TIME RESOLVED RELATIVISTIC ELECTRON DIFFRACTION

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Abstract

We report on the use of a ultrashort high brightness relativistic beam from the UCLA Pegasus laboratory RF photoinjector source for probing matter transformation at the atomic scale with sub-100 fs time resolution. The high accelerating gradient and the relativistic electron energy allow to pack more than 107 electrons in less than 100 fs bunch length, enabling the study of irreversible ultrafast phenomena by single-shot diffraction patterns. The experimental setup, and the preliminary results from the first ever relativistic electron diffraction time-resolved study are discussed.

INTRODUCTION

Ultrafast electron diffraction holds the promise to enable real time atomic scale resolution studies of ultrafast structural dynamics[1-2]. Even though all of the scientific results so far have been obtained using non relativistic electron beams, relativistic electron diffraction has the advantages of yielding shorter and much more intense probe pulses.

RF photoinjectors can deliver 10^7 electrons in less than 100 fs and a lot of interest has gathered around the possibility of using such electron sources for this new application. After first preliminary experiments showed the feasibility of obtaining diffraction patterns from thin metal foils[3-4], at the Pegasus Laboratory we set out for the task of demonstrating time-resolved diffraction patterns.

In this paper we report the first preliminary results of these efforts.

The UCLA Pegasus photoinjector is an advanced photoinjector laboratory characterized by a ultrashort laser pulse (<50 fs rms) photocathode drive laser. When the laser energy generates pulses of tens of pC the electron beam undergoes a space charge driven expansion and evolves into a final nearly ideal uniformly filled ellipsoidal distribution. As this distribution is characterized by self-fields linear in the coordinate offset this regime of operation is of great interest in the quest for higher brightness beams for a variety of applications such as FEL injectors[5].

For ultrafast electron diffraction, the laser energy is lowered in order to extract only few pC of charge from the cathode and limit as much as possible the longitudinal expansion and obtain a sub-100 fs electron pulse at the sample plane 80 cm from the cathode.

We report in the Table the operating parameter of the Pegasus RF photoinjector when operated in UED mode.

Table 1: Pegasus Photoinjector Parameters for Ultrafast Relativistic Electron Diffraction.

Parameter	Value
Beam Energy	3.5 MeV
Beam charge	1-5 pC
Bunch length	100-200 fs
Spot size at the cathode	400 um
Normalized emittance	<0.5 mm-mrad
Gun field gradient	75 MV/m
Launch phase	25 deg

The electron bunch length is measured using an X-band streak camera at 1.8 m of distance from the cathode. The results of electron bunch length vs. charge for three different cathode spot sizes are shown in Fig. 1.

The electron bunch length sets the time resolution for UED, but the spatial resolution is largely influenced by the beam emittance. The intrinsic angle divergence in the beam has to be much lower than the Bragg scattering angle. In other words, in order to obtain a good contrast in the diffraction pattern, one would like to utilize a beam with as low as possible transverse emittance.

For the very low charges employed in these experiments, most of the contribution to the beam emittance originates from the cathode or thermal emittance which is directly proportional to the laser spot size at the cathode. For these reasons, we typically operate at 400 um laser spot size in a compromise between spatial and temporal resolution.

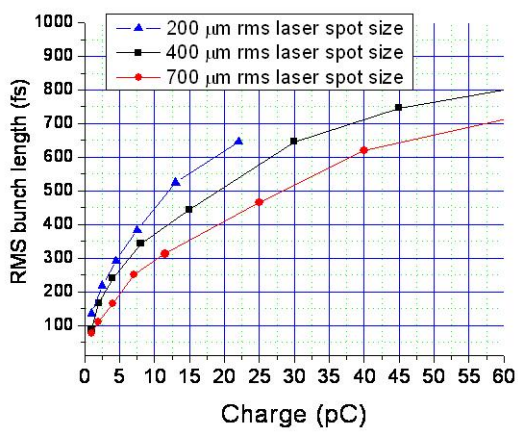

Figure 1: Bunch length as a function of charge for three different spot sizes.

