ISBN: 978-3-95450-232-5 ISSN: 2673-7000 doi:10.18429/JACoW-NAPAC2022-TUXD6

DUAL RADIOFREQUENCY CAVITY BASED MONOCHROMATIZATION FOR HIGH RESOLUTION ELECTRON ENERGY LOSS SPECTROSCOPY*

A. Kulkarni†, P. Denham‡, A. Kogar, and P. Musumeci.
Department of Physics and Astronomy, University of California, Los Angeles, CA, USA

Abstract

Reducing the energy spread of electron beams can enable breakthrough advances in electron energy loss spectroscopy (EELS) investigations of solid state samples where characteristic excitations typically have energy scales on the order of meV. In conventional electron sources the energy spread is limited by the emission process and typically on the order of a fraction of an eV. State-of-the-art energy resolution can only be achieved after significant losses in the monochromatization process. Here we propose to take advantage of photoemission from ultrashort laser pulses (~ 40 fs) so that after a longitudinal phase space manipulation that trades pulse duration for energy spread, the energy spread can be reduced by more than one order of magnitude. The scheme uses two RF cavities to accomplish this goal and can be implemented on a relatively short (~ 1 m) beamline. Analytical predictions and results of 3D self consistent beam dynamics simulations are presented to support the findings.

INTRODUCTION

In conventional EELS instruments, fine energy resolution is obtained after a monochromator rids the initial beam of most of its electrons, as the initial energy spread from the cathode is typically on the order of 1 eV. After monochromatization, the beam can approach energy scales in the meV range, which are of most relevance to condensed matter systems [1]. For continuous beams, the main contribution to the spread in energies of the electrons is the initial spread in energies at the cathode coupled with the fluctuations in the high voltage power supply. If the initial beam is pulsed, the longitudinal phase space of the beam can be manipulated to trade off bunch length for energy spread as proposed in Duncan et al. [2]. As discussed in that paper, the limit in energy spread reduction is then set by the Liouville theorem-based requirement that the final longitudinal phase space area must be conserved. It follows then that if we could stretch the beam by 100 times in length, it would be possible to reduce the initial energy spread by a factor of 100 without any current loss. This approach is closely related to the work of Zeitler et al. [3] to reduce to eV levels the energy spread in relativistic RF photoinjectors beamlines.

In this paper we discuss a pulsed transmission electron energy loss spectroscopy (EELS) scheme which studies the implementation of this transformation for non-relativistic 40-100 keV table-top setups enabling the possibility to access meV-scale energy resolution and outperform current EELS instruments by orders of magnitude in terms of beam brightness, a highly desirable result for time-resolved EELS [4]. The scheme is based on the use of a very short (40 fs full width half maximum -FWHM) laser pulse to emit a short burst of electrons from a flat photocathode in a high-tension DC gun. Two properly phased radiofrequency (RF) cavities are then used as a temporally magnifying telescope to stretch and collimate the electron bunch by nearly two orders of magnitude to 5-10 ps, while at the same time proportionally reduce its energy spread. Compared to the original approach presented in Duncan et al. [2] where there is a strong coupling between the transverse and longitudinal dynamics imprinted by the nanoscale dimensions of the electron emitter, here the flat photocathode allows to decouple the beam size evolution and to consider sizable electron charges in each bunch. The simplification also makes possible to analytically describe the longitudinal dynamics, uncovering the scaling laws and limits in energy spread reduction.

THEORY

A cartoon of the proposed beamline is depicted in Fig. 1. Therein, we suppose that a short (40 fs) electron bunch (< 5000e potentially down to a single electron per pulse) is emitted by a DC 40 keV photo-injector. The beam is then accelerated down stream through two TM010-like cavities. The first cavity is set to act as a temporal defocusing lens which stretches the beam. According to Liouville theorem, a temporally stretching beam must simultaneously have its intrinsic energy spread compressed to conserve phase space area, a shown in the cartoon as the thickness of the longitudinal phase space ellipse decreases after the first cavity. As the beam exits the first cavity, the longitudinal phase space exhibits a positive correlation between position and energy. During the drift to the second cavity, the pulse stretches in time, meanwhile the uncorrelated energy spread (the thickness of the longitudinal phase space ellipse) decreases. The beam finally enters the second cavity at the correct phase to fully compensate the positive chirp with the end result of a significantly reduced energy spread.

