ACCELERATOR BASED ULTRAFAST ELECTRON DIFFRACTION AND MICROSCOPY AT SJTU*

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Abstract

Historically particle accelerators are instrumental for high energy physics (accelerator based colliders) and photon science (accelerator based synchrotron light sources and free electron lasers). Now there is growing interest in applying accelerator technology to solve the grand challenge in probing matter at ultrafast temporal and ultrasmall spatial scales. In this paper we discuss how one can use MeV electrons produced in accelerators (e.g. photocathode rf guns) to study ultrafast dynamics at atomic scale through ultrafast electron diffraction and microscopy techniques. We will also describe the current status of ENTROPY (cENter for ulTRafast diffractiOn and microscoPY) at Shanghai Jiao Tong University (SJTU). This center is expected to provide access to new sciences by producing kHz rep-rate ultrafast and ultrabright electron beams that will give researchers unparalleled power and precision in examining the fundamental nature of matter.

INTRODUCTION

The ability to probe matter with both ultrahigh temporal and ultrahigh spatial resolution is one of the ultimate goals in scientific research. This capability is perhaps best enabled by large scientific facilities, such as free-electron lasers (FELs) where intense x-rays with femtosecond pulse width are produced by sending GeV electrons through long undulators (see. e.g. [1]). There are small facilities with significantly reduced cost and size, yet still providing sufficient temporal and spatial resolution for studies of a wide array of ultrafast science. For instance, ultrafast electron diffraction (UED), ultrafast electron crystallography (UEC) and ultrafast electron microscopy (UEM) in which ultrashort electron pulses are used as the probes, have also been developed to study structural dynamics (see, e.g. [2, 3]). In this paper, we discuss the key physics of UED and UEM and show how one may enhance the performance of these facilities with accelerator technologies.

MOTIVATION FOR RELATIVISTIC UED AND UEM

Fast time-dependent phenomena are typically studied with pump-probe techniques in which the dynamics are initiated by pumping lasers and then probed by delayed pulses. In UED the diffraction pattern is the Fourier transform of the nuclei and electron density distribution of the sample. Therefore, by studying the diffraction pattern variation as a function of time, one may investigate the structural dynamics. The temporal resolution depends on the pumping laser pulse width, the probing electron pulse width, the velocity mismatch and the timing jitter between the pumping and probing pulses.

Conventional DC-gun based UED has limited beam energy ($\sim$ 100 keV and below) and low acceleration gradient ($\sim$ 10 MV/m). The serious space charge force tends to increase the electron pulse width that limited the number of electrons that can be produced in an ultrashort pulse. While one may shorten the distance between the cathode and sample, or use rf cavity to reverse the energy chirp to compress the beam, the velocity mismatch still limited the temporal resolution to picosecond in studies of gas phase samples.

For conventional UEMs, there are two major configurations for achieving high temporal resolution, both of which replace the DC electron source in a transmission electron microscope (TEM) with a laser triggered photocathode gun to produce bunched electrons. The first configuration operates in stroboscopic mode [4] in which a femtosecond beam with only a single electron (on average) to avoid space-charge effect is used to probe the sample after a femotsecond pump laser. Typically one useful image corresponding to a specific time delay between the pump laser and probe electron beam is obtained with integration over about 10^8 shots. While very high temporal resolution and spatial resolution can be achieved with this configuration, it only applies to studies of perfectly reversible process, because the sample needs to be pumped $\sim 10^8$ times and the sample must completely recover after each shot. Alternatively, a useful image may be obtained in a single shot with a longer pulse that contains enough electrons. With the beam peak current several orders of magnitude higher than a conventional TEM, the temporal resolution and spatial resolution in this configuration is degraded by space charge effects and the limited electron beam brightness, etc. For instance, the recently developed dynamic TEM (DTEM) has achieved about 15 nanosecond (ns) temporal resolution and 10 nanometer (nm) spatial resolution (corresponding to a product of temporal and spatial resolution 10^{-16} m*s) using a 200 kV electron beam produced with a nanosecond laser pulse [5].

Apparently there are several distinct advantages to use accelerator technologies (e.g. photocathode rf gun as used in many FELs facilities as the electron sources) for UED and UEM. First, higher accelerating field gradient allows extraction of a given charge from a source with smaller area that reduces thermal emittance and increases the space charge limited maximal beam brightness. Second, higher

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gradient also allows electrons to be accelerated to relativistic within a shorter distance that mitigates space charge induced emittance growth, which is useful to preserve the beam brightness. Third, electrons with MeV energy have velocity close to the speed of light, which also effectively mitigates the velocity mismatch problem that may allow one to push the resolution in studies of gas phase samples to beyond 100 fs.

