Lossless monochromation for electron microscopy with pulsed photoemission sources and rf cavities

C. J. R. Duncan,1 D. A. Muller,2 and J. M. Maxson1∗

1Cornell Laboratory for Accelerator-Based Sciences and Education, Cornell University, Ithaca, NY 14853, USA
2School of Applied and Engineering Physics, Cornell University, Ithaca, NY 14853 USA

Resonant radiofrequency cavities enable exquisite time-energy control of electron beams when synchronized with laser driven photoemission. We present a lossless monochromator design that exploits this fine control in the one-electron-per-pulse regime. The theoretically achievable maximum beam current on target is orders of magnitude greater than state-of-the-art monochromators for the same spatial and energy resolution. This improvement is the result of monochromating in the time domain, unconstrained by the transverse brightness of the electron source. We show analytically and confirm numerically that cavity parameters chosen to minimize energy spread perform the additional function of undoing the appreciable effect of chromatic aberration in the upstream optics. We argue that our design has significant applications in both ultra-fast and non-time-resolved microscopy, provided photoelectron sources of sufficiently small size and laser sources of sufficiently high repetition rate. Our design achieves in simulations more than two orders of magnitude reduction in beam energy spread, down to single digit meV. Overcoming the minimum probe-size limit that chromatic aberration imposes, our design clears a path for high-current, high-resolution electron beam applications at primary energies from single to hundreds of keV.

I. INTRODUCTION

The electron-optical properties of time varying fields have long been of fundamental interest to electron microscopists and accelerator physicists [1–6]. Resonant radiofrequency (rf) cavities in particular have found significant use over the past two decades in time-resolved electron diffraction and microscopy. Highlights of a versatile range of applications include: compressing electron pulses in time to the femtosecond scale and below [7–14], temporal magnification of ultrafast events [15, 16], impulsive acceleration and deceleration of beams over short distances [13, 17–20], chopping continuous beams into short-pulses [20–23], and the controlled introduction of time-of-arrival correlations for performing energy measurements [13, 24]. Radiofrequency cavities are the workhorses of particle acceleration at primary energies above 1 MeV. Efforts toward adopting rf technology in electron microscopy have historically struggled to achieve the required precision in the timing of electron bunches [6]. Today, advances in broadband laser sources and photocathode materials have largely solved the problem of synchronising electron pulses with rf phases [25], setting the stage for a fruitful exchange of expertise between the accelerator and microscopy research communities.

The focus of the present work is the use of cavity fields to compress beam energy spreads in the single-electron-per-pulse regime. Lower energy spread beams are advantageous in all electron microscopy, both static and time-resolved. The importance to electron energy loss spectroscopy (EELS) is clear: monochromation of continuous wave (CW) cold field emission (CFE) sources to the single meV scale has enabled measurements of phonon spectra with atomic spatial resolution in the scanning transmission electron microscope (STEM) [26, 27]. Ultrafast pulsed beams of equally narrow energy spread would make it possible to resolve the different contributions of coupled lattice, charge, and spin dynamics to the spectrum of quantum materials [28]. In applications beyond spectroscopy, source energy spread limits the spatial resolution of electron microscopes. Chromatic aberration is the barrier to achieving atomic diameter probes at low primary energies of less than 5 keV [29], a commonplace regime for scanning electron microscopy and industrial metrology methods such as time resolved cathodoluminescence. Finally, designs for damage-mitigating, pulsed multipass electron microscopes employ monochromation [30].

The challenge in producing low energy spread beams is that the best sources have intrinsic spreads of hundreds of meV. To date, monochromator designs that reach the single meV energy scale have relied on apertures in energy-dispersive locations [31, 32]. Apertures cause a loss of beam current by a factor equal to the ratio between the desired energy spread and the source energy spread. A factor 100 reduction leaves little current for imaging in the continuous case [26], and prohibitively low current in the ultrafast case. Lossy monochromation at low voltages is infeasible because beam current must increase as accelerating voltage decreases to maintain tolerable detector signal.

Pulsed sources combined with rf fields provide a direct experimental handle on the beam’s longitudinal phase space, comprising the conjugate dynamical variables of forward momentum and time of arrival at a given transverse plane [12, 33]. Photoemission is capable of delivering sub-picosecond electron pulses with femtosecond timing precision to experimental targets. Lossless energy spread reduction is therefore possible because the time of arrival — and hence rf accelerating phase — is tightly

∗jm586@cornell.edu
correlated with the energy of the particle.

The body of this paper begins in Sec. II with an analysis of the trade-off between current and energy spread in photo-emission and the statement of a fundamental lower bound on energy spread as function of current on target. Considering photoemission from a planar source, Sec. III derives and solves analytic conditions on the cavity parameters for energy-spread minimization. Particle tracking simulations confirm these analytic results. Precise synchronization is essential to minimising energy spread in our scheme and Sec. IV analyses the effect of timing jitter at the 10 fs scale, precision that has been achieved with bunching cavities in ultrafast diffraction beam lines [25]. We show that 10 fs timing uncertainty shifts the peak of the final energy distribution by 1 meV and broadens the tails by order 10 meV. Section V investigates the effect of the cavities on the transverse coherence of the beam. Analytic results show that the same cavity parameters that are optimal for energy spread reduction also perfectly cancel the effects of spherical and chromatic aberrations in the electron gun. We compare this prediction of our analytical formula with particle tracking simulations.

II. FUNDAMENTAL TRADE-OFF BETWEEN ENERGY RESOLUTION AND BEAM CURRENT

Monochromation entails a trade-off between final energy spread and average current on target, both in existing aperture-based energy-selectors and our proposed lossless design. Figure 1 shows a schematic of our design side-by-side an energy-selector. The constraints that impose the current-energy trade-off are different between the two devices. A comparison helps to situate our design in relation to the state of the art. A first analysis is simplified by neglecting the contribution that transverse momenta make to total particle energy. The end of this section returns to the complication introduced by accounting for the transverse store of energy.

