

Design and Implementation of a Flexible Beamline for fs Electron Diffraction Experiments

G. F. Mancini¹, B. Mansart¹, S. Pagano¹, B. van der Geer², M. de Loos², and F. Carbone¹

¹ *Laboratory for Ultrafast Microscopy and Electron Scattering, ICMP, École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland*

² *Pulsar Physics Eindhoven, The Netherlands*
Author e-mail address: giulia.mancini@epfl.ch

Abstract: We report the design and implementation of a table-top apparatus for Ultrafast Electron Diffraction capable of experiments in both transmission and reflection geometry with electron packets of 30 keV. The electron pulse properties in terms of charge per pulse, transverse spot-size and temporal duration on the sample can be controlled with the use of radiofrequency technology combined with a set of electron optics. The characterization of the beam is performed via a light-electrons cross-correlation experiment and we demonstrate an overall temporal resolution around 300 fs for bunches containing up to 10^5 electrons at 20 kHz repetition rate.

OCIS codes: (320.7160) Ultrafast technology; (320.7100) Ultrafast measurements.

1. Introduction

Within the last few years Ultrafast Electron Diffraction (UED) has been successfully used to investigate the dynamics of surfaces and thin films upon photoexcitation, owing to the high cross-section of electrons for interaction with matter [1,2], and simple implementation in table-top set-ups. The main limitation for this technique lies in the space-charge effects affecting time resolution. On the other hand, radiofrequency (RF) technology has been used to improve electron diffraction performances, allowing to obtain few tens to few hundreds of fs electron bunches containing up to 10^6 electrons at 100 kV acceleration voltage, enough for single-shot experiments on materials [3]. We report the design and implementation of a table-top electron diffractometer operating at 30 kV acceleration voltage capable of fs-resolved experiments both in transmission and reflection geometry. The overall layout of the experiment is presented in Fig.1A. To treat the longitudinal beam properties, a RF technology is employed, comprising an RF cavity, a temperature control unit and a phase-locked loop circuit [3]. The RF cavity is oscillating on the TM_{010} mode with a resonant frequency of 3 GHz. By synchronizing the phase offset of the RF field to the laser pulses [4], the RF cavity imparts a chirp to the probing electron bunches which compensates their broadening during propagation [3].

2. Simulation Methods

The UED apparatus has been designed following a bottom-up approach consisting mainly of three steps: (i) The beam propagation is first described within a one-to-one analogy with geometrical optics, neglecting all space-charge effects and the finite dimensions of the electron source. (ii) The finite size and emittance of the source are taken into account, to retrieve an estimate of both the temporal duration and transverse spot-size; space-charge effects are neglected. (iii) The full simulation of the beam propagation is performed with the General Particle Tracer (GPT) code [5], and is used to refine the design parameters. In the simulation the following parameters are considered: the real field profile and emittance of the electron source, the thickness of the electronic lenses (solenoids and RF cavity), the exact field profile in the RF cavity, and space-charge effects during propagation. The number of electrons per pulse used for the simulation is $6 \cdot 10^5$, the highest that can be obtained from our DC gun.

The results are shown in Fig.1B. The evolution of the beam diameter, pink line, shows the initial expansion after emission from the gun aperture; the first collimating solenoid compensates for the initial beam divergence. An almost 0.5 mm beam reaches the second focusing solenoid and enters the RF cavity receiving a slight transverse defocusing. The transverse beam dimension on the sample is around 160 μm . In the same figure, the longitudinal evolution of the beam is displayed (blue line). After emission from the cathode, the electron bunch spreads in time, and it reaches the RF cavity with a duration longer than 10 ps. The longitudinal focus is at the same propagation distance as the transverse focus for a field in the cavity around 0.54 MV/m, and the final bunch length is approximately 200 fs. These simulations suggest the possibility to perform UED experiments with approximately 10^5 electrons per pulse confined in a spot of 160 μm size and 200 fs duration. The experimental implementation procedure with a detailed description of the UED set-up is reported in [6].