CURRENT STATUS OF ACCELERATOR BASED UED AND UEM

Since the proposals of MeV UED in the early 2000's [6, 7], photocathode rf gun based UEDs have been demonstrated at many laboratories. Static MeV electron diffraction patterns have been observed in SLAC [8], UCLA [9], Tsinghua University [10], Osaka University [11], BNL [12], DESY [13] and Shanghai Jiao Tong University (SJTU) [14]; and pump-probe experiments with MeV UED have been tested in UCLA [9], Osaka University [11], BNL [12] and SJTU [15]. Currently, most of the MeV UED operate at low rep-rate (e.g. $\sim$ 10 Hz), and the resolution is typically on the order of a few hundred femtosecond, limited either by the electron bunch length, or the timing jitter (related with long-term stability of the accelerator). The samples studied so far are limited to solid, and no results on liquid or gas phase sample have been reported.

Accelerator based UEMs are still in the very early development stage. One prototype UEM with electron beam produced in a photocathode rf gun has been constructed [16], and very recently two theoretical studies have shown the feasibilities of reaching 10 ps temporal resolution and 10 nm spatial resolution in photocathde rf gun based UEMs [17, 18]. The main challenges include, but are not limited to, producing electron beams with high energy stability, low emittance and low energy spread, fabrication of high-field focusing elements such as superconducting solenoids and permanent magnet quadrupoles, high accuracy in alignment of all the magnets, etc.

ENTROPY AT SJTU

The ENTROPY (cENter for ulTRafast diffractiOn and microscoPY) being constructed at SJTU will host a user facility with photocathode rf gun based UED and UEM [14]. A prototype machine, schematically shown in Fig. 1, has already been built to test the critical technologies. The electron beam is produced in an S-band (with frequency at 2856 MHz) 1.6-cell photocathode rf gun with a femtosecond UV laser. The beam is focused by a solenoid to form convergent electron diffraction pattern at a fluorescent screen about 3.4 m downstream of the sample.

The femtosecond laser system can operate at 1 kHz and the maximal energy for each laser pulse is about 18 mJ. After a beam splitter, 10% of the laser is tripled to 266 nm with third harmonic generation crystals to drive the photocathode rf gun, and the rest of the laser can either be di-

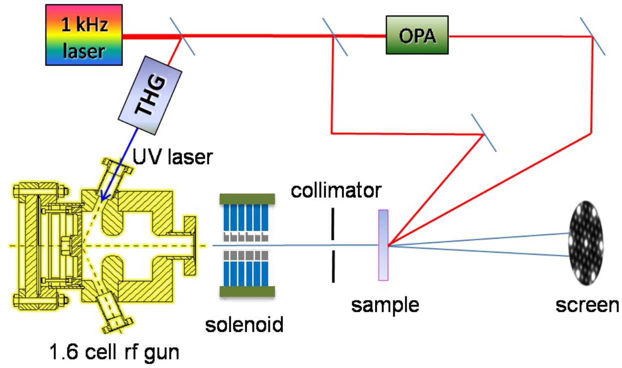

Figure 1: The schematic layout of the MeV UED at SJTU.

rectly used as the pump at 800 nm or be converted to a pump with variable wavelength with an OPA. The laser is synchronized with the rf oscillator, and the laser-rf timing jitter has been estimated to be about 100 fs (rms) from the phase jitter measurement using a mixer and an rf phase detector. The S-band photocathode rf gun is powered by a 5 MW klystron, which is driven by a solid-state modulator that can operate at 500 Hz and whose pulse-to-pulse stability is better than 0.1% (rms). Currently, the system is operated at 2$\sim$10 Hz because of the limited radiation shielding capability of the experimental hall. A new 2.4 cell gun is being made which will allow us to operate the UED at up to 1 kHz. Efforts are also being devoted to upgrading the shielding wall that will allow operation at full rep-rate.

The sample chamber is designed to hold both solid state foil and gas phase sample. For metallic films, a built-in temperature cooler could precisely control the temperature from a few K to 500 K. For studies of gas phase sample, two differential pumping chambers are used to maintain the relatively high vacuum in the electron gun (about 10^{-9} Torr) and the detector chamber (about 10^{-8} Torr), while the vacuum is about 10^{-3} Torr in the gas phase sample chamber, in which a nozzle with a turbo pump is used to continuously generate gas phase sample. SF6 will be used to test the design of the differential pumping system soon.

