Computational and experimental characterization of high-brightness beams for femtosecond electron imaging and spectroscopy

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Computational and experimental characterization of high-brightness beams for femtosecond electron imaging and spectroscopy

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Using a multilevel fast multipole method, coupled with the shadow imaging of femtosecond photoelectron pulses for validation, we quantitatively elucidate the photocathode, space charge, and virtual cathode physics, which fundamentally limit the spatiotemporal and spectroscopic resolution and throughput of ultrafast electron microscope (UEM) systems. We present a simple microscopic description to capture the nonlinear beam dynamics based on a two-fluid picture and elucidate an unexpected dominant role of image potential pinning in accelerating the emittance growth process. These calculations set theoretical limits on the performance of UEM systems and provide useful guides for photocathode design for high-brightness electron beam systems.

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High-resolution transmission electron microscopy (TEM) using coherent electron beams from field emission guns (FEGs), along with aberration correction, has enabled the atomic resolution imaging of complex materials. More recently, advances in developing femtosecond (fs) photoemitters and retrofitting them into diffraction and microscopy systems has further opened up the prospects of multi-functional TEMs.

The simulation is directly compared with ultrafast shadow imaging experiments designed to characterize the photoelectron dynamics in the same regime, in order to validate the ability of the MLFMM approach to faithfully reproduce nonlinear beam dynamics without resorting to fitting parameters. We provide a microscopic description of the virtual cathode formation as driven by instabilities and image potential that promote a dramatic emittance growth and current limitation. We show that the transient phase space structure of the electron bunches is well described as a two-fluid system with a dominant ellipsoidal component undergoing laminar flow under moderately high acceleration fields, while a smaller turbulent component is determined by the initial phase space structure. This unique two-fluid characteristic subtly controlled by the source geometry and initial conditions provides a handle for controlling the emittance growth at high-intensity beam generation without significantly sacrificing the spatial and temporal-spectroscopic resolutions.

As a grid-free approach, MLFMM treats the local interaction in a natural way without artificial smoothing, allowing crucial handling of the nonlinear dynamics near the photocathode where the electron beam is at low energy and high density. During this period, the electron density distributions are most sensitive to the initial phase space, which are probed directly using the ultrafast shadow imaging techniques as shown in Fig. 1, providing the crucial validation for MLFMM (see Ref. 11 for experimental details). In Fig. 1(a), the longitudinal density profiles (circles at 50 and 80 ps) extracted from the shadow images evidence the accelerated expansion driven by space-charge fields at $N_p = 1 \times 10^8$, and are compared with the MLFMM simulations reproduced under the same conditions. In the simulation, we use the three-step model for creating the photoelectrons, in which we set the Fermi energy $5$ eV, and work function $4.45$ eV to...
model the gold photocathode, and photon energy 4.66 eV. The laser pulses are Gaussian spatially and temporally with standard deviations: \( \sigma_x = 91 \mu m \) and \( \sigma_z = 21 \) fs, respectively, according to the experiments. A fine time step of 0.06\( \sigma_t \) is used for the initial period of 6\( \sigma_t \) during photoemission, whereas, in the following 120 ps of the beam dynamics, the step size is progressively increased up to 0.5 ps. Macroparticles are used in simulating \( N_e > 10^6 \) after checking with the corresponding N-particle simulations for consistency. The dynamics equations are solved by the fourth order Runge-Kutta integrator.\(^{19}\)

The agreement between the simulation and experiment is excellent for the leading portion of the beams. In the trailing portion close to the cathode (Fig. 1(a), shaded regions), the charge density is not fully accessible experimentally due to strong surface scattering of the probe beam.\(^{11}\) MLFMM shows that near the surface the charge density deviates from Gaussian, reflecting the formation of VC. Previously, to understand the space-charge-led expansion, shadow imaging experiments indicated an unusual universality in sub-linear scaling of bunch longitudinal size \( \sigma_z \) versus number of electrons emitted \( (N_e^{emit}) \) taken at 120 ps from MLFMM simulations (left panel), and compared to the shadow imaging data (right panel).\(^{11}\) \( N_e^{emit} \) versus the number of generated electrons \( (N_e^0) \) for various extraction fields \( (F_a) \), showing evidence of virtual cathode formation. Inset: Threshold number of electrons \( (N_e^{crit}) \) for virtual cathode formation as a function of \( F_a \).

