Ultrafast electron microscopy for investigating fundamental physics phenomena

Prawesh Dahal[a], Akrit Mudvari[a], Aashwin Basnet[a], Parker W. Brown[a], Brett Barwick[a]*
[a]Dept. of Physics, Trinity College, Hartford, CT USA 06106

ABSTRACT

Recent experiments have shown that by combining a femtosecond laser with an electron microscope, dynamics at the nanoscale with femtosecond temporal resolution can be followed. These ultrafast electron microscopy (UEM) techniques have allowed a variety of experimental studies on nanoscale material systems to be conducted, including the ability to image plasmon dynamics in real time. We will discuss how UEM can also be used to study fundamental quantum problems, present some recent experimental results and we will discuss how future experimental improvements will push the temporal resolution from hundreds of femtoseconds down to the single femtosecond regime and below.

Keywords: ultrafast electron microscopy, femtosecond electron point projection microscopy, electron orbital angular momentum, Kapitza-Dirac effect, electron pulse compression

INTRODUCTION

Overview

Microscopy has long served as an integral part of scientific research and its technology has advanced with time, developing for instance from simple devices such as the optical microscope to more complex ones such as scanning and transmission electron microscopes (SEM and TEM, respectively). Further modifications in the functioning of electron microscopes have opened up a field of research dubbed ultrafast electron microscopy or 4D electron microscopy; UEM techniques allow images to be captured with high temporal and spatial resolutions, which are currently in the *femtosecond* (fs) and *nanometer* (nm) regimes respectively[1]. In a UEM, a femtosecond laser is integrated with the electron microscope where the short laser pulses are split with one pulse used to generate an electron pulse and the second laser pulse directed to a specimen that is being imaged in the microscope. The laser pulse that is focused on the specimen initiates the dynamics which are then imaged by femtosecond electron pulses. By delaying the time between the dynamics initiating pump laser pulse and the electron probe pulse images can be captured at well-defined delay times[2]. This technique can achieve a frame rate of approximately 10^{12} frames per second, which provides the aforementioned temporal resolution, while maintaining nanometer spatial resolution[1]. This pump-probe UEM capability has been extended to a variety of electron microscopy techniques including diffraction, dark field imaging, electron energy loss spectroscopy and imaging, electron tomography, scanning TEM, SEM, and others[1]. With UEM, the characterization of materials and the study of laser induced dynamics in condensed matter systems are achieved and in general the responses of samples to intense optical stimuli can be observed on the nano-scale and smaller. Investigating and developing electron pulse compression techniques are important in extending the capabilities of UEM to the few fs temporal regime which will open up a variety of new possible experiments.

Methods and fundamental applications

One technique that has been made possible with UEM's based on TEM's is the recently developed photon-induced near-field electron microscopy (PINEM)[3]. PINEM is an imaging technique that exploits the ability of electrons to absorb/emit photons that have been radiated from a stimulated nanoscale sample[3]. This absorption/emission is mediated by the plasmonic near-field and by selecting only the electrons that have absorbed photons to construct images the excited near-field of the nanoscale specimen can be observed[3,4,5,6,7,8]. While UEM and PINEM have numerous condensed matter applications, it can also be used for more fundamental research. An example of UEM being used to study more fundamental quantum behavior has been accomplished recently with PINEM by capturing both the quantized energy exchange of the near-field while at the same time imaging its spatial interference[9]. This simultaneous observation of the

Ultrafast Nonlinear Imaging and Spectroscopy IV, edited by Zhiwen Liu, Iam Choon Khoo, Demetri Psaltis, Kebin Shi,
Proc. of SPIE Vol. 9956, 995604 · © 2016 SPIE · CCC code: 0277-786X/16/$18 · doi: 10.1117/12.2237392

energy quantization and interference behavior is of a fundamental interest and shows the power of UEM to investigate more fundamental quantum effects. A second example demonstrating UEM's flexibility to study more fundamental problems was in the recent observation of Rabbi-oscillations of an electron beam[10]. That study demonstrated that the electron beam can be coherently controlled with plasmonic near fields. The resulting coherent electron packets may be used in quantum information technology research, including studies investigating entanglement in near-field interactions[10].