We start by assuming that we can neglect transverse effects, focusing only on longitudinal single particle dynamics. In addition, we will ignore for now space charge effects (which will always be true in the limit of single-electron pulses). For an on-axis particle the effect of a cavity can be simply described by a sinusoidal change in its kinetic energy depending on the phase experienced by the particle. The energy gain imparted by the first cavity as a function of

* Work supported by National Science Foundation under Grant No. DMR-1548924.
† athkulkarni@gmail.com
‡ pdenham@physics.ucla.edu

ISBN: 978-3-95450-232-5 ISSN: 2673-7000 doi:10.18429/JACoW-NAPAC2022-TUXD6

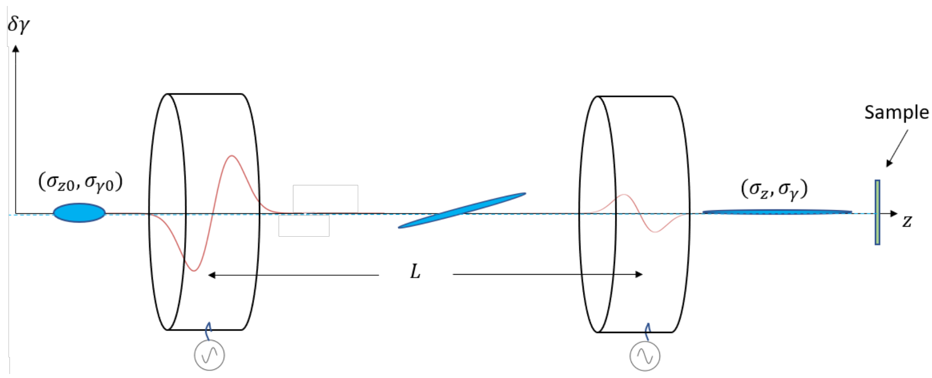

Figure 1: Cartoon depiction of the compensation scheme.

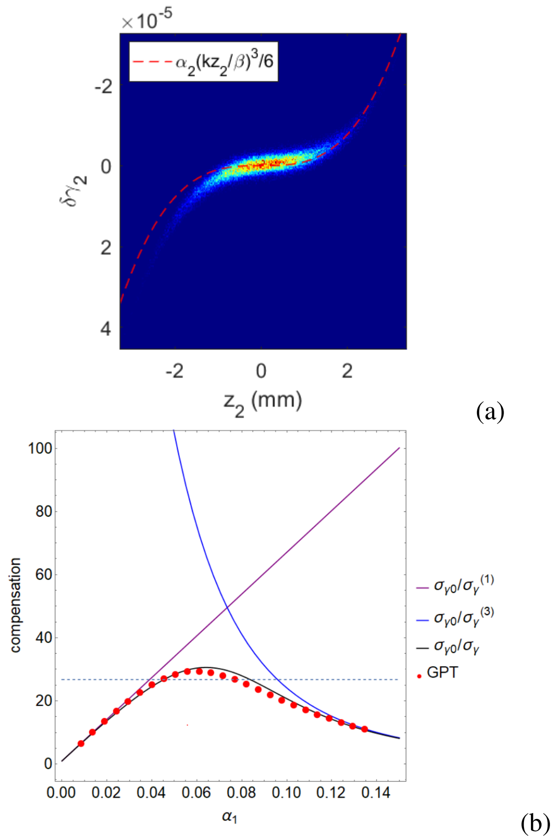

Figure 2: a) Final phase space of mildly over-compensated simulation. Space charge effects are turned off. This phase space should be considered an aggregate of many shots. b) Final RMS energy spread (normalized to initial energy spread) near optimum as a function of the first cavities amplitude.

