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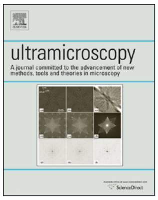

Full length article

GHz laser-free time-resolved transmission electron microscopy: A stroboscopic high-duty-cycle method

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ABSTRACT

A device and a method for producing ultrashort electron pulses with GHz repetition rates via pulsing an input direct current (dc) electron beam are provided. The device and the method are based on an electromagnetic-mechanical pulser (EMMP) that consists of a series of transverse deflecting cavities and magnetic quadrupoles. The EMMP modulates and chops the incoming dc electron beam and converts it into pico- and sub-pico-second electron pulse sequences (pulse trains) at >1 GHz repetition rates, as well as controllably manipulates the resulting pulses. Ultimately, it leads to negligible electron pulse phase-space degradation compared to the incoming dc beam parameters. The temporal pulse length and repetition rate for the EMMP can be continuously tunable over wide ranges.

Applying the EMMP to a transmission electron microscope (TEM) with *any* dc electron source (e.g. thermionic, Schottky, or field-emission source), a GHz stroboscopic high-duty-cycle TEM can be realized. Unlike in many recent developments in time-resolved TEM that rely on a sample pumping laser paired with a laser launching electrons from a photocathode to probe the sample, there is no laser in the presented experimental set-up. This is expected to be a significant relief for electron microscopists who are not familiar with laser systems. The EMMP and the sample are externally driven by a radiofrequency (RF) source synchronized through a delay line. With no laser pumping the sample, the problem of the pump laser induced residual heating/damaging the sample is eliminated. As many RF-driven processes can be cycled indefinitely, sampling rates of 1–50 GHz become accessible. Such a GHz stroboscopic TEM would open up a new paradigm for *in situ* and *in operando* experiments to study samples externally driven electromagnetically. Complementary to the lower (MHz) repetition rates experiments enabled by laser photocathode TEM, new experiments in the multi-GHz regime will be enabled by the proposed RF design. Because TEM is also a platform for various analytical methods, there are infinite application opportunities in energy and electronics to resolve charge (electronic and ionic) transport, and magnetic, plasmonic and excitonic dynamics in advanced functional materials. In addition, because the beam duty-cycle can be as high as $\sim 10^{-1}$ (or 10%), detection can be accomplished by commercially available detectors.

In this article, we report an optimal design of the EMMP. The optimal design was found using an analytical generalized matrix approach in the thin lens approximation along with detailed beam dynamics taking actual realistic dc beam parameters in a TEM operating at 200 keV.

1. Introduction

The past 10 years has seen enormous advancement in time-resolved transmission electron microscopes (TEMs), driven largely by advances in pulsed laser systems. Time-resolved ultrafast TEM (UTEM) [1] and dynamic TEM (DTEM) [2] are steadily establishing new measurement capabilities to observe, understand and control

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solid or soft materials at the most basic level under equilibrium, far-from-equilibrium, extreme, or *in situ* and *in operando* conditions. In a broader context, UTEM and DTEM are electron methods to probe structural dynamics. Even though electrons interact with matter 10^5 times more strongly than photons (electrons are $\sim 10^5$ stronger in scattering power than photons), ultrafast science instrumentation is currently dominated by synchrotron x-ray and x-ray free electron laser facilities, as well as by the use of rare and highly customized high harmonic generation laser microscopes. Since electron- and photon-based tools are fundamentally different, a comparison can be drawn using a combined resolution metric called space-time resolution (STR, spatial resolution times temporal resolution) – in both classes STR can be $\leq 10^{-20}$ m·s. Nevertheless, there are two other parameters that are important: the sampling rate (repetition rate) and beam duty cycle of the electron or photon pulse sequences. In the modern technological era, many breakthroughs rely on understanding how advanced nanoscopic materials or devices operate at rates approaching or exceeding 1 GHz. For example, spintronics exploit ferromagnetic resonance in magnetic materials which occurs in the GHz regime [3]. Understanding how magnetic eigenmodes and magnons interact with real material, defects, and interfaces is a major impendent to the advancement of spintronics. Another example is MEMS/NEMS, which exploit mechanical oscillation of micro-/nano-structures in the MHz and GHz [4]. *In operando* measurements of mechanical properties of these structures will lead to breakthroughs for smaller and faster devices. These examples illustrate the need for adequate instrumentation. At the moment, none of the aforementioned techniques/tools is able to provide GHz-scale sampling rates.