While most of the previous experimental work has been focused on UEM's with relatively high-energy electrons, low-energy UEM's with sub-keV kinetic energies are also in use and can be advantageous for a variety of reasons. Lower energy electrons have high-scattering cross sections allowing for increased sensitivity in both imaging and diffraction approaches[11]. Furthermore, the high sensitivity of low energy electrons to weak electric fields makes the direct investigation of electrostatic fields around nanoscale systems possible[11]. The advantages of low energy electrons motivate their use in femtosecond point projection microscopy, electron diffraction and other experiments, which can be used to study not only condensed matter systems, but are also uniquely suited for completing fundamental quantum experiments. One experiment that is proposed in more detail below pertains to the use of the Kapitza-Dirac (KD) effect to control the quantum properties of an electron beam[12]. To date the KD effect has only been observed with low energy electrons (~500 eV), motivating the use of low energy UEM to complete KD based experiments. A recent study has proposed that the KD effect can be used to transfer quantized amounts of orbital angular momentum (OAM) from photons to electrons and an experimental demonstration of this effect would be interesting both for fundamental quantum aspects and for the practical use of the pulsed OAM electron beams. These types of experiments require experimental advances in low energy UEM, including the reduction of electron pulse durations and the development of high coherence femtosecond pulse electron beams.

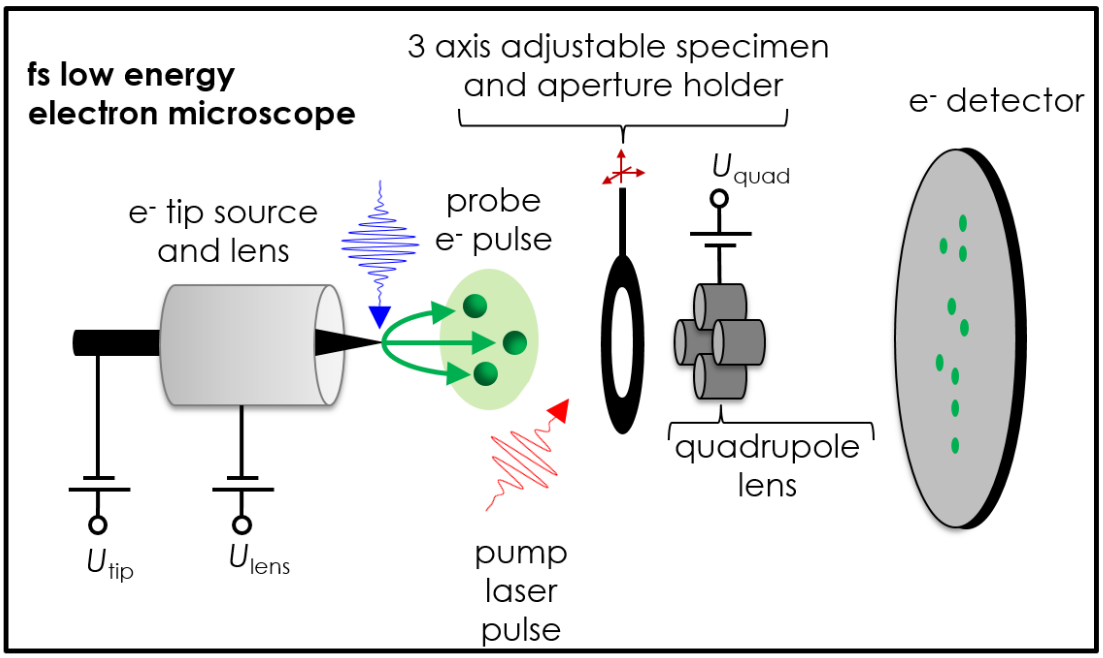

Figure 1. Diagram of a femtosecond low energy electron microscope with a tip-lens source. A laser pulse initiates an electron probe pulse from the emission tip, while the pump laser pulse excites the specimen. The collimation of the electron beam can be controlled with a micro lens which surrounds the tip. Apertures and specimens can be independently positioned in 3-d with micrometer precision with respect to the tip. A quadrupole lens allows the electron beam to be focused as appropriate on the 2-d electron detector.

