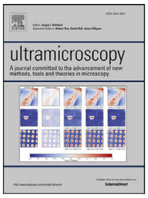
Ultramicroscopy 000 (2017) 1–7

Contents lists available at ScienceDirect

Ultramicroscopy

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



A novel electron mirror pulse compressor

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ARTICLE INFO

Article history:
Received 29 November 2016
Revised 5 April 2017
Accepted 9 May 2017
Available online xxx

Keywords:
Ultrafast electron diffraction
Dynamic transmission electron microscopy
Pulse compressor
Electron mirror
Magnetic beam separator
Aberration correction
Electron optics

ABSTRACT

An electron mirror-based pulse compressor design has been developed for improving the temporal resolution of dynamic/ultrafast transmission electron microscopes and ultrafast electron diffraction cameras. The improvement will enable electron microscopes and diffraction cameras to better resolve the dynamics of reactions in the areas of solid state physics, chemistry, and biology. The design utilizes a combination of mirror optics and a magnetic beam separator, which exploits the symmetry inherent in reversing the electron trajectory in the mirror in order to compress the pulse. This system can also simultaneously correct the spherical and chromatic aberration of the objective lens for improved spatial resolution. For pulsed experiments with a practical bunch charge, the correction of the chromatic aberration coefficient counters the spread in the electron energies induced by the space charge of the pulse to make possible the probing of the sample with high spatial resolution. The pulse compressor can accommodate pulses with a range of electron densities and energy spreads. Furthermore, it is designed to fit into both ultrafast electron diffraction cameras and dynamic/ultrafast transmission electron microscopes. Consequently, this instrument is suitable for enhancing the study of the structure, composition, and bonding states of new materials at ultrafast time scales.

1. Introduction

Advanced materials are one of the key drivers across a wide spectrum of applications including biotechnology, semiconductors, as well as energy storage and transfer. The successful implementation of new materials in these applications depends on the nanoscale exploration of their structures, composition, and chemical properties in materials science R&D. These explorations have been the primary scientific motivations for both the improvements in spatial resolution of electron microscopes to the sub-Angstrom regime and the new developments in element and oxidation state specific contrast [1]. In addition to these more traditional frontiers for development, there has recently been parallel efforts aimed at improving the temporal resolution of electron microscopes through the implementation of pulsed photoelectron sources [2–4]. The dramatic improvements in the temporal resolution of electron microscopy instrumentation over the last decade has made visible many atomic-scale processes that occur on timescales approaching hundred femtoseconds [5–9].

Pulsed electron techniques, like ultrafast electron diffraction (UED) and dynamic transmission electron microscopy (DTEM), have achieved an unprecedented combination of spatial and temporal resolution by introducing ultrashort photoelectron pulses into electron microscope instrumentation. This involves illuminating a photocathode with an ultrashort laser pulse, accelerating the photoemitted electrons, and illuminating a specimen with the ultrafast electron pulse to obtain time-resolved electron diffraction patterns or images. Electron sources most commonly used in UEDs and DTEMs utilize photo-electron emission from metal photocathodes, typically Cu or W, which produce an ultrafast electron pulse with an initial pulse width in the range of 1 to 100 femtoseconds. Unfortunately, repulsive Coulomb interactions between the electrons broaden the temporal and spatial extent of the pulse during the travel to the specimen [10–12]. A large fraction of the Coulomb broadening occurs near the cathode surface, before the electrons are accelerated to the beam energy. The Coulomb interactions increase the beam energy spread (known as the Boersch effect) from a fraction of an electron-Volt to hundreds or even thousands of electron-Volts. The Boersch effect has a two-fold impact on the electron optics: it spreads the arrival time window of the pulse from tens of femtoseconds to picoseconds; and it increases the objective lens chromatic aberration, which reduces the spatial resolution. These adverse effects have placed a limit on the total amount of charge that can be used in the pulse, thereby placing burdensome requirements on the number of pulses for obtaining data with adequate signal-to-noise. However, Siwick [10] has shown that the pulse of electrons emitted by the photocathode

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http://dx.doi.org/10.1016/j.ultramic.2017.05.004
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Please cite this article as: M. Mankos et al., A novel electron mirror pulse compressor, Ultramicroscopy (2017), http://dx.doi.org/10.1016/j.ultramic.2017.05.004

- given sufficient propagation - becomes highly correlated in the space of the electron kinetic energy and travel time. This correlation makes it possible to reverse much of the longitudinal spread in the pulse.

1.1. *Pulse compression techniques*

Several approaches have been developed in the past twenty years that minimize the longitudinal expansion of the pulse. One option is to reduce or eliminate this broadening by greatly reducing the number of electrons in the pulse [13]. However, the lower electron count increases the data acquisition time and requires that the sample be reproducibly pumped and probed 10^6 times to obtain images or diffraction patterns of sufficient quality. Another option is to increase the beam energy into the MeV range to minimize the electron interaction time and thus the Coulomb interactions [14]. However, electrons with such high kinetic energies not only have low cross-sections for scattering within the sample, they also cause radiation damage on many types of specimens when they do interact. Hence, both approaches tend to degrade the sample as it is being diagnosed, thereby motivating the search for alternatives.