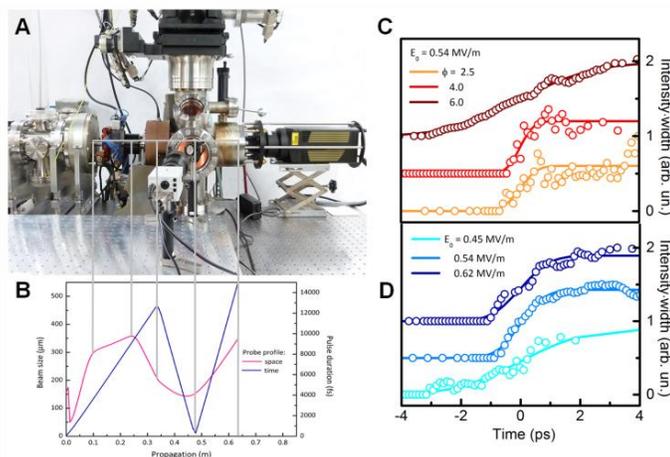


Fig.1. (A) Implementation of the UED setup consisting of a DC photo-gun, two solenoidal magnetic lenses, two steering coils, and a RF cavity. (B) GPT simulation. The longitudinal evolution of the electron beam (blue line) together with its transverse profile (pink line) is displayed as a function of the distance from the cathode. (C) Experimental results for electron pulses with 30 keV energy. Dynamics of the photoinduced dip for the phases corresponding to voltage values of 2.5, 4 and 6 V, at the optimized RF field strength value of 0.54 MV/m. (D) The effect of the RF field strength in the cavity is displayed at a constant phase value. Figures adapted from [6].

3. Experimental Characterization and Results

A cross-correlation experiment between electron and photon pulses was carried out to characterize the performances of the apparatus. A shadow image of a copper grid on the CCD was created by focusing the electrons before the same grid, and intense laser pulses were used to photoemit electrons via a multi-photon process from the copper. The charge created by light pulses produces a lensing effect on the electrons which is visible as a distortion of the grid image. In this experiment the longitudinal evolution of the beam is governed by the RF field strength and phase in the RF cavity and determines the final time-resolution of the set-up. The dip caused in the image counts by the laser pulse is fitted to a Gaussian profile whose area (intensity · width) is displayed for different values of the phase and field strength respectively in Fig. 1C and D. These transients evidence the optimal phase and field as simulated via GPT, and are fitted to a step function convoluted to a Gaussian, the latter simulating the effect of time-resolution. These fits yield a temporal resolution of 360 fs for the overall area. The same experimental parameters have been used to simulate in GPT the spatial and temporal evolution of the beam in the apparatus.

The overall temporal resolution of a UED experiment depends on the statistic average between the arrival time jitter of the pulses, the temporal duration of the electron bunches on the sample and the Group Velocity Mismatch (GVM). Therefore it can be generally defined as:

$$t_{res,UED} = \sqrt{\Delta t_{jitter}^2 + \Delta t_{duration}^2 + \Delta t_{GVM}^2} \quad (1)$$

A complete experiment reported in Ref. [3] shows that the synchronization of the RF cavity to the laser has a pulse-to-pulse jitter around 80 fs. For this reason, we consider $\Delta t_{jitter} = 80$ fs in Eq. (1). The probing pulse duration in the focus of the cavity is estimated via GPT simulation, and for the highest charge we considered, $6 \cdot 10^5$ electrons per pulse, we obtain 288 fs. Therefore, $\Delta t_{duration} = 288$ fs in Eq. (1). In our geometry the resulting GVM has been estimated 242 fs, taking into account an excitation laser fluence of 48.9 mJ/cm² over a photoexcited area of 125 μm diameter ($\Delta t_{GVM} = 242$ fs). According to these estimates, which are done considering the highest charge we can extract from our cathode and a non-optimized geometry to limit GVM effects, we obtain an overall time resolution around 380 fs, in good agreement with the lowest value obtained experimentally of 360 fs.

References

- [1] A. H. Zewail, *Annu. Rev. Phys. Chem.* **57**, 65-103 (2006).
- [2] D.-S. Yang, N. Gedik, A. H. Zewail, *J. Phys. Chem. C* **111**, 4889-4919 (2007).
- [3] T. van Oudheusden *et al.*, *PRL* **105**, 264801 (2010).
- [4] F. B. Kiewiet, A. H. Kemper, O. J. Luiten, G. J. H. Brussaard, M. J. van der Wiel, *Nucl. Instrum. Methods Phys. Res. A* **484**, 619-624 (2002).
- [5] S. B. van der Geer, M. J. de Loos, The General Particle Tracer code, <<http://www.pulsar.nl/gpt>>.
- [6] G. F. Mancini *et al.*, *Nucl. Instrum. Methods Phys. Res. A* **691**, 113-122 (2012).