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self-field-driven beam expansion can be fully corrected by deploying appropriate beam transport optics, including compression and aberration corrections, to achieve the desired resolution at the target. Reducing the Boersch effect at beam generation is, therefore, critical for enhancing performance of high-brightness UEM designs.

We analyze the nature of the coexisting two fluids (turbulent and laminar) observed in the space-charge-dominated beams. The transverse charge density profiles of the beams generated at \( F_a = 0.32, 1, \) and 10 MV/m are extracted and fitted with a linear combination of Gaussian (G) and uniform-filled ellipsoidal (E) functions, as depicted in Fig. 2(e). At \( F_a = 10 \text{ MV/m} \), the laminar beam is perfectly described by the ellipsoidal function, signifying the formation of the 2D ellipse at this stage. In contrast, at \( F_a = 0.32 \) and 1 MV/m, the flow is not fully laminar, and the nonlaminar portion can be well described by a Gaussian with extended tails representative of the Maxwellian-like (thermal) population, which coexists with the 2D ellipse as evidenced by the fits. The predominance of the 2D ellipse, which accounts for more than 65% of the electrons even under the lowest \( F_a \), is somewhat surprising, as ellipsoidal beam formation fully driven by self-fields is expected only at the zero surface charge limit, i.e., \( F_s \ll F_a \) (Refs. 22 and 23).

We further examine the scaling of the normalized rms emittance versus charge filling, namely, transverse emittance \( \epsilon_x(N_{\text{emit}}) \) and longitudinal emittance \( \epsilon_z(N_{\text{emit}}) \). We consider two image charge models where the first model considers pinned surface charges and the second treats the image charges as fully responsive to the motion of the electrons, providing reasonable limiting cases for the response dynamics of the image charges. Fig. 3 depicts the emittance at 120 ps calculated for various \( N_{\text{emit}} \) and \( F_a \) and for the image charge pinning scenario. In \( \epsilon_x(N_{\text{emit}}) \) [Fig. 3(a)], we can clearly see a strong increase of \( \epsilon_x \) after \( N_{\text{crit}} \) is reached in all \( F_a \) data, evidencing the VC-driven \( \epsilon_x \) growth. When the image charges are pinned, \( \epsilon_x \) growth is nearly doubled, but \( N_{\text{crit}} \) is the same (see comparison in the \( F_a = 0.32 \text{ MV/m} \) cases). The inset of Fig. 3(a) shows the temporal evolutions of \( \epsilon_x \) at \( F_a = 0.32 \) and 1.0 MV/m (all with \( 10^7 \) electrons), where generally \( \epsilon_x \) reaches a steady state after 40 ps. The region exhibiting emittance-growth-free dynamics is well correlated with the region where the 2D elliptical pulse structure dominates. In contrast, in \( \epsilon_z(N_{\text{emit}}) \), as presented in Fig. 3(b), we see little or no VC effect at play as increases in \( \epsilon_z \) seem to follow a universal trend, uninterrupted by the VC formation. This insensitivity to external field settings is an indicator that with pancake bunch generation the longitudinal emittance is primarily driven by the strong internal longitudinal fields and nonlinearities. This is particularly evident from the very similar \( \epsilon_z(t) \) calculated for \( N_{\text{emit}} = 10^7 \) under three very different external field values (\( F_s \)), as shown in the inset.

