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Accelerator-based single-shot ultrafast transmission electron microscope with picosecond temporal resolution and nanometer spatial resolution

D. Xiang [a,*], F. Fu [a], J. Zhang [a], X. Huang [b], L. Wang [b], X. Wang [b], W. Wan [c]

[a] Key Laboratory for Laser Plasmas (Ministry of Education), Department of Physics and Astronomy, Shanghai Jiao Tong University, Shanghai 200240, China
[b] SLAC National Accelerator Laboratory, Menlo Park, CA 94025, USA
[c] Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

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ABSTRACT

We present feasibility study of an accelerator-based ultrafast transmission electron microscope (u-TEM) capable of producing a full field image in a single-shot with simultaneous picosecond temporal resolution and nanometer spatial resolution. We study key physics related to performance of u-TEMs and discuss major challenges as well as possible solutions for practical realization of u-TEMs. The feasibility of u-TEMs is confirmed through simulations using realistic electron beam parameters. We anticipate that u-TEMs with a product of temporal and spatial resolution beyond 10^{-19} ms will open up new opportunities in probing matter at ultrafast temporal and ultrasmall spatial scales.

1. Introduction

Transmission electron microscope (TEM [1]) has played an important role in development of physics, chemistry, biology and material sciences. In a conventional TEM (see, for example Refs. [2,3]), the electrons are produced in an electron gun, accelerated with the DC field and then focused onto a sample with a condenser lens system. The electron beam distribution at the sample is further magnified (by up to a few millions) with the imaging lens system and finally measured with an area detector. Over several decades TEMs have been widely used to probe molecules, atoms, crystals, and innovative new materials with atomic resolution in order to better understand their properties and behaviors. Some breakthroughs enabled by TEMs include discovery of the single-walled nanotube [4], detection of tumor viruses [5], imaging of individual atoms [6], just to name a few.

In conventional full field TEMs, the electron beam is typically produced with thermionic, Schottky, or field emission. The voltage of routine TEMs is below 200 kV and medium-voltage TEMs work at 200–500 kV to provide better resolution. In high-voltage electron microscopes, the voltage reaches 500 kV to 3 MV [7], which provides much better transmission such that materials and large biological cells that are difficult to prepare in thin enough layers can be studied. As the electrons are emitted continuously, the temporal resolution in conventional TEMs is only achieved at the millisecond level using a fast framing camera. Though millisecond is not fast, it has already allowed real-time study of some slow dynamics, e.g. the motion of thermally activated vortices in a superconductor has been observed with an electron microscope at 30 frames per second [8]. The temporal resolution can be significantly improved if the electrons are bunched (e.g. illuminating a photocathode with a short pulse laser to produce a short electron beam) rather than emitted in constant stream. This also removes the need for a fast detector and the temporal resolution is simply determined by the electron bunch length, which in many cases is comparable to the laser pulse width.

Currently, there are two major configurations for achieving high temporal resolution in TEMs. The first configuration operates in a stroboscopic mode [9] in which a femtosecond beam with only a single electron (on average) to avoid a space–charge effect is used to probe the sample after a femtosecond pump laser. Typically one useful image corresponding to a specific time delay between the pump laser and the 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,

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[*] Corresponding author.
E-mail address: dxiang@sjtu.edu.cn (D. Xiang).

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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 spatial resolution in this configuration is degraded by space charge effects, 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} ms) using a 200 kV electron beam produced with a nanosecond laser pulse [10].

Increasing both the beam energy and the beam brightness may further push the product of temporal and spatial resolution by a few orders of magnitude (see, for example [11]). For instance, a possible path to reach 10 ps temporal resolution and 10 nm spatial resolution with a 5 MV TEM has been briefly discussed in Ref. [12]. A prototype u-TEM using an electron beam produced in a photocathode rf gun [13,14] has been developed and about 300 nm spatial resolution has been achieved [15,16]. Very recently, an u-TEM using quadrupoles for imaging has been briefly discussed in Ref. [17]. In this paper, we present a detailed study of the feasibility of an accelerator-based u-TEM capable of providing a few picosecond temporal resolution and a few nanometer spatial resolution in a single-shot. We study key physics related to performance of u-TEMs and discuss major challenges (such as beam emittance, beam energy spread, beam energy stability, and space charge effect) as well as possible solutions for practical realization of u-TEMs. Using a representative set of parameters, the feasibility of achieving a product of temporal and spatial resolution beyond 10^{-19} ms is confirmed through simulation.