phase offset within the bunch can then be written as:

$$\delta\gamma_1 = \delta\gamma_0 + \alpha_1 \sin(k_1 z_0/\beta), \tag{1}$$

where $\delta\gamma_1$ is the energy deviation from the reference particle at the exit of the first cavity, $\delta\gamma_0$ is the energy deviation prior entering the RF fields, typically associated with the emission process, $\alpha_1 = e\Delta V_1/mc^2$, ΔV is the peak accelerating voltage with m and e the electron charge and mass, c is the speed of light, and z_0 is the particles longitudinal coordinate relative to the reference particle (which is assumed to be at the center of the bunch). Here, we also adopt the impulse approximation, assuming that the cavities occupy a small enough distance such that their effect is only to change the particle energy.

By design, the initial bunch length entering the first cavity is sufficiently small so that we can consider only the first order in the Taylor series expansion i.e. $\sin(k_1 z_0/\beta) \approx k_1 z_0/\beta$. Each electron then ballistically propagates for a distance L to the second cavity. Its relative longitudinal position in the bunch after the drift can be written as:

$$z_2 = z_0 + L\frac{\Delta\beta}{\beta} \tag{2}$$

$$\cong z_0 + L\left(\eta_1\delta\gamma_1 + \eta_2\delta\gamma_1^2\right), \tag{3}$$

where we use the expansion $\frac{\Delta\beta}{\beta} = \sum_m \eta_m(\delta\gamma)^m$ [3] and the relativistic factors η_1 and η_2 are explicitly given by

$$\eta_1 = \frac{1}{\beta^2\gamma^3} \tag{4a}$$

$$\eta_2 = \frac{2-3\gamma^2}{2\gamma^6\beta^4}. \tag{4b}$$

The second cavity is tuned so that the reference particle arrives at a zero crossing where the fields impart a negative chirp on the beam so that the resulting final energy difference is:

$$\delta\gamma_2 = \delta\gamma_1 - \alpha_2 \sin(k_2 z_2/\beta). \tag{5}$$

The amplitude of the second cavity is set to exactly cancel the positive linear chirp imparted by the first cavity, but at this point it is important to note that the beam is much longer at the second cavity, so we can not simply ignore the non-linear terms in the Taylor series expansion. Figure 2a) showcases these 3rd-order effects that result from an elongated bunch that samples too much RF phase at the second cavity. Keeping the third order contribution, we get:

$$\delta\gamma_2 = \delta\gamma_1 - \alpha_2\left(k_2 z_2/\beta - (k_2 z_2/\beta)^3/6\right). \tag{6}$$

Combining together Eqs. (6) and (3), we obtain a symplectic map in terms of the initial coordinates which enables

ISBN: 978-3-95450-232-5 ISSN: 2673-7000 doi:10.18429/JACoW-NAPAC2022-TUXD6

a straightforward evaluation of the second moments of the longitudinal phase space at the exit of the second cavity once we know the input beam distribution. We can then use it as a guide to evaluate optimal settings for the energy spread reduction EELS scheme. For example, only retaining the linear terms, we can see, to linear order, that the residual energy spread is given by:

$$\delta\gamma_2 = \frac{\delta\gamma_0}{1 + \eta_1 \alpha_1 L k_1 / \beta}, \tag{7}$$

which points out how the compensation of energy spread can be enhanced by increasing the first cavity frequency, increasing the drift length, or increasing the amplitude of the first cavity. Figure 2b) demonstrates the analytical optimum as verified numerically.