STATIC DIFFRACTION PATTERNS

In order to clearly define the probed area on the sample, a 1 mm diameter aperture hole is inserted in the beam path at a distance 80 cm from the cathode. The transverse dimensions of the propagating beam are then minimized with the emittance compensation solenoid on the Yag crystal fluorescent detector screen located 1.9 m from the cathode in order to reduce the beam divergence. Typically we obtain a rms beam size of 300 um rms (full beam 1.2 mm diameter) in these conditions, and a corresponding rms spread in beam angles of ~0.1 mrad. The amount of charge passing through the hole is ~ 1 pC and it is only weakly dependent on the charge on the cathode. A beam with larger charge on the cathode in fact is collimated by the solenoid to a larger spot size so that a smaller fraction gets transmitted through the hole.

A 100 nm thick metal foil is then lowered in the beam path just few cm downstream of the hole aperture. The diffraction rings are immediately visualized on the screen, and very fine tuning of the solenoid and launch phase is at this point required only to maximize the pattern contrast. This technique has the advantage of allowing a prompt and repeatable tuning for the diffraction camera. It also generates diffraction patterns with circularly symmetric rings, as opposed to our previous technique involving quadrupole magnets [4].

In order to maximize the quality of the diffraction pattern, it is required to subtract from the images a dark-current only background shot acquired blocking the driver laser before the cathode. Even if the field-emission is minimized by employing a short RF macropulse (1.5 us), due to the very large aperture of the lens employed to image the Yag detector screen, a significant dark current signal is observed even after the aperturing hole.

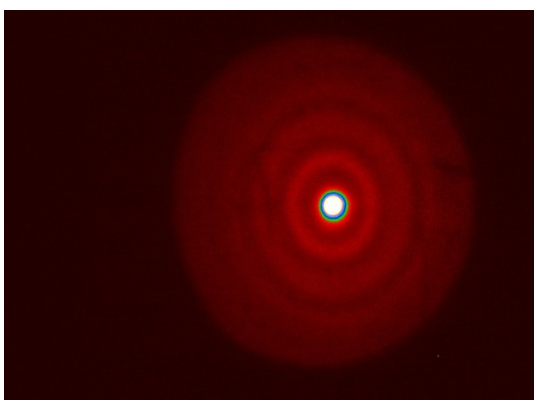

Figure 2: Single shot diffraction rings obtained at the Pegasus laboratory with 3.5 MeV beam from a 100 nm thick gold foil.

Diffraction patterns from gold, silver, titanium and aluminium have been recorded using single-shot exposure thanks to the large number of probing electron per pulse (~10^7). The width of the rings, is related to the undiffracted beam dimensions. Even though the pattern quality if significantly worse compared to the images that can be obtained with TEMs, the Bragg peaks position and amplitude are easily identified. The uncertainty on their determination is sufficient to appreciate any structural change, especially the fairly large ones typical of the irreversible processes.

PUMP AND PROBE EXPERIMENTAL SETUP

The pump laser energy is obtained splitting the IR pulse before the third harmonic generation stage. After the beamsplitter about 1 mJ of 800 nm light are available for exciting an ultrafast phase change in the metal under study. The pump pulse is routed to the accelerator beam line through a trombone based delay line (shown in Fig. 3). In order to maximize the gold absorption a 500 um thick doubling BBO crystal is used just before the final vacuum window to generate 0.4 mJ of 400 nm light. A f = 36 cm lens is used to focus the pump laser on the back surface of the target to a 500 um diameter spot size. This yields an illuminating fluence of 20 mJ/cm^2. Ideally one would require the cross section of the pump beam to be much larger of the probe beam size. Unfortunately in our case, we would need a higher pump energy in order to maintain the fluence above the threshold for melting. For this reason, we choose to focus the pump laser to a spot size smaller than the probing beam.

Given the fact that there is 12 meters long transport line between the delay line and the target, we found the need to install a remote controlled alignment feedback system for the translation stage shown in Fig. 3. After the delay line is moved the laser is brought back in less than 1 s within 50 um of its initial position by a computer software. (see Fig. 4).

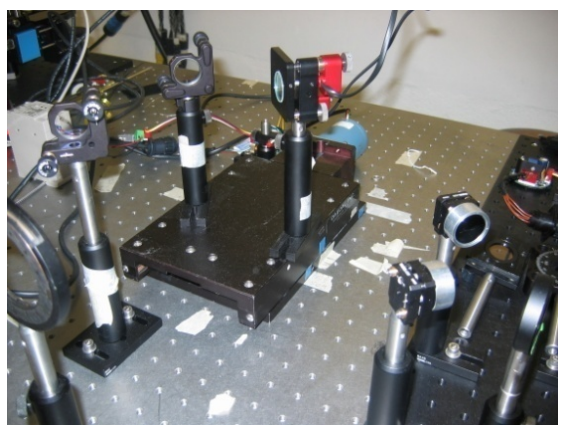

Figure 3: Translation stage with remote controlled mirror for varying the pump-probe time-delay.