2. Considerations for an u-TEM

The performance of a conventional TEM is mainly determined by the electron beam quality and the spherical and chromatic aberrations of the imaging system. The spatial resolution limited by spherical aberration is approximately $r_s = C_s\theta^3$, where C_s is the spherical aberration coefficient and θ is the divergence of the detected electrons at the sample. Similarly, the spatial resolution limited by chromatical aberration is approximately $r_c = C_c\theta\delta$, where C_c is the chromatical aberration coefficient and δ is the relative energy spread of the beam at the exit of the sample. Both C_s and C_c are on the order of the focal length of the objective lens. In accelerator terminology, C_s and C_c are related to the U_{1222} and T_{126} elements of the third order and second order transfer matrix, respectively. The biggest difference between an u-TEM and a conventional TEM is, perhaps, that the beam peak current is many orders of magnitude higher such that collective self-interactions of the electrons may play a role. In this section we discuss the general considerations for a single-shot u-TEM and give an order-of-magnitude estimate for the beam and imaging system requirements for realization of an u-TEM.

In view of producing a beam with high brightness, a high accelerating field gradient is required, regardless of if the electrons are produced in a DC diode or a laser-driven photocathode rf gun. When electrons are produced, they are affected by their self-fields. For sufficiently high current density, the electric field at the cathode from space charge may equal to the external field, and the current density cannot be increased further. For a planar diode with gap d and voltage V, the maximal current density limited by space charge is proportional to $V^{3/2}/d^2$ [18]. For a laser-driven photocathode rf gun, the maximal current density is proportional to the external field. Therefore, higher accelerating field gradient allows extraction of a given charge from a source with smaller area that reduces thermal emittance (given the same transverse thermal energy at the cathode) and increases the space charge limited maximal beam brightness [19]. Furthermore, higher 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.

In view of mitigating collective self-interaction of the electrons that may change electron trajectory and energy in the imaging system to degrade the spatial resolution, a high beam energy is required. In general, the transverse space charge force that changes particle's trajectory is proportional to $1/\gamma^2$ due to the cancelation of electric and magnetic forces, where γ is the relativistic factor of the beam (see, for example [20]). The longitudinal space charge force that changes particle's energy is proportional to $I'(z)/\gamma^2$, where $I'(z)$ is the derivative of the beam current at a longitudinal position z. Therefore, a beam with high energy and flat current distribution is desired for mitigating longitudinal space charge force that may increase beam energy spread. Note, for a coasting beam as in conventional TEMs, the electric and magnetic fields have only transverse components by symmetry. So longitudinal space charge is not an issue of concern for conventional TEMs and the transverse space charge force acts like a weak defocusing lens and can be readily compensated by increasing the strength of the solenoids.

In view of reducing spherical and chromatic aberrations of the imaging system, solenoids with strong strengths to provide a short focal length (thus smaller spherical and chromatic aberration coefficients) are needed. This will also loosen the requirements on beam emittance, energy spread and energy stability.

Based on these considerations, the proposed single-shot u-TEM is based on the accelerator that provides a much higher accelerating field than a conventional TEM. Similar to conventional TEMs, an u-TEM consists of three main systems: electron source, condenser lens system, and imaging system, as shown schematically in Fig. 1. To provide picosecond temporal resolution, the electron beam is produced in a photocathode rf gun illuminated with picosecond laser pulse. The beam energy at the exit of the gun is typically a few MeV, which effectively mitigates the temporal broadening from the space charge effect and makes it possible to preserve the picosecond pulse width during transport to the sample. In our representative design the electron beam is produced in a standard S-band photocathode rf gun (with frequency at 2.856 GHz), and the beam global energy spread is further reduced in a harmonic rf cavity (e.g. C-band cavity with frequency at 5.712 GHz). A solenoid (S) following the gun is used to minimize emittance growth from the space charge effect and to control the beam size. The condenser lens (C) used to focus the beam onto a sample and the imaging system, composed of an objective lens (O), an intermediate lens (I) and a projection lens (P), that magnifies the beam, play similar roles as in conventional u-TEM, except that their strengths are much higher to provide sufficient focusing for MeV beam (depending on beam energy and magnetic field strength, superconducting solenoids may be used). Finally the

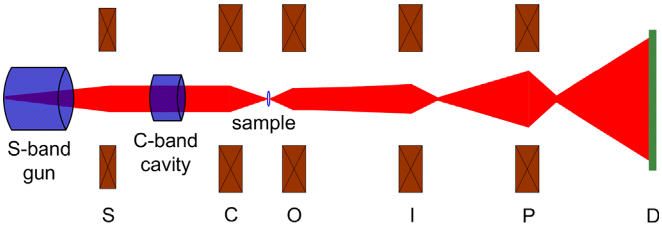

Fig. 1. Schematic of an accelerator-based u-TEM. It consists of a photocathode rf gun to produce picosecond electron beam, a condenser lens (C) to focus the electron beam onto the sample, an objective lens (O), an intermediate lens (I) and a projection lens (P) to magnify the electron beam, and an area detector (D) to measure the magnified beam distribution.