First demonstration of ultrafast-electron diffraction using a 3-GHz electro-optical comb generator as a UV photoinjector laser.

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Abstract: We demonstrate ultrafast-electron diffraction using an electro-optic comb-generator, emitting a 3 GHz train of ps pulses at 257 nm. This new operation mode improves electron coherence and reduces space-charge effects.

1. Introduction

Ultrafast Electron Diffraction (UED) is a technique to investigate ultrafast structural dynamics using a laser-pump, electron-probe scheme [1]. Complementary to other diffraction techniques like X-rays diffraction, electron diffraction is particularly well suited to investigate materials of low atomic number and therefor of interest for research fields like organic chemistry or biology. At DESY, the UED facility REGAE (Relativistic Electron Gun for Atomic Exploration) incorporates a 3 GHz S-band photoinjector which can generate electron bunches with a kinetic energy of up to 5 MeV [2]. A 12.5 Hz repetition rate Titanium-Sapphire laser currently is used both to generate the optical pump pulses and the UV pulses to create electron bunches from a Cs_2Te photocathode.

A major limiting factor in UED is the mutual repulsion of the electrons within an electron bunch due to their negative charge. While a high number of electrons is desirable for the diffraction signal in terms of signal-to-noise ratio, the so-called space charge effect reduces the spatial resolution (spatial coherence length) for high-charge bunches. We here demonstrate, for the first time to our knowledge, a method which significantly reduces space-charge effects on the expense of lower time resolution, but without compromising the signal-to-noise ratio of the diffraction pattern: Instead of using one single high charge electron bunch, we create an up to 1.5 µs long train of electron bunches of relatively low individual bunch charge (and thus higher coherence) using a 3 GHz

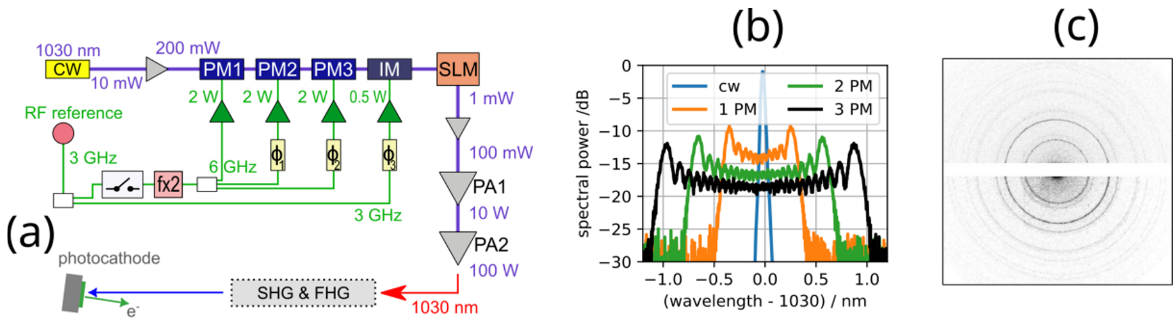

Figure 1: (a) Schematic of the laser system. cw: continuous wave seed laser, PM: phase modulator, SLM: phase-and amplitude shaper (spatial light modulator) , PA: power amplifier, SHG: second-harmonic generation, FHG: fourth-harmonic generation. (b) Output optical spectra of the laser frontend: unmodulated seed (cw) and output with one, two and three phase modulators (PM) active. (c) A first diffraction of a gold (Au) powder reference sample, generated using the new laser system.

ultrafast UV laser, filling each accelerating bucket of our gun. To achieve this, we developed and commissioned a new laser system based on electro-optic comb generation. This new low space-charge operation mode will significantly expand the scientific capabilities of REGAE

2. Laser system

To generate the maximum number of electron bunches within the 1.5 µs-long radio-frequency (RF) pulse accelerating the electrons at REGAE, the laser was designed to have a repetition frequency equal to the 3 GHz driving RF of the electron gun. This was implemented using the technique of an Electro-optical frequency comb generator (EO comb) [3,4]. While easily allowing such high repetition rate, this approach has the additional advantage that the RF field driving the EO comb can be derived directly from the accelerator RF, thus making the laser inherently synchronized to the accelerator.

A schematic overview our laser system is shown in Figure 1a), most details were reported last Europhoton Conference [5]. In brief: A single-mode, polarization maintaining (pm)-fiber pulse generator centered at 1030 nm comprises a narrow bandwidth continuous-wave fiber seed laser, amplification, phase- and amplitude modulation stages and a phase-and amplitude shaper for chirp removal. The output pulse train (1.8 nm bandwidth, 2 ps duration, see Figure 1b) is amplified in two large-mode ytterbium-doped pm fiber amplifier stages to the 100W average power. The RF field driving the modulators is derived from the REGAE main RF oscillator (MO) and amplified using several RF amplifiers. Using two conversion crystals for second (SHG, 15 mm LBO crystal) and subsequent fourth-harmonic generation (FHG, 5 mm CLBO crystal), the IR light is converted to UV at 257 nm.

To generate a pulse train of defined length in the UV, the RF modulation section of the laser incorporates a fast RF switch. Using this switch, the infrared output of the laser can be toggled between a fully modulated pulse train and quasi-continuous wave, with a transition time in the order of about 30 ns. Due to the differences in UV conversion efficiency, effectively this allows to generate a burst of UV pulses with controllable duration.

The laser output pulses have an energy of about 33 nJ (100 W average power), with a maximum conversion efficiency of about 1% resulting in 0.33 nJ pulses in the UV.

3. Results

We recently installed our laser in the REGAE facility. The UV beam paths of both 12.5 Hz and 3 GHz laser were combined via a flip mirror, followed by a common transport beamline to the photo cathode of the electron gun.

Compared to the synchronization schemes for oscillator-based photoinjector lasers [6], we found the synchronization and timing procedure was vastly simplified using the new EO comb system, as it is inherently synchronized to the facility RF. The correct timing was achieved by scanning the gun RF phase once and additionally shift a trigger signal defining the start of the UV burst.

With the newly installed laser system and beam transport, we were quickly able to generate first electrons, proving the feasibility of an EO comb-based photoinjector laser. Here we present the first static (probe-only) diffraction images of reference samples. As an example, we show in Figure 1c) a diffraction pattern from a thin sample of gold (Au) powder. Very recently we were able to generate first diffraction data using small apertures for the electron beam, utilizing the reduced space charge and thus increasing the spatial coherence of the electron beam. We are currently evaluating those imaging and will present the results at the conference.

Even without any active feedback loop, the passive stability of the laser itself as well as the relative stability with respect to the electron gun is excellent. We could measure diffraction data for several hours without any adjustments on the laser system – a crucial requirement for extensive parameter scans in pump-probe experiments. We are currently working on optimizing the UV conversion and the transmission through the UV beam transport for more UV power at the gun, as well as on fully integrating all laser controls and diagnostics into the facility control system.

In conclusion, we demonstrated, to our knowledge for the first time, ultrafast electron diffraction based on an electro-optical frequency comb laser driven electron gun. This laser concept vastly simplifies laser-accelerator synchronization. We furthermore showed reliable operation for hours without intervention, allowing e.g. extensive parameter scans in pump-probe UED experiments. In the coming months, we intend to fully quantify laser and electron beam parameters and identify possible points of improvements, with focus on the UV energy and beam shape. A quantitative back-to-back comparison with the old laser system, is planned will support those studies. Ultimately, the new laser system shall extend the operation modes for REGAE, allowing new parameter ranges for UED experiments.

4. References

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