The conservation of longitudinal emittance in a pulsed beam relates the minimum energy spread achievable in lossless transport $\Delta E_{\text{in}}$ to the initial laser pulse length $t_l$ at the source, the final electron pulse length $t_f$, and the initial electron energy spread $\Delta K$:

$$\Delta E_{\text{in}} = \frac{t_l}{t_f} \Delta K \geq \frac{\hbar}{2t_f}.$$  \hspace{1cm} (1)

The rightmost inequality is a consequence of the Heisenberg uncertainty principle, which sets the fundamental limit to longitudinal emittance. The factor $t_l$ includes the response time of the photocathode: on the scale of 10 fs [34] for typical metallic photocathodes, and extending much longer (up to 100fs and above) for semiconductor photocathodes [35, 36]. The single-electron-per-pulse regime reaches the lowest possible emittances (both transverse and longitudinal) because of the absence of Coulomb interactions that would otherwise broaden the energy distribution and spoil transverse coherence. In this regime, the maximum average current of a laser driven system synchronized to rf cavities is $I_{av} = e f$, where $e$ is the electron charge and $f$ is the resonant frequency of the cavities. In time-resolved, pump-probe systems, the minimum practicable rf period is set by the time it takes the sample to relax to the ground state after pump excitation. Relaxation times vary significantly depending on the sample and the desired excitation strength. For non-time-resolved systems, the maximum repetition rate is equal to the cavity resonance frequency.

Pulses that stretch to fill the rf cycle acquire unwanted non-linear energy-time correlations, and thus these higher-order effects bound the allowable final pulse length. Let the duty cycle $D$ denote the ratio of final pulse length to rf period. Substituting the duty cycle into Eq. (1) yields an expression for our design’s maximum average current:

$$I_{av} = \frac{D e}{t_l} \frac{\Delta E_{\text{in}}}{\Delta K} = I_{pk} D \frac{\Delta E_{\text{in}}}{\Delta K},$$  \hspace{1cm} (2)

where $I_{pk} := e/t_l$ is the peak current at the cathode. Energy-selecting monochromators also show a linear scaling of average current with the fractional reduction in energy spread. The optimal performance of our monochromator is thus equivalent to an an energy-selector with an effective input current of $I_{pk} D$. An estimate of the allowable values of $D$ depends on the details of our monochro-
an initial pulse length that approaches the quantum limit, mator can deliver to an experimental target, we suppose theoretical ceiling on the current that the rf monochromator presented in this paper balances an orthogonal trade-off between temporal resolution and energy selector delivers to the experimental target is a constraint on target with energy selectors because the transverse brightness of the highest-resolution instruments is favorably with the order 1 nA current delivered from CFE sources to state of the art TEM monochromators. There is little scope for future increases in current with peak current in Eq. (2) points to the potential superiority of pulsed beams with lossless monochromation as higher-brightness photoemission sources become available. The two dimensions of active research toward higher-brightness photo-emitters are lower source energy spread and smaller source size. Measurements of photo-emission from cryo-cooled alkali-antimonide photocathodes have shown source energy spreads on the 10 meV scale, an order of magnitude smaller than CFE sources. Photo-emitting tips yield nanometer source sizes, smaller than the diffraction limited laser spot diameter. A hypothetical alternative to a tip geometry is to layer a photoemission mask on planar cathodes, exposing a photoemitting disc with a diameter on the scale of 10 nm. The simulation results we present in Sec. III make practical assumptions about the photoemission source that anticipate future trends. We consider a planar cathode geometry with an RMS source size of 12 nm and initial uniform energy spreads of 0.1, 0.5 and 1 eV. The physics that makes rf monochromation possible, which the next section describes, does not depend

\[ t_i \Delta K = \hbar/2. \]

Then, letting the duty-cycle be \( D = 0.01 \) and the final energy spread 10 meV, the current on target is 20 nA, three orders of magnitude potential improvement.

Both metallic photoemission and cold field emission sources in use today produce energy spreads of multiple hundreds of meV [26, 39–41]. A final spread of 5 meV is an appropriate benchmark, being the resolution required to resolve phonons in EELS and to reduce chromatic aberration in objective lenses by more than an order of magnitude \( A \) target \( \Delta E_{\text{min}} \) of 5 meV from a \( \Delta K = 500 \) meV source with initial pulse length \( t_i = 30 \) fs implies a final pulse length of 3 ps, resonant cavity frequency of \( \sim 3 \) GHz, and an average current of 500 pA. An average current of 500 pA is more than sufficient for imaging above 10 keV primary energy, and cavities and rf sources at 3 GHz are well-explored in both accelerator and time-resolved experimental work. Additionally, with the reduction in laser repetition rate (by pulse-picking, for example) to accommodate sample recovery times in pump probe experiments, 3 ps resolution enables the tracking of phonon population evolution in time [42].

In the applications of interest, the cathode makes the dominant contribution to energy spread, with the sub-leading contribution coming from fluctuations in the accelerating voltage. Our design specifically corrects source energy spread, relying on the correlation between initial kinetic energy and time and position of arrival. Section IV, on jitter, suggests how to incorporate into our design fast feedback from existing beam diagnostic devices so as to compensate sub-leading sources of energy spread.

The advantage of lossless monochromation over energy selectors for time resolved applications is that users can obtain improved energy resolution without paying a cost in average current. For applications that at present obtain the best performance from CW beams, the scaling with peak current in Eq. (2) points to the potential superiority of pulsed beams with lossless monochromation as higher-brightness photoemission sources become available. The two dimensions of active research toward higher-brightness photo-emitters are lower source energy spread and smaller source size. Measurements of photo-emission from cryo-cooled alkali-antimonide photocathodes have shown source energy spreads on the 10 meV scale, an order of magnitude smaller than CFE sources. Photo-emitting tips yield nanometer source sizes, smaller than the diffraction limited laser spot diameter. A hypothetical alternative to a tip geometry is to layer a photoemission mask on planar cathodes, exposing a photoemitting disc with a diameter on the scale of 10 nm. The simulation results we present in Sec. III make practical assumptions about the photoemission source that anticipate future trends. We consider a planar cathode geometry with an RMS source size of 12 nm and initial uniform energy spreads of 0.1, 0.5 and 1 eV. The physics that makes rf monochromation possible, which the next section describes, does not depend.

