Ultramicroscopy 188 (2018) 85–89

Contents lists available at ScienceDirect

Ultramicroscopy

journal homepage: www.elsevier.com/locate/ultramic

High quality ultrafast transmission electron microscopy using resonant microwave cavities

W. Verhoeven[a], J.F.M. van Rens[a], E.R. Kieft[b], P.H.A. Mutsaers[a], O.J. Luiten[a,*]

[a] *Department of Applied Physics, Coherence and Quantum Technology Group, Eindhoven University of Technology, P.O. Box 513, 5600, MB Eindhoven, Netherlands*
[b] *Thermo Fisher Scientific, Achtseweg Noord 5, 5651, GG Eindhoven, Netherlands*

ARTICLE INFO

Article history:
Received 1 August 2017
Revised 7 March 2018
Accepted 9 March 2018
Available online 10 March 2018

Keywords:
Ultrafast transmission electron microscopy
Pump–probe
Microwave cavities
Coherent ultrashort electron pulses

ABSTRACT

Ultrashort, low-emittance electron pulses can be created at a high repetition rate by using a TM_{110} deflection cavity to sweep a continuous beam across an aperture. These pulses can be used for time-resolved electron microscopy with atomic spatial and temporal resolution at relatively large average currents. In order to demonstrate this, a cavity has been inserted in a transmission electron microscope, and picosecond pulses have been created. No significant increase of either emittance or energy spread has been measured for these pulses. At a peak current of 814 ± 2 pA, the root-mean-square transverse normalized emittance of the electron pulses is $\varepsilon_{n,x} = (2.7 \pm 0.1) \cdot 10^{-12}$ m rad in the direction parallel to the streak of the cavity, and $\varepsilon_{n,y} = (2.5 \pm 0.1) \cdot 10^{-12}$ m rad in the perpendicular direction for pulses with a pulse length of 1.1–1.3 ps. Under the same conditions, the emittance of the continuous beam is $\varepsilon_{n,x} = \varepsilon_{n,y} = (2.5 \pm 0.1) \cdot 10^{-12}$ m rad. Furthermore, for both the pulsed and the continuous beam a full width at half maximum energy spread of 0.95 ± 0.05 eV has been measured.

1. Introduction

Ultrashort high quality electron pulses at energies ranging from 30 to 200 keV have become a useful and powerful tool to investigate dynamical systems on sub-picosecond timescales through diffraction [1], imaging [2] or spectroscopy [3], offering a vast amount of new information. Typically, inside an ultrafast transmission electron microscope (UTEM) electron pulses are extracted from a photocathode using an intense pulsed laser. Accurately timed with a clocking laser pulse, dynamic processes can then be investigated with pump–probe measurements. Using photoemission, a very large operational parameter-space can be spanned [4]. Furthermore, by using sideways illumination of a Schottky emitter, the emission characteristics of the source are maintained, allowing for high quality electron pulses to be created [5].

Although photoemission is commonly used in UTEM systems, there is an interesting alternative to use a blanking method, where a continuous beam is periodically swept across a slit or aperture [6–8]. Creating pulses in this way has the advantages that amplified laser systems are no longer required, and that no intrusive alterations to the source have to be made. Instead, the system benefits from the vast amount of research done on state-of-the-art continuous sources, including recent developments that promise a higher brightness in the future [9]. Furthermore, any possible instabilities in electron emission due to the intrinsic pointing stability of a drive laser are circumvented.

Recently, it has been shown that pulsing a beam can be done using a microwave cavity oscillating in the TM_{110} mode while maintaining the low emittance of a continuous source [10–12]. This can be accomplished using a conjugate blanking scheme, where the electron beam is focused at the center of the cavity, allowing for 100 fs pulses to be created with a high beam quality. Since the power in the cavity can easily be adjusted, the pulse length can also be changed without influencing the electron emission process. In Fig. 1(a) this chopping principle is shown.

In order to perform pump–probe experiments, the phase of these microwave cavities can be accurately synchronized to a pump laser pulse. Using state-of-the-art synchronization schemes, timing jitter between the electron pulses and the laser pulses can be suppressed to levels well below 100 fs [13,14].

Alternatively, it has been proposed to use a microwave signal as a pump pulse to drive electronic or semiconductor devices for laser-free stroboscopic imaging with repetition rates in the GHz regime [15]. This is an interesting aspect of using microwave cavities, as they can provide a higher repetition rate and therefore a higher average current for samples with a fast relaxation time.

* Corresponding author.
E-mail address: o.j.luiten@tue.nl (O.J. Luiten).

https://doi.org/10.1016/j.ultramic.2018.03.012

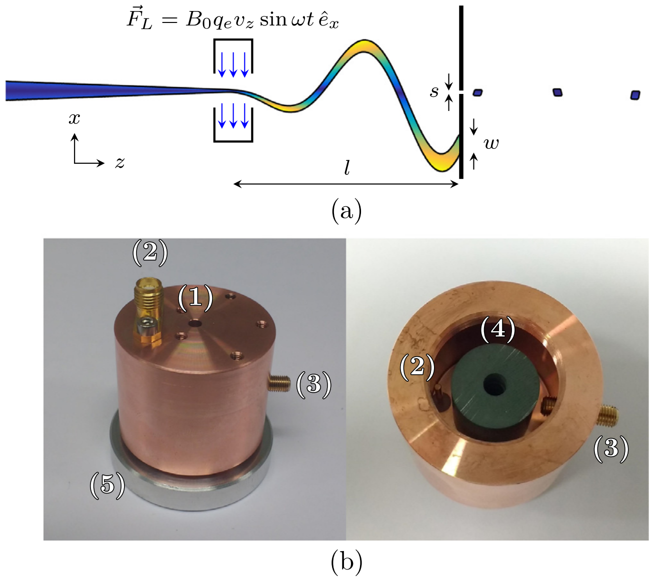

Fig. 1. (a) General principle of the creation of pulses using a TM_{110} deflection cavity, where a continuous beam is deflected over a chopping aperture. Definition of the parameters is discussed in Section 2.2. (b) A typical cavity, with (1) the entrance aperture, (2) the antenna, (3) the tuning stub, (4) the dielectric material, and (5) the lid used to close the cavity. Shown left is the side of the cavity, and right is the bottom of the cavity with the lid removed.