In the last decade, Radio Frequency (RF), 360° deflector, and mirror pulse compressors have been developed that overcome the pulse broadening without reducing the number of electrons in the pulse and without increasing the beam energy above the typical range for TEM (100 keV). In the RF approach, the time-varying field of an RF cavity is used to compress the pulse to the 100 femtosecond level [15]. As the longitudinally-broadened pulse enters the cavity, the leading higher energy electrons are decelerated, while the trailing lower energy electrons are accelerated. The cavity can be easily incorporated into a rectilinear column. The challenge with RF methods is in the synchronization of the RF cavity with the pulsed laser [16,17] and in maintaining stability. These methods lack the technical simplicity of static electron optics.

In the 360° deflector approach, the pulse is made to traverse a circular path by either dipole magnets [18] or electrostatic capacitors [19]. For the configuration with dipole magnets, the deflection disperses the beam in energy, forcing the higher energy electrons to traverse a longer path, resulting in pulse compression. Here, temporal compressions to about $\sim$200 femtoseconds has been achieved. Although the incident and exiting electron paths are parallel, they do not coincide due to a required out of-plane offset between the magnets in the first and the second half of the deflection. This offset introduces aberrations and makes alignment and beam set up challenging. For the configuration with an electrostatic, spherical capacitor, a central-force field produces a longer path for the higher energy electrons in order to compress the electron pulse. This system has mirror symmetry, which cancels the deflector aberrations, and has compact in-line construction, which provides a straight line-of-sight from source to sample. However, the symmetry that is introduced by the capacitor also results in a temporal focus of the pulse at its midplane, after a 180° deflection, which significantly enhances the effects of Coulomb repulsion.

In the mirror compressor approach, there have been two proposals so far [20,21]. The two share a common feature: the pulse enters the mirror at a relatively large entrance angle in order to separate the incoming and outgoing pulse. The skewed entry complicates the column alignment, as there is no direct line of sight from the photocathode to the specimen. In addition, the large entrance angle introduces significant aberrations and a significant tilt of the pulse front, which further complicates the experimental setup. Last, the presence of a turnaround point at the mirror reflection plane has the potential for inducing undesirable Coulomb blur: as the electrons slow down in the mirror, they are more likely to get deflected (Loeffler effect) or loose or gain energy (Boersch effect) due to interactions with neighboring electrons, which results in added spatial blur and energy spread of the pulse.

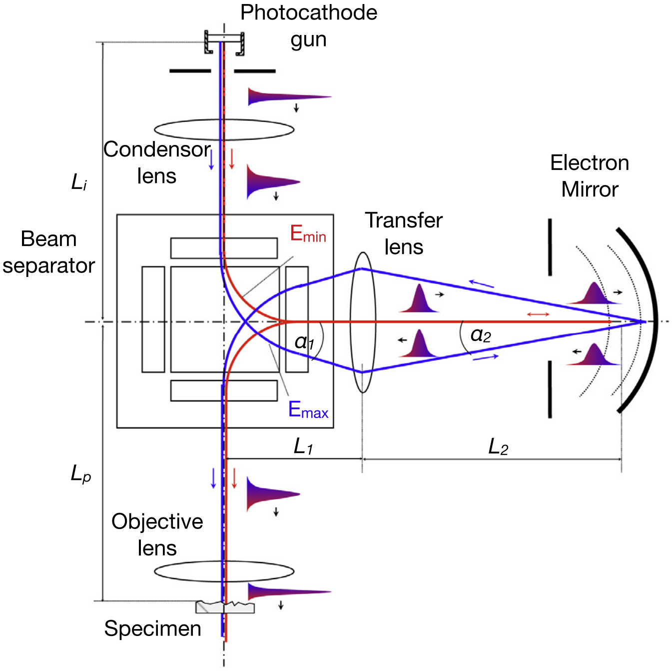

Fig. 1. Schematic layout of the mirror pulse compressor.

2. Electron optics of the mirror pulse compressor

The novel mirror pulse compressor proposed here aims to overcome the disadvantages of the above-mentioned competing designs while also implementing the desired features and providing an additional capability: aberration correction. A layout of the electron pulse compressor is shown in Fig. 1. The layout represents the conceptual design of the pulse compressor and includes only the critical electron-optical elements needed to illustrate the imaging principles. Many of the elements typically present in a detailed column design, e.g. transfer and field lenses, alignment and stigmation coils, etc. are omitted here for clarity.

The key feature of this novel pulse compressor is the combination of a magnetic beam separator [22,23] that does not introduce significant aberrations and an electron mirror, which can compress pulses with varying number of electrons and can be tuned to match different column geometries [24]. This approach utilizes static electron-optical components to achieve the pulse compression and thus does not suffer from the jitter problems encountered in the RF approaches. Another key advantage is the use of a magnetic beam separator that allows the compressor to be readily incorporated into the straight column axis of existing UED and DTEM instruments, thereby significantly simplifying the column alignment and improving its stability. The beam separator also allows the pulse to enter the electron mirror at normal incidence, which minimizes the aberrations introduced by the mirror. The symmetry in the geometry of the unit comprising the beam separator and the mirror eliminates any energy dispersion that would further deteriorate the quality of the pulse front. The beam envelope in the mirror is carefully designed to minimize Coulomb interactions by spreading the pulse in both the longitudinal and transverse directions at the turnaround point. Furthermore, the mirror provides the capability to correct the aberrations of the objective lens in the UED and DTEM instruments.

Please cite this article as: M. Mankos et al., A novel electron mirror pulse compressor, Ultramicroscopy (2017), http://dx.doi.org/10.1016/j.ultramic.2017.05.004