![Figure 2](image-url)

**FIG. 2.** Normalized energy gain vs single particle time of arrival, expressed as a fraction of the full rf period. The quadratic coefficient of the energy gain is subtracted, leaving only unwanted higher order terms. Vertical lines show the initial pulse length \( t_i \) (solid) and final pulse length \( t_f \) (dashed). The duty cycle in Eq. (2) is \( f t_f \). Inset shows the full sinusoidal energy gain vs scaled time of arrival, with the quadratic term restored.

mator design. As shown in Sec. III, minimising final energy spread requires that the energy gain from the cavities depend quadratically on time. Figure 2 plots the cubic and higher order time dependence of the work done by a single cavity on a logarithmic scale. Inspection of the figure indicates that choosing \( D < 0.1 \) suppresses the higher order contributions to the 1% level or lower. Modern ultrafast laser oscillator sources can provide multiple GHz repetition rates with \(< 20 \) fs pulse durations [37]. For a total emission time of \( t_i = 30 \) fs, and assuming \( D = 0.01 \), we arrive at \( I_{pk} D = 50 \) nA, which compares favorably with the order 1 nA current delivered from CFE sources to state of the art TEM monochromators [26], of like design to the diagram shown in Fig. 1(a).

Constrained by the transverse brightness of the electron source, energy selection in the spatial domain confronts a trade-off between energy and spatial resolution. The 10 pA scale of the maximum current the CFE energy selector delivers to the experimental target is a consequence of balancing the objectives of angstrom scale spatial resolution against 10 meV scale energy resolution [38]. There is little scope for future increases in current on target with energy selectors because the transverse emittance of the highest-resolution instruments is approaching the quantum limit. By contrast, the rf monochromator presented in this paper balances an orthogonal trade-off between temporal resolution and energy resolution, constrained by longitudinal brightness. Hence, holding the pulse length and energy spread at the sample constant, a reduction in the initial pulse length results in higher current on the target. To calculate the theoretical ceiling on the current that the rf monochromator can deliver to an experimental target, we suppose an initial pulse length that approaches the quantum limit,
on assumptions about source quality.

The discussion in this section is completed by considering transverse spatial degrees of freedom. The contribution that the transverse momenta make to total energy spread implies a parallel trade-off between transverse beam-size and energy-spread. Therefore, including the transverse contribution makes possible a reduction in energy spread without a compensating increase in pulse length. A natural mechanism to imagine realizing this possibility is a radial electric field that performs work to reduce the energy stored in the transverse momentum and, as a bi-product, collimates the beam. Our design employs a similar mechanism, conceptually more complicated but simpler to engineer. Our design, after expanding the beam, applies a spatially varying longitudinal field so as to balance an energy surplus in the transverse direction by creating an energy deficit in the longitudinal direction.

Altogether, our design achieves its final energy spread by stretching the beam both transversely and longitudinally. The spatial and temporal trade-offs do not originate from two distinct constraints but are instead both consequences of the conservation of six-dimensional phase space volume. Thus, if the expanded, reduced energy spread beam is again focused down to its source size or smaller by magnetostatic lenses, then the pulse length stretches as the beam size shrinks. Magnetostatic lenses do no work and hence have no effect on energy spread, but produce pulse stretching due to variations in the path lengths traced by particles that are now travelling at much closer to the same velocity. In the applications of our monochromator previewed in the introduction, there is more experimental flexibility to trade longer pulses for lower energy spread than there is to vary beam size at the experimental target. Hence, Eq. (1) formulated only in terms of the longitudinal variables captures the essential physics of our monochromator design.

III. ENERGY EQUALIZATION

A. Correlation between emission energy, arrival time and position

Our monochromator design operates in two steps. First, the system of electron source and optical column correlates energy with time and position of arrival, stretching the electron pulse in time and space. The uncorrelated energy spread decreases as the pulse is stretched. In the second step, the rf fields remove the correlated energy spread.

Correlations between energy, time and space naturally arises near zero energy in the low to moderate extraction fields ($1 - 10$ MV/m) of a dc gun, which our design exploits. Figure 3 shows our simulation gun field map alongside a plot of the position of downstream beamline elements. Illustrative snap-shots of the evolution of energy correlations in simulation are shown in Fig. 4. An alternative method for stretching electron pulses is to insert dedicate device comprising rf cavities and drift space [11, 45]. Our omission of dedicated stretching cavities has the advantage of simplicity. Furthermore, by relying on the gun field to stretch the pulse, our design does not require significant drift space for stretching to develop. In this section, we derive an analytical model of stretching in an idealized gun field and confirm the model’s accuracy in particle tracking simulations.

Two rf cavities correct the kinetic energy spread contained in the time (cavity one) and transverse position (cavity two) degrees of freedom. Both cavities are identical in design, comparable to the device described in [12]. A 3D rendering of our cavity is shown in Fig. 5. This section computes to good accuracy the settings of cavity phases and amplitudes that minimize final energy spread.

Simply knowing the initial kinetic energy of a particle does not uniquely determine its time and position of arrival at the cavities. Instead, we derive the inverse relationship: the initial kinetic energy as a function of the time and position of arrival at the cavities. The exit of the gun serves as the primary reference plane for particle coordinates. Any equipotential plane downstream of the anode would fulfill this role equally well. Our analytic model ignores fringing effects and thus takes the anode to be the gun exit. The initial kinetic energy is non-relativistic and thus proportional to the sum of squares of initial momenta. Our problem thus reduces to expressing each of the three initial momenta separately in terms of their conjugate coordinates at the gun exit.

We begin by deriving the initial longitudinal momen-
Finally, the stopping time of an electron with velocity $v > 1$ keV final energies — the smallest term is the stopping time. If we have $E_z = -1$ MV/m, the stopping time of an electron with velocity $v_z/c = 0.002$ is 3 ps, the ultra-relativistic time of flight across a cathode gap of 1 cm is 30 ps, and the corresponding Newtonian time of flight is 500 ps.