Factors limiting sampling rates at light sources are complex, and even if they are solved, limited beam time allocations will still limit ubiquity of x-ray GHz methods. In contrast, UTEM and ultrafast electron diffraction [5] approaches can be developed at the scale of single principal investigators, and in this case the barrier to GHz sampling arises from the inherent limitations of driving processes with a pump laser. UTEM is a stroboscopic pump-probe method in which data are repeatedly collected over extended periods of time and thermal load from the pump laser must be handled so a process under study stays reversible [6]. Therefore, even though lasers with higher repetition rates are available, UTEM systems typically operate at much less than 0.1 GHz, and sometimes even at ~ 0.1 MHz, depending on the experiment. In contrast to most laser-driven processes, many processes driven electrically/magnetically or both can be cycled indefinitely at GHz frequencies, one example being switching in a semiconductor device. This class of problems, characterized by electrical stimulus and lack of an extended cool-down time, is largely distinct from the class of problems typically studied in laser-based UTEM systems. Thus a purely electrical approach to pump/probe electron microscopy can complement existing laser-based approaches by reaching much higher repetition rates in the study of processes that can be so driven. Higher repetition rates have an immediate advantage in terms of the amount of time required to accumulate a measured signal because of the potential for much higher duty cycles and thus much higher time-averaged probe current.

As an alternative to the laser-photocathode combination method of producing electron pulses, blanking of a direct current (dc) electron beam can produce periodic electron pulse sequences with a flexible temporal structure that can be perfectly synchronized with the clock signal driving a high-frequency nanoscale device (be it a transistor, a plasmonic laser diode, a nano-electromechanical system, a spin-transfer torque memory, or any other device). The basic principle of such stroboscopic TEM is presented in Fig. 1. A small part of the RF signal to the dc beam pulsing device is diverted to the sample through a phase-locked delay line. Similar to UTEM, issues like heating can be anticipated, but even so, essential difference here is that near- *in operando* examination of devices or device structures is feasible, since their realistic operation is itself electromagnetically driven and long thermal cool-down can be avoided. More specifically, the stroboscopic TEM would reveal the inner workings of such devices in unprecedented ways, by bringing all of the high-resolution imaging, analytical, nanoscale diffraction (including strain measurements), and other capabilities (such as holographic imaging of electric fields or spectroscopic imaging of plasmonic fields combined with tomography, making it truly 4D) of a modern TEM down to the time scale of the device's normal operation. In the method being proposed, the pulsed laser system is eliminated, which significantly reduces system complexity and enhances the ease of use. These are important factors to consider when studying already highly-complicated systems for high-technology applications.

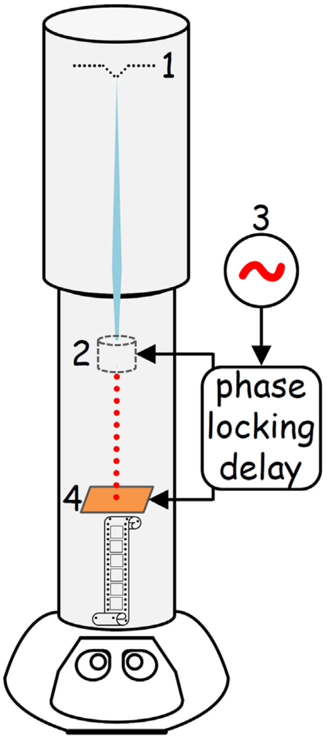

Fig. 1. Basic principle of the stroboscopic laser-free high-duty cycle TEM: 1 is the electron dc source; 2 is a dc beam pulsing device; 3 is an RF source; 4 is the sample.

The first prototypes of time-resolved electron microscopes operating in a stroboscopic regime via blanking the dc beam were dates back to the 1960s. Those were scanning electron microscopes (SEM) used to image electron current propagation in novel (for that time) semiconductor devices such as MOSFETs [7] and Gunn-effect devices [8]. The achievable sampling rates were driven by progress in semiconductor technology – from MHz in 1968 [7] to GHz in 1978 [8]. Thus, by early 1980s stroboscopic SEMs looking into processes with temporal resolution as low as 10 ps were widely prototyped. While ns-pulses at MHz repetition rates were easy to generate using a standard deflecting plate system [7], ps-pulses at GHz repetition rates required development of new strategies, which included a specialty meander traveling-wave line [9], specialty fast capacitors [10], and an RF pillbox cavity with a deflecting mode operating at 1 GHz applied for practical use in electron microscopy for the first time [8]. The aforementioned examples made use of SEMs with lateral resolution $< \sim 1$ µm. While imaging with spatial resolution between 1 Å and 1 nm is routine in a modern TEM, recent interest in time-resolved and pump-probe experiments sets much more challenging