LOW ENERGY ELECTRON EXPERIMENTAL APPARATUS

Currently, low energy UEM's have reached resolutions of ~100 nm spatial, along with ~100 fs temporal resolution[11,13,14]. These femtosecond low energy electron microscopes generally rely on laser-triggered electron tip sources that can deliver ~100-1000 fs electron pulses onto a specimen and have been used in studies on both ultrafast electronic and structural dynamics[11], including diffraction studies of graphene[14]. However, one limitation of low energy UEM's is that low energy electron pulses are susceptible to pulse dispersion which can limit their resolution in time-resolved experiments[11].

We have developed a low energy UEM, depicted in Fig. 1 that utilizes photoemission from a metal nanotip that provides a coherent femtosecond electron emission source for our microscope. The tungsten nanotips have radii less than 100 nm and are electrochemically etched and prepared by a lamella drop-off technique from polycrystalline tungsten wire[15]. The tips are thoroughly sonicated in order to remove dust particles and to avoid the formation of an oxidation layer. The shape and nature of the tip apex are carefully studied using an SEM to ensure that the tips are both undamaged and of the proper radius. These tips are then mounted within a gold-coated microlens with the length of the tip exposed being approximately equal to the radius of the lens, as shown in Figure 2a. The electrostatic lens is used to focus the electron beam[16], and allows the UEM to be operated in either a diffraction or projection imaging mode Fig. 2a-b.

The photoelectrons are generated by focusing femtosecond laser pulses (~1 nJ, 800 nm, 50 fs) onto the negatively biased tip. The resulting electron pulses are then accelerated towards the grounded sample and in time-resolved experiments a second laser pulse is focused onto the sample after traveling through an optical delay stage. By varying the delay stage pump-probe experiments are possible and either projection images or diffraction patterns, are observed on the multichannel plate phosphor screen detector and recorded with a CCD camera[11,13]. Current experiments are focused on optimizing the microlens/tip, Fig. 2a, for diffraction along with optimization of the quadrupole lens which will allow KD type experiments to be completed in our apparatus.

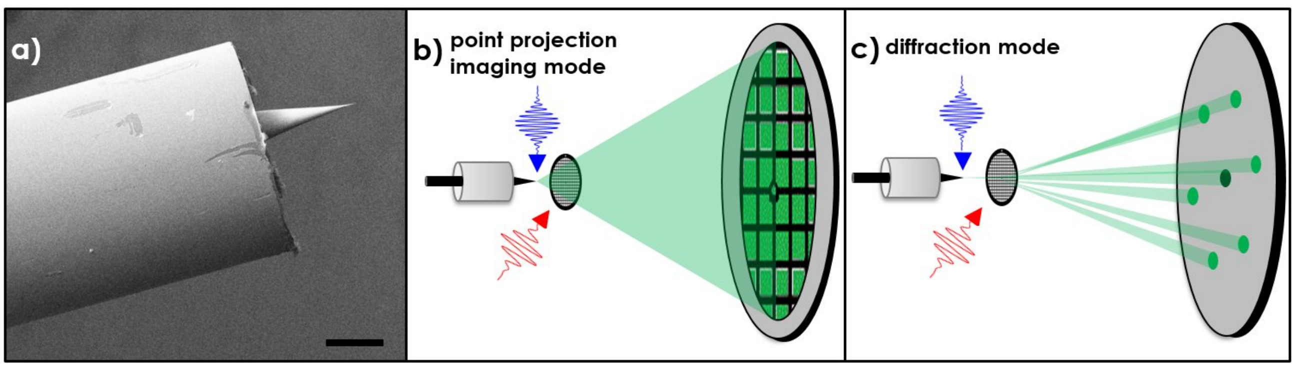

Figure 2. a) SEM image of an electrochemically etched tungsten tip mounted in a gold coated microlens. The tip protrudes from the lens to a distance approximately equal to the radius of the lens. The scale bar is 200 µm. Setup for low-energy electron b) point projection and c) diffraction imaging modes. The electrons emitted by the field emission tips are excited by the ultrashort laser pulse and then are accelerated towards a nanoscale sample placed a few micrometers away from the tip. The gold coated electrostatic lens is used to collimate the electron beam allowing both diffraction and point projection imaging in the same apparatus.