In order to estimate the error that arises from neglecting the stopping time, we can expand the square root in powers of the small quantity

$$\delta = \frac{p_{z0}^2}{e^2 E_z^2} \left( \frac{z^2}{c^2} - \frac{2z\gamma_0 m_e}{e E_z} + \frac{p_{z0}^2}{(e E_z)^2} \right)^{-1},$$

which is proportional to the square of the stopping time. Then, for the example values cited above, the correction term first order in $\delta$ is $\sim 10$ as. The relevant scale to compare is the uncertainty in the time of emission of a single electron packet $t_l$, equal to the the laser pulse length and photoemission response time, which are on the order of 10 fs. Thus, the stopping time can safely be neglected inside the square root. Under the assumption that all initial kinetic energies are of single eV scale or less, it also safe to assume $\gamma_0 = 1$, and no remaining term inside the square root appearing in Eq. (3) depends on the initial conditions. Thus, the square root drops out from the difference in arrival times $\Delta t$ between two particles, and $\Delta t$ becomes linearly proportional to the difference in their initial longitudinal momenta $\Delta p_{z0}$.

$$\Delta t = \frac{\Delta p_{z0}}{e E_z} + \Delta t_0,$$

with $\Delta t_0$ the difference in emission times (relative to the arrival time of the laser pulse).
The most convenient choice of reference particle is the particle with zero initial kinetic energy. The arrival time therefore gives us the $p_{x0}$ of all particles to within a precision set by $t_\ast$, which is 1\% for the example values. By comparison, fluctuations in the accelerating gradient, typically at the $10^{-5}$ level or below, have a negligible effect on time of arrival.

The coefficient of proportionality appearing in Eq. (5) is independent of $z$ and thus the gun length. A dependence on gun length is absent because the relative time of arrival is frozen for $z$ large enough that $\delta \ll 1$, per Eq. (4). The freezing of time of arrival differences makes it possible to generalize Eq (5) to non-uniform accelerating fields by replacing $E_z$ with the photocathode field. The resulting expression is accurate so long as (i) the photocathode field is approximately uniform over a distance such that $\delta \ll 1$ and (ii) the particle velocities are increasing functions of $z$ for the remaining length of the gun.

As for the transverse coordinates, the solution to the equation of motion in $x$ (without transverse focusing) is,

$$x = \frac{p_x}{m_e} \tau + x_0,$$

with $x_0$ the coordinate of the particle at emission and $\tau$ the proper time that elapses between emission of the particle and its crossing the transverse plane at $z$. The coordinate $y$ follows from cylindrical symmetry. Let $\tau_\ast(z)$ be the proper time to reach $z$ of a reference particle (initially at rest). Expanding $\tau$ in $p_{x0}$ around $\tau_\ast(z)$,

$$\tau = \tau_\ast + \frac{d\tau}{dp_{x0}} p_{x0} + O\left(p_{x0}^2\right)$$

$$= \frac{m_e c}{eE_z} \mathcal{A}(\gamma_\ast) + \frac{p_x}{eE_z} + O\left(p_{x0}^2\right).$$

(8)

Here, the variable $\gamma_\ast(z)$ is the relativistic factor for the reference particle and,

$$\mathcal{A}(\gamma_\ast) := \ln (\gamma_\ast) + \ln (1 + \beta_\ast),$$

(9)

with $\beta_\ast(z)c$ the velocity of the reference particle. Dividing Eq. (6) by Eq. (5) shows that $\mathcal{A}$ approximates the aspect ratio $x/c\Delta t$, hence the choice of notation.

To find the initial kinetic energy $K$ as a function of a particle’s position and time of arrival, we simply square both sides of Eq. (5) to obtain $\gamma_\ast^2 = 1 + \beta_\ast^2$ and $x/c\Delta t = \tau_\ast$. By the equation of motion, $x/c\Delta t = \beta_\ast c$, with $\beta_\ast$ the velocity of a particle at time $\tau_\ast$.

Knowing the initial kinetic energy of a particle as a function of the coordinates $\Delta t, x, y$ at the plane $z$, we must derive the matching expression for the work done by the cavity fields on transiting particles. For the purpose of this derivation, we make the following rigid beam approximation, which is exact in the limit that the cavity impulse is small compared to the mean particle momentum. Namely, we assume that particles transit with constant velocity $\beta c$ parallel to the symmetry axis. In this limit, only the longitudinal component of the cavity electric field contributes to the work done. Figure 6 shows the electric field of the TM010 mode supported by our cavity design.

B. Cavity fields

The relative size of this cubic term is,

$$\frac{\partial^3 K}{\partial t \partial r^2} \frac{\Delta t}{(\partial^2 K)/(\partial r^2)} = \frac{2 \gamma_\ast \beta_\ast}{\ln (\gamma_\ast) + \ln (1 + \beta_\ast)} \sim \frac{\beta_\ast}{\beta}.\quad (12)$$

The initial longitudinal particle velocity $\beta_\ast$ is on the order $10^{-3}$ for electron emitted with kinetic energies less than 1 eV. The final velocity $\beta_\ast \approx 0.5$ at an accelerating voltage of 50 kV. Thus, for pulse stretching factors approaching 1000 or more, cubic order correlations put a ceiling on the final energy spread achievable with our two-cavity solution at parts per thousand the initial energy spread.

The longitudinal component of any axially symmetric transverse magnetic mode expands in powers of derivatives of the on axis longitudinal field $E_{0z}$ as,

$$E_z(t, z, r) = J_0 \left[ r \sqrt{\frac{\partial^2}{\partial z^2} + \frac{\omega^2}{c^2}} \right] E_{0z}(z) \cos (\omega t + \phi_0),$$

(13)
where $J_0$ is the zeroth order Bessel function, which for operator arguments is defined by the power series,
\[ J_0 \left[ \frac{\partial}{\partial z} \right] := \sum_{k=0}^{\infty} \left( \frac{-1}{4} \right)^k \frac{1}{(k!)^2} \frac{\partial^{2k}}{\partial z^{2k}}. \]

We define the free-parameter $\phi_0$ such that at $\phi_0 = 0$ the reference particle undergoes the maximum change in energy. Integrating the right-hand-side of Eq. (13) by parts to all orders in the derivative expansion gives the work $W$ as a function of the radial coordinate $r = \sqrt{x^2 + y^2}$ and the time of arrival $t$.

\[ W(r, t) = -\int_{-\infty}^{\infty} eE \cdot ds \approx -\int_{-\infty}^{\infty} eE_z(t(z), z, r) dz \]
\[ = -J_0 \left( \frac{2\pi i r}{\beta \gamma} \right) \cos(\phi) \int_{-\infty}^{\infty} eE_{0,z}(z) \cos \left( \frac{2\pi z}{\beta \lambda} \right) dz, \]

with $\phi = \omega \Delta t + \phi_0$ and $\Delta t$ the difference in time of arrival at the gun exit, the same variable appearing in Eq. (10). Referring the time of arrival to the gun exit in this way is an approximation valid in the limit that drift sections between elements after the anode are short, which is the regime of interest. The integrand on the right-hand-side of Eq. (16) is independent of $r, t$. Hence, neglecting the small spread in $\beta$, it is possible to define an effective cavity length $d_{RF}$ and voltage [12]. Letting $E_{max}$ be the maximum longitudinal field on axis,

\[ d_{RF} := \frac{1}{E_{max}} \int_{-\infty}^{\infty} E_{0,z}(z) \cos \left( \frac{2\pi z}{\beta \lambda} \right) dz. \]

