) for $\Delta E = 0.1$ eV given in chapter 7 of Ref. 21. This does not contradict the work of Jarvis²² who claims to have

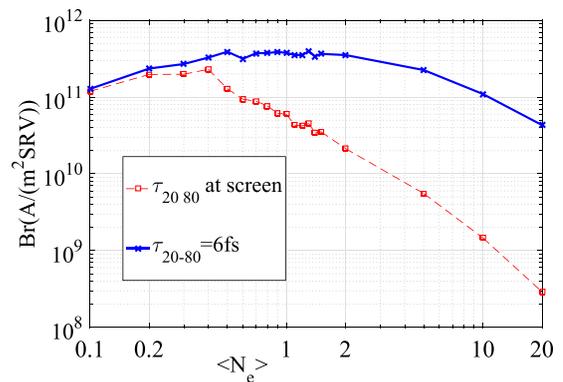


FIG. 3. Reduced brightness of photofield emitter in Figure 1 as a function $\langle N_e \rangle$. The solid blue line is the B_r as calculated without any increase in τ_{20-80} ; this represents the theoretically achievable limit if we could perform perfect pulse compression. The dotted red line is the B_r with τ_{20-80} measured at the screen.

experimentally created pulses with B_r at the quantum limit. If you can measure individual pulses with $\langle N_e \rangle = 1$, then some will truly have just one electron and therefore no Coulomb interactions; however when many pulses are taken on average, then our simulations suggest that B_r will be limited by Coulomb interactions. Also, reducing the initial pulse length will, for low $\langle N_e \rangle$, increase B_r but will also cause the maximum B_r to occur for lower $\langle N_e \rangle$ than here. Poisson statistics and Coulomb interactions limit the usable B_r , in this case the maximum $B_r = 2.4 \times 10^{11} \text{ A}/(\text{m}^2 \text{ sr V})$. Note that this is substantially higher than the $1.6 \times 10^9 \text{ A}/(\text{m}^2 \text{ sr V})$ that has experimentally been obtained in DC field emission sources²³ which are limited by the emission process and the maximum extractor voltage usable before arcing occurs. In Ref. 3 by Ehberger *et al.*, they find an emittance $8 \times 10^{-11} \text{ mrad}$ at 44 eV with a 130 fs pulse length which is similar to the emittances we are discussing here.

We already identified that the energy spread is probably the area that gives the most issues. Here, we will discuss simulations of the energy spread of the photofield emitter caused by Coulomb effects and what might be done to it. Figure 4 shows the energy distribution for $\langle N_e \rangle = 20$ caused only by Coulomb interactions (since our initial energy spread was 0). The FW50 (the full width containing 50% of the current) energy spread measured at the detection plane is 8 eV. If we compare this to the intrinsic energy spread of a field emitter (about 0.3 eV), this is large. At this moment, it is unclear what the intrinsic energy spread of a photofield emitter is; in experiments such as Ref. 24, there is absorption of additional photons or effects from the rapidly changing ponderomotive forces. Our results show that at $\langle N_e \rangle = 1$ there is very little effect from Coulomb interactions in line with the published results from the experimental photofield set-ups of Refs. 9 and 8 where there is typically only a single electron per pulse. It should be noted that both Refs. 8 and 9 are using a much larger tip, so the current density at the tip is smaller than here, which means smaller B_r and less Coulomb effects. In Figure 4(a), the electrons velocity in the z-direction is plotted versus the time of detection for pulses with an average of 20 electrons, showing as expected, electrons in the

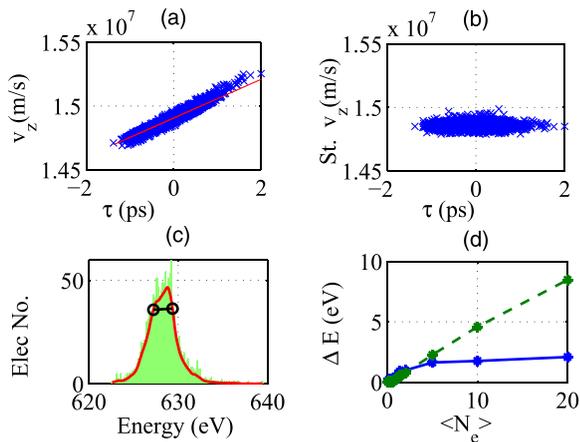


FIG. 4. Method to linearly reduce ΔE . (a): Raw data from $\langle N_e \rangle = 20$ pulse with linear fit in red. (b): data after subtracting the linear fit. (c): An example energy spread from which we find the FW50. (d): The (green) dotted line is the FW50 energy spread without correction, and the (blue) solid line is the stochastic energy spread left over after correction.

front of the pulse are accelerated and electrons in the tail are decelerated. This opens the path to pulse compression.

Recently, it has been demonstrated that ultra-fast diffraction can be performed with a bunch of electrons that have been compressed with a time varying electromagnetic field. In Ref. 25, the authors create a bunch of electrons with linear space charge and allow it to expand, until it reaches a radio frequency cavity, where an oscillating electric field reverses the linear expansion. We will look at the total energy spread, and also a corrected energy spread which approximates the action of the radio frequency cavity, we call the corrected energy spread the stochastic energy spread. To correct the energy spread, we used the least square method to fit a straight line through the z velocity data plotted against time 4(a). The linear dependence on time was removed from the velocity data 4(b), and we found the new FW50 4(d).

Figure 4(d) shows that the total energy spread quickly grows to a massive 8 eV yet the stochastic energy spread levels out at around 2 eV.

What should be noted for the stochastic ΔE is that we did not actually compress the electrons, and when compressing the electrons, they will undergo further stochastic interactions which would broaden the energy spread.

The slow stochastic growth roughly follows the theory of Jansen.²⁶ For dc beams in the Lorentzian regime $\Delta E = 1.90 \times 10^5 \frac{J\pi r}{\alpha U_{ext}^{1/2}}$, here U_{ext} is the extractor voltage, α is the final half angle (10 mrad), r is the initial radius of the electrons (15 nm), and $J = \frac{\langle N_e \rangle e}{\tau_{20-80} \pi r^2}$ with τ_{20-80} measured at the screen. The theory has been tested experimentally with Schottky emitters,¹⁶ and we have compared it to N-body simulations of cold field emitters in Ref. 19. Here, it predicts $\Delta E = 2.6 \text{ eV}$ at $\langle N_e \rangle = 20$, given the naive application to our pulsed system this is surprisingly good.

We have shown that the stochastic Coulomb interactions can affect the energy spread and brightness of a photofield emitter. For our system, the maximum B_r allowed by Coulomb Interactions was $B_r = 4 \times 10^{11} \text{ A}/(\text{m}^2 \text{ sr V})$ near $\langle N_e \rangle = 1$. The stochastic energy spread was $\approx 2 \text{ eV}$ more than double that of a Schottky emitter or four times a cold field emitter both typically used in high resolution imaging. The total uncorrected energy spread grows almost linearly with $\langle N_e \rangle$, the average number of electrons per pulse.

See [supplementary material](#) for more information on the calculation method. Method of calculating the fields, ray tracing, and numerical errors.

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