SIMULATION

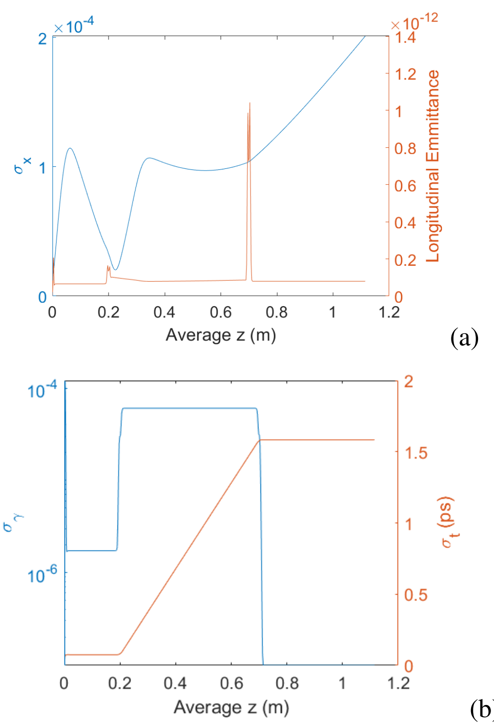

Figure 3: a) σ_x and longitudinal emmittance b) σ_t and σ_γ plotted as a function of the bunch's z-coordinate.

Defining energy spread compensation as $\sigma_{\gamma 0}/\sigma_\gamma$, in the 3D case, with a spot size of 30 um, we are able to achieve an compensation of 17.6. Figure 3 shows the evolution of the beam. The bunch length evolves as expected with an initial uncorrelated phase space which minimally stretches before the first cavity. At the first cavity (z = 0.2 m), a large linear correlation is imparted to the bunch causing the bunch to elongate over the drift. Additional energy spread is added to the bunch by accelerating the front of the bunch accelerate while decelerating the tail. This correlation is effectively nullified by the second cavity (z = 0.7 m), pushing the energy spread to be lower and preventing further bunch elongation.

The transverse effects are kept under control using two solenoids at z = 0.045 m, 0.319 m with the spot size consistently on the order of 10^{-4} ensuring that the transverse components of the beam momentum have minimal impact on the monochromatization scheme. The longitudinal emittance has some small growth due to the RF non linearities as expected over the beamline, but largely remains constant. The two spikes on the Fig. 3a) correspond to the cavities.

In general, the scheme is shown to be relatively robust under fluctuations for both phases and the electric field amplitudes. Assuming 0.1 % amplitude fluctuations and a 0.1 degrees phase jitter, 45 % of the shots still show a compensation above 15.

Although the initial implementation will be with single-electron pulses, the addition of space charge effects is noteworthy since it will contribute to additional increase in bunch length, imparting an initial linear correlation before the bunch even reaches the first cavity. This pushes the bunch to be far longer than we expect causing the bunch to sample a far greater RF phase from the second cavity, generating higher-order effects that would reduce the achievable compensation.

CONCLUSION

In this paper we present a novel scheme based on the use of two RF cavities that was developed for the purpose of improving the performances of time-resolved EELS beamline, with low energy spread being the primary target. We have shown that theoretically large compensations are possible given the correct setup and provided figures of merit that can allow one to use similar optics for lower energy spread. Monochromatization in our approach comes at a cost of bunch length so the best cases for this scheme use ultrashort initial bunches, longer drifts and a low number of particles. The final temporal resolution is inversely proportional to the energy spread, but in the examples presented here is still in the 10 ps range. In principle this scheme can be combined with ultrafast electron diffraction to resolve in momentum space the electron energy loss in the sample.

REFERENCES

[1] R. F. Egerton, *Electron energy-loss spectroscopy in the electron microscope*, Springer Science, 2011.

[2] C. J. R. Duncan, D. A. Muller, and J. M. Maxson, "Lossless Monochromation for Electron Microscopy with Pulsed Photoemission Sources and Radio-Frequency Cavities", *Physical Review Applied*, vol. 14, no. 1, pp. 014060, 2020. doi:10.1103/PhysRevApplied.14.014060

[3] B. Zeitler, K. Floettmann, and F. Grüner, "Linearization of the longitudinal phase space without higher harmonic field", *Phys. Rev. Spec. Top. Accel. Beams*, vol. 18, no. 12, pp. 120102, 2015. doi:10.1103/PhysRevSTAB.18.120102

[4] F. Carbone, O. Kwon, and A.H. Zewail, "Dynamics of chemical bonding mapped by energy-resolved 4D electron microscopy", *Science*, vol. 325, no. 5937, pp. 181-184, 2009. doi:10.1126/science.1175005