Choosing $\phi_0 = 0$ simplifies the conditions for canceling the quadratic dependence of energy on time appearing in Eq. (10). With $\phi_0 = 0$, the expression for $W$ expands to quadratic order in $r, t$ as,

\[ W(r, t) = -d_{RF} eE_{max} \left( 1 - \frac{\omega^2}{2} \Delta t^2 + \frac{\pi^2}{\beta^2 \gamma^2 \lambda^2} r^2 \right). \]

The term in $r$ enters Eq. (18) with the opposite sign to its counterpart in $t$ because Eq. (13) requires the peak of $E_{z,0}(z)$ to be a saddle-point in three dimensions.

C. Analytic prediction of cavity parameters

By incorporating two cavities in the monochromator, it is possible to make the cumulative energy gain a concave down quadratic function of both time and space. A negative quadratic dependence cancels the positive dependence imprinted at the electron source. This strategy introduces the design problem of optimising beam transport between the cavities. It is conceptually simplest to suppose transfer optics that image the reference plane of the gun exit successively at the mid planes of the two cavities. If the imaging condition is satisfied, the transfer map that relates the particle transverse coordinates at the gun exit to the particle coordinate at a given cavity mid-plane is described by a single parameter, namely, the magnification factor $M$, $x \mapsto Mx$. Let $M_1$ and $M_2$ denote the magnification factors at each respective cavity, and $E_{max,1}, E_{max,2}$ the two cavity amplitudes. It follows from Eqs. (10) and (18) that the difference in energy $\Delta E$ between any particle and the reference particle, referred to the particle coordinates at the plane of the gun exit, splits into two terms:

\[ \Delta E(t, r) = \Delta E(t) + \Delta E(r), \]

where, ignoring terms in $t_0, x_0, y_0$, as well as longitudinal drift,

\[ \Delta E(t) = \left[ \frac{(eE_z)^2}{2m_e} + d_{RF} e (E_{max,1} - E_{max,2}) \omega^2 \right] \Delta t^2, \]

\[ \Delta E(r) = \left[ \frac{(eE_z)^2}{2m_e c^2 A^2} - \frac{\pi^2 d_{RF} e}{\beta^2 \gamma^2 \lambda^2} (E_{max,1} M_1^2 - E_{max,2} M_2^2) \right] r^2. \]

Again, $\beta, \gamma_*, \gamma_*$ are the values for a particle initially at rest. To optimize the cavities for monochromation, we set $\Delta E(t), \Delta E(r)$ to zero, yielding an under-constrained system of equations in the free variables $E_{max,1,2}, M_1, M_2$. That the system is under-constrained suggests the freedom to optimize a second design objective beyond energy spread. The most relevant second objective for microscopy applications is transverse emittance preservation, which we consider below in Sec. V. Though under-constrained, Equation (20) does predict the net energy added to the beam by the monochromator. The net energy added is a self-consistency test on the assumption that particle velocities remain approximately constant as they transit the cavities, which was made in deriving Eq. (21). The total energy gain of the beam is

\[ -ed_{RF} (E_{max,1} - E_{max,2}) = \frac{1}{4\pi^2} \frac{(eE_z \lambda)^2}{m_e c^2}. \]

Given a 3 GHz cavity and an accelerating gradient of 1 MV/m, the total energy added is 500eV. This energy scale indicates that rf amplitude noise at the $10^{-9}$ level or larger makes a non-negligible contribution to the final energy spread.

Equation (20) ignores the uncertainty in emission time $t_0$ set by the laser pulse length and the photocathode response time $t_l$. Non-zero laser pulse length imposes another limit on the lowest achievable energy spread. If we take the laser pulse length into account then at the earliest arrival times the final longitudinal distribution is linearly correlated between the emission time and energy. The associated energy spread induced by non-zero...
emission time is,

$$\Delta E(t) = d_R F \varepsilon (E_{\text{max},1} - E_{\text{max},2}) \omega_0^2 \frac{p_{\text{0}x}}{c E_z} t_0.$$  \hspace{1cm} (23)$$

Substituting the expression for the net energy gain at the optimal cavity parameters in Eq. (22) yields,

$$\Delta E(t) = -\frac{e E_z}{m_e} p_{\text{00}} t_0.$$  \hspace{1cm} (24)$$

Equation (24) shows that the final energy spread is bounded by the longitudinal emittance at the source, i.e., by the product of the spread in initial longitudinal momentum and time of emission. For the residual spread $\Delta E(t)$ to be of the same size as or smaller than the residual $\Delta E(t)$, the source transverse size must be on the scale of single microns, provided beam sizes in the cavities of $\sim 100$ microns. Source sizes for the highest brightness microscopy applications, both pulsed and continuous wave, are on the nanometer scale, and the simulation results presented in Sec. III assume a nanometer scale source. Thus, we can neglect $\Delta E(t)$ in estimating the final energy spread.

D. Simulation results

This section presents simulation results for four sets of initial beam conditions, to elucidate the beam dynamics and indicate the practical utility of our monochromator design. The first three sets are highly idealized and designed to isolate in turn the longitudinal and transverse degrees of freedom (Figs. 7, 8), as well as the effects of cubic and higher order non-linear cavity fields on final energy spread (Fig. 9). The fourth set corresponds to realistic photoemission beam size and momentum spread and demonstrates monochromation from 1 eV to final energy spread on the few meV scale (Fig. 10). The cases considered do not include the effects of timing jitter, which are accounted for in Sec. IV. Simulation beamline elements and field maps are shown in Figs.

