Tunable electron beam pulser for picoseconds stroboscopic microscopy in transmission electron microscopes

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ABSTRACT

For two decades, time-resolved transmission electron microscopes (TEM) have relied on pulsed-laser photoemission to generate electron bunches to explore sub-microsecond to sub-picosecond dynamics. Despite the vast successes of photoemission time-resolved TEMs, laser-based systems are inherently complex, thus tend not to be turn-key. In this paper, we report on the successful retrofit of a commercial 200 keV TEM, without an external laser, capable of producing continuously tunable pulsed electron beams with repetition rates from 0.1 GHz up to 12 GHz and a tunable bunch length from tens of nanoseconds down to 10 ps. This innovation enables temporal access into previously inaccessible regimes: i.e., high repetition rate stroboscopic experiments. Combination of a pair of RF-driven traveling wave stripline elements, quadrupole magnets, and a variable beam aperture enables operation of the instrument in (1) continuous waveform (CW) mode as though the instrument was never modified (i.e. convention TEM operation mode, where the electrons from the emission cathode randomly arrive at the sample without resolvable time information), (2) stroboscopic (pump-probe) mode, and (3) pulsed beam mode for dose rate sensitive materials. To assess the effect of a pulsed beam on image quality, we examined Au nanoparticles using bright field, high-resolution TEM imaging and selected area diffraction in both continuous and pulsed-beam mode. In comparison of conventional TEMs, the add-on beam pulser enables the observation of ultrafast dynamic behavior in materials that are reversible under synchronized excitation.

1. Introduction

In the previous two decades, important technological advancements have expanded the range of temporal resolution in transmission electron microscopes (TEM) [1,2]. Direct-counting and single-electron detectors have revealed dynamics in the millisecond timescale, and laser-assisted photoemission microscopes [3] combined with beam scanning, spatially-parsed large area detectors [4], and sparse-sensing algorithms [5], can now unlock phenomena at the µs to femtosecond (fs) time-scales. Further optimization of the photoemission stage and beam bunching technologies promise to extend the temporal resolution into the deep fs and even attosecond regime [6–8].

Beyond pulsed laser-based approach, others have proposed to use dielectric RF cavities to modulate and pulse the electron beam [9–11]. However, a resonant cavity, by definition, operates at a particular resonance frequency; it is not a broadband device and cannot be arbitrarily tuned without significant power loss. The resulting pulsed beam (e.g., the probe beam in a stroboscopic mode) is therefore also single-frequency and not broadly applicable to subjects with dynamic periods beyond the probe frequency. In addition the resonance frequency of a dielectric cavity is known to be prone to thermal drift [12,13], thus additional engineering strategies are required for the probe frequency-drift mitigation.

Advancing our earlier concept paper [14], we recently developed a RF-driven electron beam pulser system and deployed it on a JEOL JEM-2010F TEM (a 200 keV Schottky-emission instrument) [15]. The Pulser is shown in Fig. 1a as the element within the dotted square. The Pulser is inserted between the electron gun and the microscope’s first condenser lens. Its construct is radiation and vacuum compatible with the manufacturer’s requirement. Fig. 1b is a schematic of the internal elements of the Pulser, which consists of three principal elements: electron beam modulator (K1) /demodulator (K2), beam chopping aperture (A), and quadrupole magnets (Q1 through Q3). The incoming CW (unmodified) beam is assumed to enter the Pulser on-optical axis (z-
When the Pulser is activated, several diode RF amplifiers (not shown) are excited to modulate the incoming electron beam. The modulator circuitry can independently adjust the repetition rate (Prf) and the center frequency (rf) of the RF signal. Here, Prf is set to be equal to 50 MHz, and rf is set to 2 GHz for the examples shown in this paper. The tuning range of rf is limited by the maximum RF power output from the pulser. The curve in Fig. 1c shows the full band, which ranges from 100 MHz up to 12 GHz. In this mode the duty cycle D, as illustrated by Fig. 1d, is the preferred method because increasing D instead of Prf will reduce the induced transverse momentum of the electrons leaving K1. This is particularly important when a large aperture is used. The dual-beam mode, K2 (at 90° phase lag with respect to K1) adds to the induced transverse momentum of the electrons from K1. The result is two spatially separated pulsed beams, both with the repetition rate of 2Prf as illustrated by Fig. 1d. In the dual-beam mode, the electron pulses land in two separate spatial locations (that are temporarily nPrf out of phase) as observed on the image plane. This mode is primarily used for the characterization of the Pulser, however, it may be possible to probe the specimen at Prf instead of 2Prf by eliminating one of the twins with an aperture.

The most straightforward use case of the Pulser is the stroboscopic pump-probe experiments, which is an extension of the pulsed-beam mode (as shown in Fig. 1e). A daughter RF signal is tapped from the primary K1 drive signal to provide stimulus or "pump" to the sample through a phase-locked delay line while the sample is probed by synchronized electron pulses. By changing the electrical delay between the pump signal and the probing beam, the complete time evolution of a cyclical dynamic phenomenon can be captured. The full band, 100 MHz < 2Prf < 12 GHz, is available in the stroboscopic pump-probe mode.