OPTICAL COMPRESSION OF ELECTRONS

For experiments utilizing UEM's one of the limiting factors is the electron pulse duration. Electron sources have an intrinsic energy spread (~1 eV) which leads to a distribution in electron velocities and is inherent to all pulsed electron sources. This spread in electron velocities causes the pulse to spread, or undergo dispersion as it propagates resulting in longer duration pulses. One technique to deal with this dispersion is to place the electron source and specimen extremely close together, where the electron pulse does not have time to disperse before it interacts with a specimen[13]. A second option is to 'correct' the dispersion by recompressing the electron pulse. This can be accomplished by influencing the electron pulse with a transient potential that speeds up the trailing slow electrons and slows down the fast leading electrons[17]. As this pulse then propagates it recompresses at a later position, which is dependent upon the shape and strength of the transient potential. To create the transient potentials for electron pulse compression RF[18] and

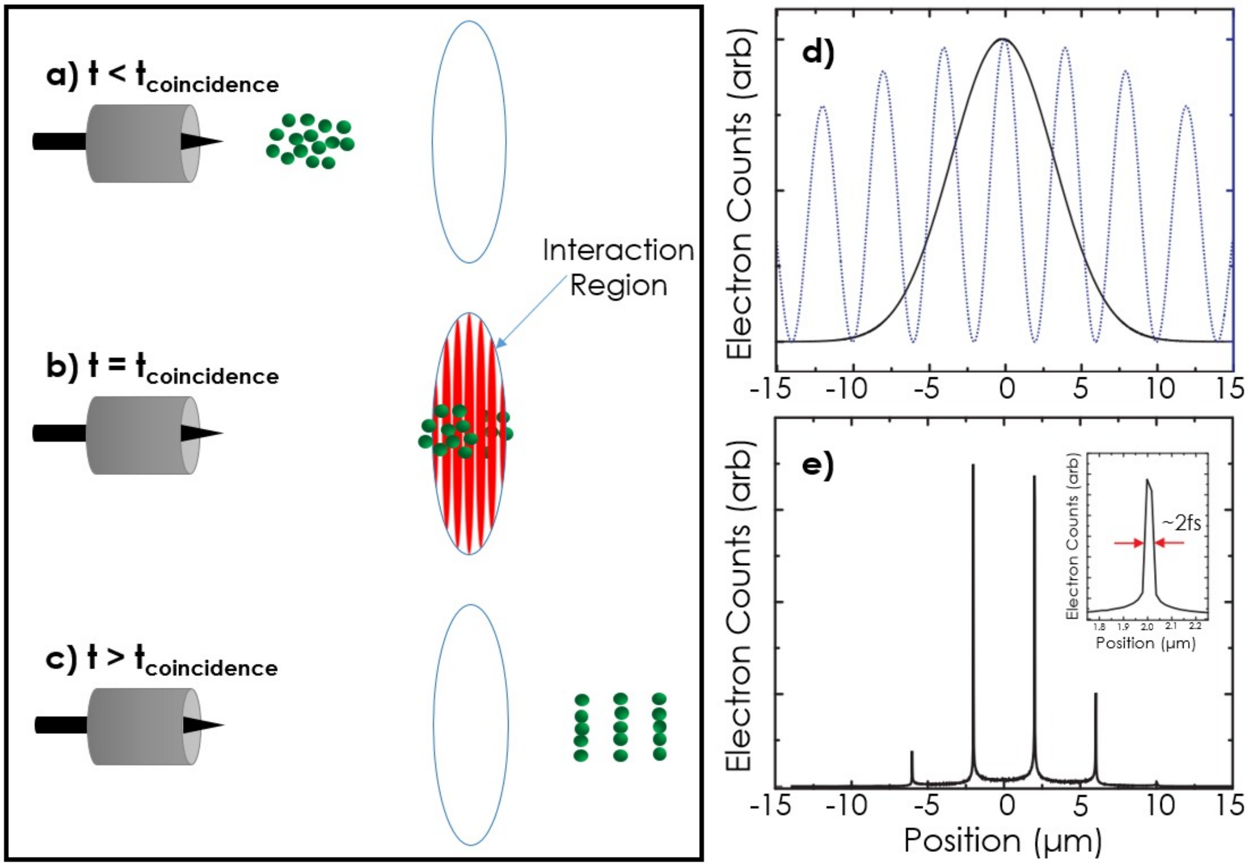

Figure 3. a) A dispersed electron packet of variable velocities is emitted from the emission tip-lens combination and directed towards the interaction region and sample. b) The electron packet reaches and passes through the interaction region, during which time it is subjected to the standing wave. This compresses the packet. c) The electron packet moves away from the interaction region, and towards the sample. d) An electron pulse is overlapped with a standing wave (Intensity) formed by the pulsed laser. e) After a certain time (2.8 ps), the electrons from d) have reached maximum compression, represented by the narrow peaks. The inset in e) shows a magnified view of one of the peaks.

Terahertz fields[19] have been demonstrated. Both techniques can deliver electron pulses to a specimen with durations of ~100 fs. An alternative method using Visible-NIR light in the form of a standing wave has been proposed to compress electron pulses from picoseconds to the few fs to attosecond regime[20]. The standing wave optical method was investigated theoretically in the context of higher energy electrons (10-100keV) with a standing wave constructed with two different frequencies of light that co-propagates with the electron pulse[20]. After interacting with standing wave through the ponderomotive force, the electrons were shown to compress into a train of pulses of ~20 attosecond durations each[20].

Here, we show that this compression technique can also be applied to lower energy electrons, ~1 keV, utilizing a standing wave made with two laser beams that have the *same* frequency. Using two beams of the same frequency greatly reduces the experimental complexity and allows for demonstrated techniques of overlapping femtosecond laser pulses to be used[21,22].