Simulating quasi-one dimensional initial beam conditions allows for easy graphical evaluation of the accuracy of Eqs. (20) and (21). The first set of initial conditions is designed to isolate the longitudinal phase space, having a source with vanishing transverse size, no transverse momentum and a laser pulse length of $\Delta t_0 = 30$ fs. Figure 7(a) shows in blue a scatter plot of this beam in energy and time as it exits the gun. The simulated gun field map is a uniform gradient with a total accelerating voltage of 50 kV over a distance of 1 cm. The results in Fig. 7(a) are in close agreement with Eq. (10), shown in the same figure as a white curve. Figure 7(a) shows in red the final distribution of the same beam in energy and time after transiting a single cavity with amplitude chosen according to Eq. (20). The cavity removes the energy spread up to a small residual linear correlation apparent in the figure at negative arrival times. Equation (24) accurately predicts the coefficient of this linear correlation, shown in Fig. 7(a) as a white line, confirming that the cavity restores the longitudinal emittance to its initial value.

Figure 7(b) shows results for the same quasi-one dimensional distribution as Fig. 7(a) but now accelerated in the non-uniform gun field map shown in Fig. 3(b). The field is non-uniform because of the electrode geometry, also shown in Fig. 3(b). The accelerating voltage is 50 kV over a distance of 1 cm. The field on the cathode is 1.8 MV/m, less than half the average gradient in the gun. The prediction of Eq. (10) is again shown by the white curve. In evaluating Eq. (10), $E_z$ is taken to be the field on the cathode. These results show that it is indeed the photocathode field, rather than the voltage or average gradient, which sets the final pulse length and the correlation between time of arrival and energy. The red curve shows the final energy time distribution after transiting a single cavity with amplitude chosen according to Eq. (20), again identifying the accelerating gradient with the field on the cathode, $E_z \mapsto E_z(z = 0)$.

The second set of initial conditions isolates the transverse direction. The beam at the source has non-zero size
arise from thick lens (non-impulsive) effects, as evidenced overestimate the optimal cavity amplitudes, which likely guesses for the optimization algorithm and consistently calculated from Eqs. (20) and (21) provide good initial and phases are optimized numerically. Parameter values alistic fields shown in Figure 6(a). The cavity amplitude the forward hemisphere. The gun is modelled with the re-

and transverse momentum spread but vanishing longitud-

dinal momentum spread, so that all particles are emitted parallel to the photocathode surface. Figure 8 shows in blue the distribution of energy against radial displacement as this beam exits the gun. The prediction of Eq. (10) is shown as a white curve and again agreement is close. In this case, the gun field map is a uniform accelerating gradient, which simplifies the transverse fo-
cusing in the gun. A non-uniform accelerating field re-



cules of Fig. 9 confirms qualitatively the analytic approx-



dition in Eq. (18) for the work done by the cavities on the beam. Stepping through the panels, the initially paraboloid energy surface in Fig. 9(a) is transformed after transit through the first cavity into a hyperboloid in Fig. 9(b). Transit through the second cavity produces a surface of constant energy in Fig. 9(c), up to cubic corre-
cctions of order $10^{-3}$ the initial energy spread, a scale consistent with the expression for the leading cubic cor-
rection in Eq. (11).

Having tested the accuracy of our analytic model with idealized beam distributions, we present the results of simulations with realistic initial conditions. Simulation parameters are shown graphically in Fig. 6. Figure 10 shows the final energy distribution obtained from simulating a source of 12 nm rms transverse radius and a uniform 1 eV energy spread distributed uniformly in solid angle over the forward hemisphere. The two curves in Fig 10 correspond to primary energies of 10 and 50 keV. The final full width at half maximum for both primary energies is 4 meV.

The evolution of energy spread as function of position down the simulated beam-line is shown in Fig. 11(b), alongside pulse length in Fig. 11(c) and (d). Comparing Fig. 11(b) with Fig. 11(c) and (d), the final value of the product $\sigma_E \sigma_t$ is less than the initial value, in apparent contradiction with Eq. (1). The contribution the trans-
verse momenta make to energy spread accounts for the discrepancy, with a significant fraction of the reduction in energy spread being made possible by expanding the transverse size of the beam. The ratio of final to initial transverse size is order $10^3$ compared to the order 10 ratio of final to initial pulse lengths. A system of mag-
etostatic lenses can subsequently demagnify the beam without affecting energy spread and in the process fur-
ther stretch the pulse length. For the initial conditions $\Delta K = 250$ meV shown in Fig. 11, the final product $\sigma_E \sigma_t$ exceeds $\hbar/2$ by only 30%. Even as the quantum limit is approached, summary statistics calculated from classical particle tracking continue to describe the size of the beam envelope [47].

IV. JITTER

Jitter in the phase of the cavities relative to the photo-
emitting laser pulse train contributes to the final energy spread. The study in [12] finds that the scale of phase jitter is set by thermal fluctuations in the length of the cavities. The cavity fundamental frequency varies with changes in the cavity length, thereby shifting the phase difference between the driving oscillator and cavity re-
sponse. In terms of a change in temperature $\Delta T$, the change in phase $\Delta \phi$ is [12],

$$\Delta \phi = -2QK_T \Delta T,$$  (25)
where $Q$ is the unloaded quality factor of the resonator, typically $10^4$ for a normal conducting copper cavity, and $\kappa_T$ is the coefficient of thermal expansion, $1.64 \times 10^{-5}$ K$^{-1}$ for copper. At a temperature stability of 1 mK, the uncertainty in phase is $\Delta \phi = 3.3 \times 10^{-4}$, or 17 fs at 3 GHz. The time averaged uncorrelated energy spread introduced by phase fluctuations is given from expanding the sinusoidal time dependence of the work done by the cavities,

$$\Delta E = -ed_{RF} (E_{max,1} + E_{max,2}) \left( \frac{1}{2} \omega^2 \sigma_{p,0} + \frac{\omega \sigma_{p,0}}{eE_2} \Delta \phi \right),$$

with $\sigma_{p,0}$ the spread in initial longitudinal momenta and $E_2$ the accelerating gradient on the cathode. The factor containing $p_{z,0}$ is the estimate of pulse length obtained from Eq. (5). For particles arriving earlier than average, the linear term in $\Delta \phi$ dominates because pulses are 100 fs to single picoseconds long for accelerating gradients below 10 MV/m at an initial energy spread of 1 eV or more. However, the statistical distribution peaks at late arrival times, if the particle ensemble has an initially uniform distribution in energy. For particles near this peak, the term quadratic in $\Delta \phi$ dominates the right-hand-side of Eq. (26). The takeaway from Eq. (26) is thus that phase fluctuations move the tails of the energy distribution much more than they do the location of the peak, by a factor 10 to 100.