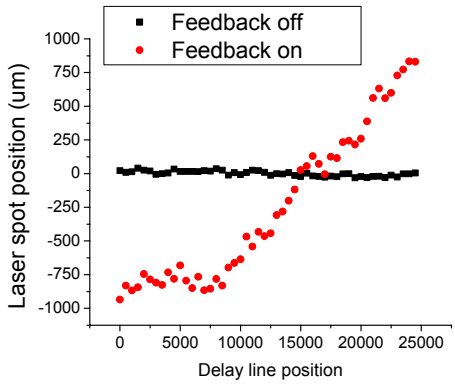

Figure 4: Measurement of the laser spot drift when the translation stage is moved with and without feedback on.

TIME RESOLVED DIFFRACTION DATA

When the laser is turned on and hits the sample before the electron beam the diffraction pattern changes as shown in Fig. 5.

There are two main effects: i) a decrease in the contrast of the diffraction peaks, as it is expected due to the loss of order in the lattice; ii) it is also noticeable a decrease of the main beam amplitude and the general increase of large angle scattered background.

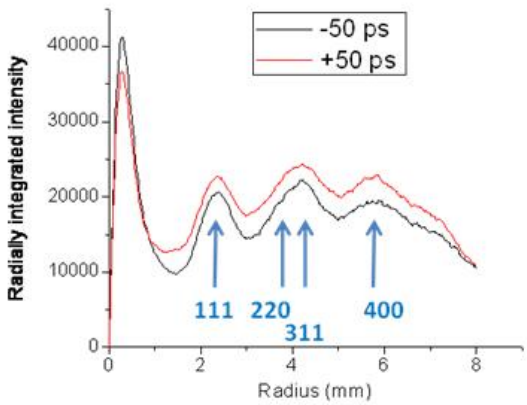

Figure 5: Radially integrated intensity before and after the laser hits the sample.

In order to take into account relative changes in the peak amplitudes, we normalize the image intensities by the total charge in the beam. This results in very stable peak amplitudes and allows us to appreciate relative changes smaller than 5 % for these quantities.

Moving the sample to a fresh spot every time, we vary the delay between the pump and probe and record a frame-by-frame movie of the metal transformation. This is shown in Fig. 6 where the normalized peak amplitudes are plotted as a function of the time delay. It is evident that at t = 200 ps the sample responds quite differently. This change could be due to a shortening of the inelastic mean free path due to laser pumping [7] or alternatively to a plasma blow-off effect[6]. We are in the process of investigating the origin of this phenomenon.

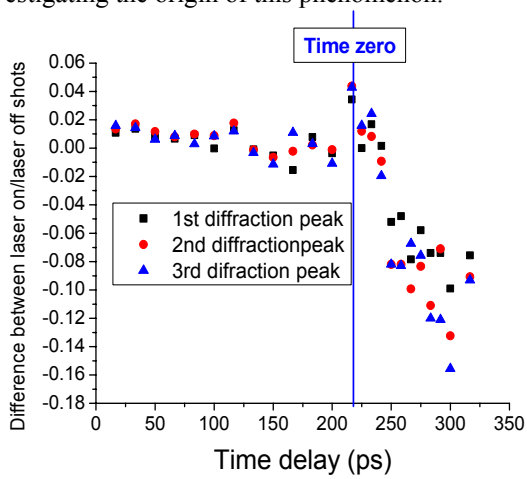

Figure 6: Time resolved relativistic electron diffraction data. The normalized amplitudes of the diffraction peaks are followed as a function of the time-delay between the pump and the probe. Only a single shot is taken for each time-delay.

CONCLUSIONS

We report for the first time to our knowledge time resolved relativistic electron diffraction data taken with the Pegasus photoinjector.

Preliminary results indicate a clear change in the diffraction patterns taking place after the laser hits the sample. Deeper studies to investigate the details of the change are underway.

The diffraction patterns are taken with single shot exposures. This technique opens the way to the study of irreversible ultrafast processes.

Due to the intrinsic jitter of an RF photoinjector we foresee the use of a non destructive time-stamping technique to record the relative time of arrival between the laser and electrons[8] and improve the resolution of the technique to sub-100 fs.

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