The quantitative emittance scalings elucidated here provide some useful guides for the design of high-brightness UEMs. First, the beam’s emittance fundamentally sets the theoretical resolution limits. The coherence length \( L_t \) is constrained by the parameter \( L_t \leq \sigma_{\epsilon_0}/2\epsilon_0 \), where \( \sigma_{\epsilon_0} = h/(m_0c) \) is the emittance quantum, \( h \) is Planck’s constant, and \( \sigma_{\epsilon_0} \) is the electron beam radius. The temporal (\( \Delta t \)) and energy (\( \Delta E \)) resolutions are constrained by \( \epsilon_\gamma: \epsilon_\gamma \leq \Delta \Lambda E/(\gamma m_0 c) \), where \( \gamma \) is
emittance is established with a cold FEG having a 6D emittance volume of $10^{-11} \mu m^3$ or $\eta \sim 10^{-3}$ [$\epsilon_x \sim 1 nm$ and $\epsilon_y \sim 10 pm$ (statistical)]. In contrast, a flat metallic photocathode widely used in ultrafast electron diffraction has a degraded $\epsilon_x \sim 20 - 200 nm$ [Fig. 3(a)]. However, the extended sources driven in the pancake regime have the emittance volume $\epsilon_x \epsilon_y \epsilon_z$ scale favorably, leading to a gain in $\eta$ for electron bunches up to $N_{\text{emitt}}^{\text{crit}} = 10^3$ for a typical laser pulse size. Using the highest acceleration fields, e.g., $F_a = 100 MV/m$ in DC guns or $F_a = 10 MV/m$ in RF guns, does not improve $\eta$ or $\epsilon$, but instead increases $N_{\text{emitt}}^{\text{crit}}$ without substantively compromising performance. In these high charge limits, the degeneracy figures are comparable to FEGs, implying that similar performance (with proper phase space manipulation) could be reached using a portion of the transverse emittance volume, for example by using apertures, whereas in the single-shot limit performance equivalent to those of thermionic guns is expected.

However, $\epsilon_z$ does not scale as favorably as $\epsilon_x$, leading to some reduction in time and energy resolution. Increasing $N_{\text{emitt}}^{\text{crit}}$ always leads to an $\epsilon_z$ increase [almost linearly, see Fig. 3(b)]. To achieve combined 100 fs–1 eV resolution no more than $N_{\text{emitt}}^{\text{crit}} = 10^3$ can be deployed based on $\Delta E \geq \epsilon_z \gamma \mu c / \Delta t$. In comparison, there is no gain in multielectron mode using sharp emitters (FEGs or atom-sized emitters) due to their poor emittance scaling with $N_e$ (as in the charge-pinning scenario). For example, when reducing the radius of the emitting area from 100 $\mu m$ to 10 $\mu m$, the emittance is initially reduced but scales unfavorably so that there is a crossover at about 100 fs after which the 6D emittance for the smaller emitter become greater. The VC regime also sets in at a lower value of $N_e^0$, leading to nearly two orders of magnitude fewer emitted electrons. Beyond optimizing the cathode geometry, laser pulse shaping of initial bunches from Gaussian to ellipsoidal can help reduce the emittance by nearly a factor of 2, moreover design of sources with lower initial thermal emittance and appropriate emittance compensation will further improve the performance.

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FIG. 3. (a) Transverse emittance $\epsilon_x$ dependence on number of emitted electrons ($N_{\text{emitt}}^{\text{crit}}$) and extraction field $F_x$. (b) Longitudinal emittance $\epsilon_z$ dependence on $N_{\text{emitt}}^{\text{crit}}$ and $F_z$ calculated at 120 ps. The insets in the panels show the time dependence of $\epsilon_x$ and $\epsilon_z$ for three selected cases at $N_{\text{emitt}}^{\text{crit}} = 10^3$. The shaded regions depict the trend for linear emittance growth.

The relativistic Lorentz factor. Lower emittance is also favored for reducing the aberrations and consequently improving the phase contrast in microscopy. In considering the sensitivity of UEMs, the emittance is linked to the 6D beam brightness: $B_{6D} = N_e / (\epsilon_x \epsilon_y \epsilon_z)$, or the degeneracy $\eta = B_{6D} \epsilon_z^3$ by normalizing $B_{6D}$ to the emittance quantum. The degeneracy for different operational regimes is depicted in Fig. 4, where the theoretical limit of $\eta$ is 2, limited by Pauli exclusion (considering both spins). One measure of improved performance is the increase in $\eta$ by increasing $N_{\text{emitt}}^{\text{crit}}$ until the emittance volume $\epsilon_x \epsilon_y \epsilon_z$ starts to increase rapidly due to VC effects. In conventional TEM, the lowest

FIG. 4. The 6D emittance $\epsilon_x \epsilon_y \epsilon_z$ versus the number of emitted electrons $N_{\text{emitt}}^{\text{crit}}$ for the extended electron sources with sizes $s$ (100 $\mu m$, 1 mm), thermionic guns, Schottky, cold field-emission guns (CFEG), and heated field-emission guns (HFEG).