For samples with slower relaxation times, it has been proposed to use two perpendicular deflecting modes at different frequencies, which can be placed in a single cavity [10]. Electrons will then be created at the difference frequency of these modes, allowing for the repetition rate to be lowered to tens of MHz. If lower frequencies are desired, a fast beam blanker can be used to pick specific pulses, which are now separated by tens of ns. In this way, microwave cavities can also provide lower repetition rates for samples with slow relaxation times, allowing for the repetition rate of the setup to be optimized for each experiment.

In order to facilitate the implementation in a TEM column, these deflection cavities can be filled with a dielectric material, which allows for a reduction in both the size and power consumption [16]. Fig. 1(b) shows a typical dielectric filled cavity used for chopping an electron beam. Shown to the left is the outside of the cavity, and to the right is a bottom view of the cavity with the lid removed.

In this paper, the implementation of a TM_{110} deflection cavity in a TEM is presented. Design considerations are discussed, and the performance of a cavity-based UTEM is demonstrated.

2. Theory

2.1. Brightness

An important figure of merit for a charged particle beam is its current density per unit of solid angle, called the transverse brightness. As the solid angle subtended by the beam, and therefore the brightness, depends on the beam energy, the beam quality is often expressed in terms of the *reduced* brightness, which can be defined in differential form as [17]

$$B_r = \frac{1}{V^*}\frac{\partial^2 I}{\partial A \partial \Omega}, \tag{1}$$

with I the current through an area A at a solid angle Ω, and $V^* = (1/2 + \gamma/2)V$ the acceleration voltage V multiplied by a relativistic correction term, with γ the Lorentz factor. The reduced brightness is a conserved quantity during acceleration of the electrons.

Since the differential reduced brightness varies throughout the beam, its maximum on-axis value is often used, called the axial or peak brightness. Within the typical working regime of a microscope, a large portion of the emitted electrons is cut away at the condenser aperture, leading to an approximately uniform current distribution. After focusing the beam at semi-angle α, this then results in a uniform angular distribution and a Gaussian position distribution within the beam waist, so that the peak brightness can be written as

$$B_r = \frac{1}{V^*}\frac{I}{2\pi^2\alpha^2\sigma_x\sigma_y} = \frac{|q_e|}{m_e c^2}\frac{I}{4\pi^2\varepsilon_{n,x}\varepsilon_{n,y}}, \tag{2}$$

with σ_x and σ_y the root-mean-square (RMS) size of the beam waist, q_e the electron charge, m_e the electron mass, c the speed of light, and $\varepsilon_{n,\,x}$ and $\varepsilon_{n,\,y}$ the RMS normalized emittance in the x and y direction respectively, given by

$$\varepsilon_{n,x} = \frac{1}{m_e c}\sqrt{\langle x^2\rangle\langle p_x^2\rangle - \langle x p_x\rangle^2} \approx \frac{\gamma v_z}{c}\sqrt{\langle x^2\rangle\langle x'^2\rangle - \langle x x'\rangle^2}, \tag{3}$$

with v_z the velocity, p_x the transverse momentum and $x' = v_x/v_z$ the angular distribution of the particles. In this equation, $\langle \ldots \rangle$ indicates the averaging over a distribution.

2.2. Beam chopping

The main advantage of using a microwave cavity is that the low emittance of the continuous beam is maintained in pulsed mode. This is only the case when using the cavity in a conjugate blanking scheme, in which all electrons deflected by the cavity originate from the same virtual image. For a regular beam blanker conjugate blanking is achieved by placing a crossover in the pivot point of the blanker.

Inside a microwave cavity the fields vary rapidly compared to the transit time of the electrons, so that it is no longer possible to distinguish a single pivot point. However, it can be shown that it is still possible to maintain the virtual image by proper placement of a crossover [10,12]. For a beam chopped by an on-axis aperture, the optimal longitudinal position of this crossover is at the center of the cavity.

This is also shown in Fig. 1(a), where the beam is focused at the center of the cavity. As a result, it arrives at the chopping aperture with a certain width w. Sweeping this beam with a magnetic field amplitude B_0 and an angular frequency ω over an aperture with width s results in a full width at half maximum (FWHM) pulse length of

$$\tau = \frac{\gamma m_e (s + w)}{4|q_e| l B_0 \sin\left(f\frac{\pi}{2}\right)}, \tag{4}$$

where l is the distance to the chopping aperture, and $f = L_{cav}/L_{max}$ the fractional length of the cavity L_{cav} compared to the maximum useful cavity length $L_{max} = v_z\pi/\omega$ for which electrons feel exactly half the oscillation period. From this equation it can be seen that in order to create short pulses, the focusing angle has to be small to restrain w from becoming too large.

Besides deterioration of the brightness, increase of the energy spread is also an important effect that has to be considered. Unfortunately, electrons moving through a cavity will probe the off-axis electric fields of the TM_{110} mode. This will not only cause the total beam energy to change, but also the energy spread to increase. Focusing the beam at the center of the cavity minimizes this increase in energy spread, but does not completely eliminate it.