The core element enabling the unique capabilities of the Pulser is a novel Traveling Wave Metallic Comb Stripline (TWMCS). The TWMCS operates in a traveling-wave mode, driven by a differential RF signal from one end and terminated at the other end with 50 Ω loads. As shown in Fig. 2a and b, the TWMCS consists of two metallic combs with their teeth facing each other. In this geometry, only the Transverse EM wave mode is allowed to propagate along the beam channel (optic axis). The Transverse EM mode has neither longitudinal electric nor magnetic component, thus in principle no induced energy spread to the incoming beam. As the transverse EM wave propagates, its velocity can be altered by the teeth structure. By optimizing the teeth thickness, spacing, and height, the phase velocity of EM wave can be adjusted to synchronize with the speed of the probe electrons so that they can travel along the z direction. (c) The simulated electron displacement vs. theRF phase for an electron traveling through the TWMCS. (d) Comparison the transverse wave dispersion in the TWMCS and the linear velocity of a 200 keV electron; e) The simulated RF power transmission coefficient S12 over a 10 GHz span.
3. Experimental demonstrations of the pulser

In Fig. 4, the tunability of the pulse length was demonstrated by varying the K1 voltage in the dual beam mode. In this mode, the two electron bunches that are \( n \) apart in time at the exit of the beam chopping aperture are alternately placed near peaks and troughs of the K2 RF drive so that they are transversely kicked in opposite directions. Thus the beam separation distance \( D_{\text{beam}} \) as measured on the detector is a reasonable proxy for temporal resolution, and the number of pixels in \( D_{\text{beam}} \) can be mapped to a time unit. However, the individual electron pulse length cannot be obtained directly from images in the dual-beam mode. For a short pulse length, if it only occupies a small fraction of the RF phase near the peak or trough, most electrons in a pulse experience a similar kicking strength, therefore, their physical size (beam width, \( W_{\text{beam}} \)) is less sensitive to the temporal profile. In contrast, for a longer pulse length, \( W_{\text{beam}} \) is strongly correlated to the real bunch length. For this reason, a clean interpretation of pulse length must be accompanied with experimental results obtained from an earlier 200 keV electron gun bench test. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Another way to tune the duty cycle is by varying the diameter of the beam chopping aperture. In the present design, the beam chopping aperture strip contains five aligned holes with different diameters, and it is mounted on piezoelectric X-Y linear stage. In Fig. 5, we picked two different diameters (\( \Phi = 40 \mu m \) and \( \Phi = 25 \mu m \)) to demonstrate tunability of the beam duty cycle. In the dual-beam mode, we kept \( f_0 \) constant at 5 GHz, i.e., the two beamlets are 100 ps apart. The duty
cycle, $D$, can be characterized by the ratio of pulse width ($W_{\text{beam}}$) over $D_{\text{beam}}$. $D$ for $\Phi \approx 40 \mu m$ is 0.24 in Fig. 5a and is 0.16 in Fig. 5b for $\Phi \approx 25 \mu m$. The reduction of duty cycle as a result of changing the aperture size from 40$\mu m$ to 25$\mu m$ is 66% which is very close to the ratio of two aperture sizes.

Finally, to test the performance of the TEM after the Pulser installation, we acquired bright-field images and diffraction patterns of a standard Au nanoparticle sample using both CW and pulsed-beam modes at $f_0 = 5.2$ GHz (refer to Fig. 6). The choice of 5.2 GHz was arbitrary. Fig. 6a is a bright-field image of Au nanoparticles at an indicated magnification of 200 kX obtained using a CW beam (Pulser off). The morphology of the nanoparticles and the amorphous nature of the carbon support can be seen clearly. We noticed that at a higher magnification of 600 kX, the lattice fringes are still visible in the CW mode (data not shown), suggesting the original spatial resolution of the TEM was not compromised by the Pulser addition. Fig. 6b is another bright-field image of the same area as Fig. 6a, captured under the same imaging condition, same image integration time (2s), but in a modified pulsed-beam mode in which K2 was deactivated (K2 was off due to equipment malfunction during our limited testing window, but it has since been resolved for a different microscope). Nanoparticles are still imaged with reasonable quality. The little contrast blur under the pulsed beam mode is mainly due to the much-lowered electron dose and the possible existence of a small transverse momentum of the pulsed electrons due to an inactive K2. Fig. 6c and d show a comparison of diffraction patterns under the CW and pulsed beam modes with an indicated camera length of 20 cm. All the characteristic Bragg rings of Au nanoparticles can be clearly distinguished in the diffraction patterns under both CW and the modified pulsed-beam modes, and the last ring at 0.57 nm on both diffraction micrographs can be readily resolved, indicating the comparable high quality of diffraction under both operation modes. Again, for the same reasons previously described, the Bragg rings do appear less sharp compared to those produced in CW mode, but the higher spatial frequencies are still preserved. The bright-field imaging and diffraction comparison tests demonstrated that the function and performance of the instrument after the column modification did not exhibit significant changes. Because the RF source, serving as a universal clock, is a versatile electrical signal, the pump (specimen excitation) synchronization using the RF tap can be deployed using a robust ecosystem of specialty specimen holders already in the market place. Therefore, the tunable RF pulsed TEM is potentially a powerful platform for ps-stroboscopic electron imaging and diffraction for exploring ultrafast dynamical processes.

In conclusion, we have demonstrated a new device which retrofits the conventional TEM and is able to chop the electron beam out of an electron gun at an ultrafast and tunable rate, thus forms electron pulses with nano-seconds to pico-seconds duration and MHz to GHz repetition rate. This device is unique because unlike a microwave cavity, it is broadband tunable. It preserves the original beam coherence, and can return to the conventional modes of operation by simply switching off the Pulser RF control. The ultrashort pulsed electrons in combination with the synchronized sample excitation enables pump-probe experiments including (but are not limited to) ferromagnetic resonance in magnetic materials, magnons, skyrmions, and atomic measurements in MEMS and NEMS systems.

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References

[15] Certain commercial equipment, instruments, or materials are identified in this paper to specify the experimental procedure adequately. Such identification is not intended to imply recommendation or endorsement by the National Institute of Standards and Technology, nor is it intended to imply that the materials or equipment identified are necessarily the best available for the purpose.