The first electron beam was produced in December 2013, and high quality single shot diffraction patterns of poly-

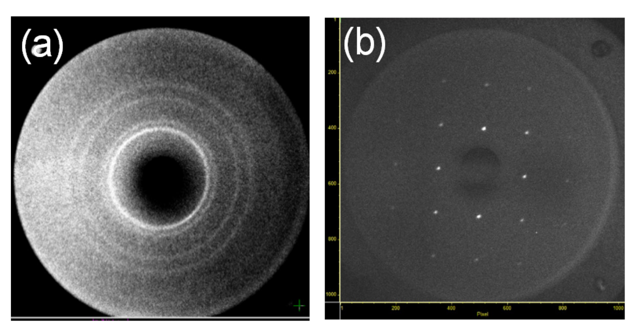

Figure 2: Single shot diffraction pattern of polycrystalline Al (a) and single crystal Gold (b) at SJTU.

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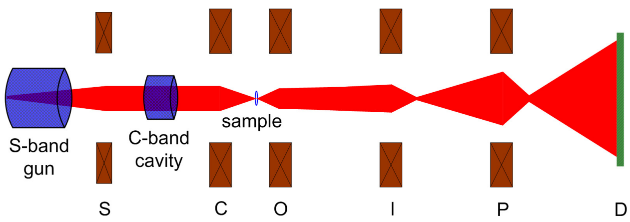

Figure 3: The schematic layout of the MeV UEM at SJTU.

crystalline Aluminium and single crystal Gold are shown in Fig. 2. Following demonstration of high quality single shot diffraction patterns, techniques are also developed to use the beam for pump-probe experiments. Specifically, we used the perturbation from the laser induced plasmas to the electron beam to determine the relative timing between the pumping laser and probing electron beam, and studied laser induced heating of single crystal gold [15].

Following the studies in [17], the schematic of the UEM is shown in Fig. 3. The electron beam is produced in an S-band photocathode rf gun. To reduce beam emittance, we will use laser shaping techniques to obtain a laser pulse with flat-top distribution (8 ps full width) in the center and Gaussian ramping (1 ps (rms)) at the head and tail. With this distribution, simulation shows that a beam with a few pC charge and normalized emittance on the order of ~ 10 nm may be obtained. Because the electron bunch extends over many degrees of the rf phase, the sinusoidal field introduces a correlated spread in beam energy, leading to a global energy spread of about 1×10^{-3} (Fig. 4a). Fortunately, the beam correlated energy spread can be corrected up to second order with a harmonic cavity. For instance, after compensation for the nonlinear rf curvature in a C-band cavity, the global energy spread reduces to 6×10^{-5}, and the energy spread for the core beam (within ± 3 ps) is only 1×10^{-5} (Fig. 4b).

A low emittance and low energy spread beam together with high-field small aberration coefficient lens ensure good performance of a UEM. As a representative example, we performed start-to-end simulations and the image of an ideal 100%-contrast sample as shown in Fig. 5a are shown in Fig. 5b, which confirms the feasibility of reaching 10 ps and 10 nm temporal/spatial resolution in accelerator based UEM.

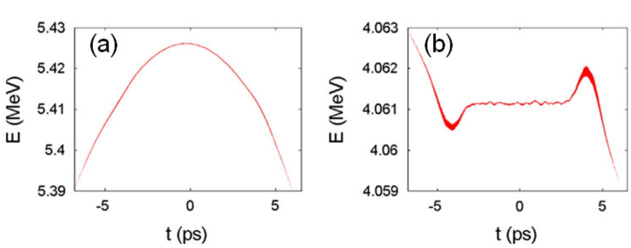

Figure 4: Beam longitudinal phase space at the exit of the s-band gun (a) and at the exit of the harmonic cavity (b).

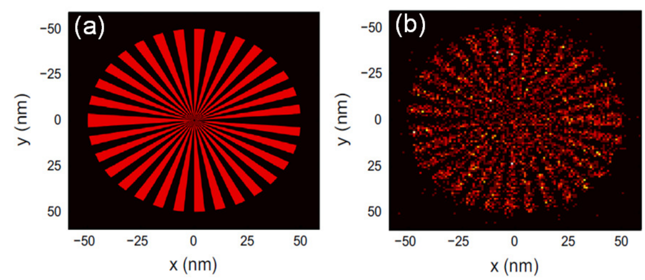

Figure 5: Distribution of the virtual sample (a) and the image (b). Note, the image is inverted.

SUMMARY

With the recent progress both in ultrafast lasers and in the generation and control of high-brightness electron beams used in modern accelerators, it is possible to significantly enhance the capabilities of UEDs and UEMs. The ENTROPY at SJTU will be completed in 2018 and we hope this facility will provide access to to new sciences with kHz rep-rate MeV UED and UEM.

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