At a phase uncertainty of $\Delta \phi = 3.3 \times 10^{-4}$, Eq. (26) predicts a broadening near the peak of energy probability distribution of less than one meV and a broadening in the tails of tens of meV. The pre-factor in Eq. (26) is 7 keV for our simulation example of a 50 kV gun with non-uniform gradient shown in Fig. 6 and 1 keV for the 10 kV example. These estimates are confirmed in simulation as shown in Fig. 12. The simulations cycle through a range of phase offsets $\Delta \phi_1, \Delta \phi_2$, one independent offset for each of the two cavities. For each pair of phase offsets, the right panel of Fig. 12 shows a scatter plot in arrival time and final energy of particles in the beam. Comparison of the scatter plots supports our interpretation of Eq. (26), indicating in particular that the jitter-induced movement in the tails of the final energy distribution is due to early arriving particles. The left panel of Fig. 12 shows the corresponding sequence of final energy histograms. On inspection, the jittery peak locations remain within order 1 meV the nominal peak location at $\Delta E = 0$.

Fluctuations in gun voltage are uncorrelated with time of arrival at the relevant order of precision, according to Eq. (3). Energy spread due to these fluctuations is therefore not removed by our rf monochromator design. One strategy for eliminating this subleading source of energy spread is to implement a feedback loop, such as is included in the magnetic prism monochromator design [31]. Our implementation would place an energy selector downstream of the cavities with a slit width chosen so that all particles are accepted at the nominal accelerating
FIG. 11. Simulation results showing beam sizes and emittance as a function of beam-line position for three different initial energy spreads; all particle ensembles have initial transverse size of 12 nm R.M.S. and momenta distributed uniformly over the forward hemisphere; in red, initial uniform energy spread of 1 eV, in blue, 500 meV, and in yellow, 100 meV. Cavity settings are chosen to achieve smallest final energy spread. (a) Transverse normalized emittance, (b) rms energy spread, (c) and (d) rms pulse duration.
FIG. 12. Simulations of cavity timing jitter. The initial particle ensemble has a uniform 1 eV energy spread distributed uniformly in solid angle over the forward hemisphere. Plots show the ensemble after transiting two 3 GHz cavities with timing offset from the optimal solution by φ₁, φ₂ respectively. Offsets scanned add in quadrature to 10 fs. At left, histograms of final energy show single meV movement in peak location and 10 meV scale movement in tails. On the right energy-time correlation are shown as a function of cavity phase, where different phases are offset on the time axis for clarity.
voltage. As the accelerating voltage changes, particles clipping the slit edge trigger the feedback mechanism to change the gun high-voltage set-point.

V. BRIGHTNESS CONSERVATION

To achieve maximum gains in average current, our design must omit transverse collimation. The ensemble of particles transported to the experimental target consequently includes large excursions from the optical axis, which are typically ignored in electron-microscope design. Brightness averaged over all emission angles and positions is therefore the more informative figure of merit for our unconventional beam, rather than the peak brightness more commonly encountered in a microscopy context. Our preferred figure of merit is the phase space brightness averaged over all emission angles occupied by the beam, which is inversely proportional to the square root of the brightness. The most general measure of phase space area is \( \text{normalized transverse emittance} \), defined as:

\[
\epsilon_{n,x} = \frac{1}{m_ec} \sqrt{\langle x^2 \rangle (p_x^2) - \langle xp_x \rangle^2}. \tag{27}
\]

Minimum physically achievable emittance corresponds to a normalized emittance equal to half the reduced Compton wavelength of the electron. At the source, the cross term in \( x \) and \( p_x \) drops out of Eq. (27) and it is convenient to define a new quantity, the mean transverse energy (MTE):

\[
\text{MTE} = \frac{\langle p_x^2 \rangle}{m_e}. \tag{28}
\]

For a statistical distribution that is uniform in energy and uniformly distributed in solid angle over the forward hemisphere, the MTE is equal to \( 2/3 \) the mean energy. Letting \( \sigma_{x0} \) be the rms source size, the source emittance is then,

\[
\epsilon_{n,x}(z = 0) = \sigma_{x0} \sqrt{\frac{\text{MTE}}{m_e c^2}}. \tag{29}
\]

Our simulations show significant emittance degradation occurring just after emission for nanometer-sized sources with energy spreads on the 100 meV scale. Surprisingly, we find that the action of the monochromator largely undoes this emittance growth. The emittance growth we observe can be understood as the contribution of the uniform accelerating field to the spherical and chromatic aberration of the optical column [41], which amounts to a series expansion of the emittance around a vanishing solid angle. However, since our design is unlike a conventional microscope, this section explicitly derives time-of-arrival dependent expressions for the emittance growth valid at all emission angles. These expressions then predict that, up to the accuracy of the analytical model of energy-equalization presented in Sec. III, the final emittance after transiting the monochromator cavities is equal to the source emittance. Simulation results reported in Fig. 11 show that, beyond the rigid-beam approximation, parameters optimised for energy spread reduction over-correct the emittance loss, imparting to the beam a correlation between time-of-arrival and divergence that is opposite in sign to the gun.

During acceleration of a pulsed beam, correlations evolve between time of arrival and beam divergence, so that projecting onto the transverse phase-space results in brightness loss. Starting from the solution to the equations of motion in \( x \) for a uniform accelerating field, the transverse emittance at proper time \( \tau \) is,

\[
\epsilon_{n,x}^2 = \frac{\langle x_0^2 \rangle (p_{x0})^2 + \langle p_{x0}^2 \rangle \langle p_{x0} \tau \rangle^2}{m_e c^2} - \frac{\langle p_{x0} \tau \rangle^2}{m_e c^2}. \tag{30}
\]

