## Effects of Electron-Gun and Laser Parameters on Collection Efficiency and Packet Duration in Ultrafast Electron Microscopy

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Real-space imaging of full-morphological, angstrom-scale structural dynamics occurring on the femtosecond (fs) timescale is possible with ultrafast electron microscopy (UEM) via the stroboscopic pump-probe methodology enabled by interfacing an otherwise standard TEM with a short-pulsed laser system [1-3]. The technology has matured to the point that commercial systems, such as the FEI Tecnai Femto, are now available [4]. The base platform for the Tecnai Femto is FEI's Tecnai G2 20 200 kV instrument with a thermionic electron gun. In UEM mode, the LaB<sub>6</sub> is operated cold such that electron emission occurs only during photo-illumination. Despite this, nanosecond single-shot imaging and diffraction – wherein >10<sup>7</sup> electrons per packet reach the specimen – are possible with a Tecnai-based UEM equipped with a conventional thermionic electron gun [5,6]. While this illustrates that sufficient photoelectrons for such experiments are generated in an otherwise unmodified Tecnai TEM, the parameter space for application-specific optimal electron-gun and photoelectron-generating laser properties remains ill-defined. Quantitatively and systematically mapping such parameter space is challenging due to the large number of variables affecting photoelectron packet properties [7-9].

Here, we describe the effects of thermionic gun configuration (e.g., Wehnelt aperture diameter and relative position of the LaB<sub>6</sub> emission source) and photoelectron-generating laser-pulse duration on electron collection efficiency (CE) and temporal duration ( $\Delta t_{50}$ ), respectively. Specifically, we use General Particle Tracer (GPT) code [10] to simulate the evolution of fs photoelectron packets generated from a cold LaB<sub>6</sub> source in an otherwise standard Wehnelt assembly, as is the case for UEM mode in the Tecnai Femto instrument. Considered in the simulations were the effects of electron path lengths, energy spread, and Coulombic interactions, and we tracked the temporal and energy spreads, brightness, and CE as a function of various parameters. Properties of the photoelectron-generating laser pulse (e.g., spot size on the LaB<sub>6</sub>, duration, and energy) and geometry of the gun (e.g., relative axial LaB<sub>6</sub> position and Wehnelt aperture diameter) were systematically varied in order to investigate the resulting electron packet properties. Three discrete UEM modes were considered: single electron (1 electron/packet), burst (10<sup>3</sup> electrons/packet), and single shot (10<sup>7</sup> electrons/packet).

Results of the simulations suggest that there exists an optimal  $LaB_6$  position relative to the Wehnelt aperture wherein a CE of 100% can be achieved for the single-electron mode and, additionally, that CE increases approximately linearly up to and beyond 90% for both burst and single-shot modes with increasing Wehnelt aperture diameter. Moreover, we found that only marginal increases in the overall temporal resolution can be achieved by decreasing the photoelectron-generating laser-pulse duration from 280 to 10 fs owing to evolution of the packet properties in the gun region and the acceleration tube. The goal of these simulations is to provide a starting point for experimentally mapping parameter space accessible with UEM instruments based on otherwise standard self-biasing thermionic electron guns while also

providing insight into challenges associated with achieving high temporal and spatial resolutions [11].

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- [11] This work was partially supported by the DOE Advanced Research Projects Agency-Energy (ARPA-E) under Contract Number 0472-1595 and in part by a 3M Nontenured Faculty Award under Award Number 13673369. Acknowledgment is made to the Donors of the American Chemical Society Petroleum Research Fund for partial support of this research under Award Number 53116-DNI7.



**Figure 1.** (a) Schematic of the thermionic gun showing the Wehnelt cylinder and LaB<sub>6</sub> with aperture diameter ( $D_{Weh}$ ) and gun-tip height ( $Z_{tip}$ ) defined. (b) Schematic of the thermionic gun geometry in the Tecnai Femto with pertinent components labeled and collection efficiency (CE) illustrated. (c) Effect of  $Z_{tip}$  position on CE for the single-electron and burst modes for  $D_{Weh} = 0.7$  mm. (d) Effect of  $D_{Weh}$  at optimal  $Z_{tip}$  position on CE for each mode studied. (e) Effect of photoelectron-generating laser-pulse duration (10, 80, and 280 fs) on temporal resolution ( $\Delta t_{50}$ ) as a function of  $Z_{tip}$  position.