The simplest physically plausible picture of emission from a flat cathode is that the initial momentum are uniformly distributed in solid angle over the forward hemisphere, implying a correlation between \( p_{x0} \) and \( p_{z0} \) and hence between \( p_{z0} \) and \( \tau \). These correlations are better disentangled by going over to polar coordinates,

\[
p_{x0} = p_0 \sin \phi \cos \theta, \tag{31}
p_{z0} = p_0 \cos \phi \cos \theta, \tag{32}
\]

and assuming that the momentum magnitude \( p \) is uncorrelated with the emission angles \( \phi, \theta \). Letting the probability of emission be uniform in azimuth \( \phi \), expanding \( \tau \) to first order in \( \Delta \tau \) per Eq. (8) and substituting the expression for \( \Delta \tau \) in terms of \( \Delta p_{z0} \) given by Eq. (5), the result is that Eq. (30) becomes,

\[
\epsilon_{n,x}^2 = \frac{\langle x_0^2 \rangle}{3} \left( \frac{\langle x_0^2 \rangle}{3} + \sigma_{x0}^2 \right), \tag{33}
\]

with

\[
\sigma_{x0}^2 = \frac{m_e c^4}{E_z} \left( \frac{1}{15} \langle \beta_0^4 \rangle - \frac{3}{64} \langle \beta_0^6 \rangle \right). \tag{34}
\]

where, again, \( E_z \) is the longitudinal field in the gun. The term \( \sigma_{x0}^2 \) is a new, critical feature of near-cathode dynamics for nanometer scale photoemission sources: an effective source size growth arising from photoemission momentum spread. The scaling of this effective source size with mean transverse energy has practical importance for active research toward higher brightness photocathodes. Assuming a uniform distribution in energy at the source, Eq. (34) simplifies to,

\[
\sigma_{x0} = 0.363 \times \frac{\langle \text{MTE} \rangle}{eE_z}. \tag{35}
\]

Thus, for nano-scale sources, photocathode emittance goes like MTE\(^{3/2}\) and not the expected MTE\(^{1/2}\), enhancing the potential impact of new low MTE materials on the future performance of photo-emitters [44].

To see that the emittance growth expressed in Eq. (33) can be undone by a time dependent lens, we invert the
The transverse focusing power of a cavity, calculated by making the same thin-lens approximation assumed in the derivation of Eq. (18), is [12],

$$f_{1,2}^{-1} = \frac{eE_{\text{max},1,2}d_{RF}}{2\gamma^3_3b_3^3m_ec^3}\omega^2 \Delta t,$$

(39)

where the cavity phase is taken to maximize (in magnitude) the reference particle.

If the gun exit is successively imaged at the midplanes of the two cavities, with magnification factors $M_1$ and $M_2$, then the condition for cancellation of the emittance growth is,

$$\frac{1}{f_{\text{gun}}} + \frac{M_1^2}{f_1} + \frac{M_2^2}{f_2} = 0.$$ 

(40)

The derivation of Eq. (40) assumes the pulse length remains constant after exiting the gun, a good approximation for the compact beam-line shown in Fig. 6(a). Substituting the expressions in Eqs. (37), (39) into into Eq. (40) gives a third equation on the system of monochromator parameters $E_{\text{max},1,2}, M_1, M_{\text{max},2}, M_2$,

$$ed_{RF}E_{\text{max},1}M_1^2 - ed_{RF}E_{\text{max},2}M_2^2 = \frac{(eE_{\gamma,\lambda})^2}{2\pi^2} \frac{\gamma^2_3b^2_3}{m_ec^4A^2}.$$ 

(41)

where $A$ is again the aspect ratio defined in Eq. (9). Equation (41) is equivalent to the equation obtained by setting the energy spread to zero in Eq. (21).

Simulation results presented in Fig. 11 show that cavity parameters optimal for reducing energy spread actually over-correct the brightness loss in the gun. Simulated particle tracks show that this over-correction is due to particles not obeying the rigid-beam approximation made in deriving Eq. (21). Thus, there is a trade-off between energy-spread-reduction and brightness conservation in a scheme involving only two cavities. Simulations show that, at the cost of reintroducing energy spread at the $10^{-1}$ level compared to the source, reducing the second cavity amplitude does perfectly restore the source emittance. Figure 11 shows simulation results for three values of the source energy spread: uniform distributions over 1 eV, 500 meV and 100 meV. The trend in energy reveals that at 250 meV and below, the excess final emittance at the minimum achievable final energy spread exceeds the source emittance by less than 30%.

VI. DISCUSSION AND CONCLUSION

This paper has shown in simulation the feasibility of a high-energy resolution, pulsed electron source design. The design utilizes radio-frequency accelerating cavities to top-off the primary energy delivered by a DC gun. Relying on correlations between energy on the one hand and time and position of arrival at the cavities on the other, the additional cavity acceleration equalizes energies in the particle ensemble. An initial energy spread of
1 eV is reduced in simulation to 4 meV at primary energies of 10 kV and 50 kV assuming arbitrary precision in cavity timing. Simulating the effect of cavity jitter at 10 fs timing precision shows the peak of the energy distribution broadening on the single meV scale. The efficacy of the design is explicable by simple yet accurate analytic expressions, strongly suggesting that our results are not sensitive to peculiar choices of simulation parameters and that the scheme could be realized experimentally with presently available technology.

A near at hand application of our design is to install two rf cavities in an existing ultra-fast electron transmission microscope. Existing UEM instruments photo-emit from nanometer scale tips, where significant field enhancement occurs. Typical gradients are 1 GeV/m at nanometer distances. The importance of field enhancement for our scheme is that the rapid acceleration of particles escaping the field-enhanced region implies a shorter final pulse length and hence a larger final energy spread, following Eq. (1). Strategies to compensate for this unwanted side-effect of field enhancement include: reducing the extraction voltage, increasing the tip radius, and introducing an extended drift space immediately following extraction.

Our results readily extend to monochromation at primary energies of hundreds of keV. At higher primary energies, the rigid beam approximation made in our analytic treatment of the cavities becomes more accurate. Conversely, the rigid beam approximation breaks down as the primary energy approaches zero. The error in the approximation becomes significant when the transit time through a single cavity is comparable to the cavity period. For a 3 GHz cavity with a 1 cm gap, the ratio of transit time to rf period is unity at a primary energy of 2.6 keV. Exploring cavity monochromation at primary energies below 3 keV thus requires a non-impulsive treatment of the work done by the cavities.

This paper also investigated, analytically and numerically, the effect of the rf cavities on transverse brightness. Fortuitously, time-dependent cavity lensing undoes brightness loss in the gun. Effects apparent in simulation that go beyond the impulsive approximation made in our analytic treatment result in the cavities over-correcting the brightness loss. Simulations show that the over-correction becomes negligible at source energy spreads of 250 meV and below, a finding that underscores the importance for future electron-beam technologies of ongoing research into photo-cathode materials with low intrinsic energy spreads [44].

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