In its simplest form an optical standing wave created by a laser has a ponderomotive potential that takes the form[23]:

$$\Phi_p = \frac{e^2 I}{2m\varepsilon_0 c\omega^2}\cos^2\left(\frac{2\pi\omega}{c}x\right). \quad (1)$$

where the intensity of the standing wave is I and the frequency of the light is ω and the corresponding ponderomotive force is:

$$F_p = -\frac{d}{dx}\Phi_p \,. \tag{2}$$

To numerically calculate the resulting compression due to the standing wave interaction we modeled an electron pulse that propagates from an electron source to a region where it interacts with the optical standing wave. During the propagation the pulse disperses due to the 1 eV initial energy spread. Then, after interacting with the optical standing wave, the electron trajectories were tracked to the point and time of maximum compression. In the calculation we used initial electron pulse duration of 100 fs, an electron energy of 1 keV and a distance of 15 mm from the source to the optical standing wave. The wavelength of laser used to create the optical standing wave was 8 μm and is present for 130 fs with a peak intensity of $5*10^{13}$ W/cm^2. Depending on the position of the electrons in the periodic potential they experience forces that can be positive or negative with respect to the propagation direction[18], see Fig. 3. For each individual potential well, the electrons that were slower experience a force in the positive direction, while the electrons in the lead experience a force in the negative direction. The result is that after interacting with the optical standing wave the electrons compress into a train of pulses of ~2 fs after propagating for 2.8 ps, Fig. 3. The compression time, along with the ultimate compressed pulse duration depends on the parameters of the standing wave, including the intensity and the shape of the potential.

In this compression scheme it is important that the electrons do not travel through multiple individual potential wells because the electrons would then experience both positive and negative forces resulting in no compression. For the parameters used in this calculation the electrons travel ~ 2 μm while the standing wave is present. This distance is less than the periodicity of the standing wave intensity, which is 4 μm and thus compression of the electron pulse still occurs. While 8 μm wavelength femtosecond laser pulses can be experimentally produced using an optical parametric amplifier, a simpler experimental realization could use 800 nm light and techniques that overlap femtosecond laser pulses to produce standing waves with arbitrary periodicities[21].

KAPITZA-DIRAC EFFECT WITH ORBITAL ANGULAR MOMENTUM

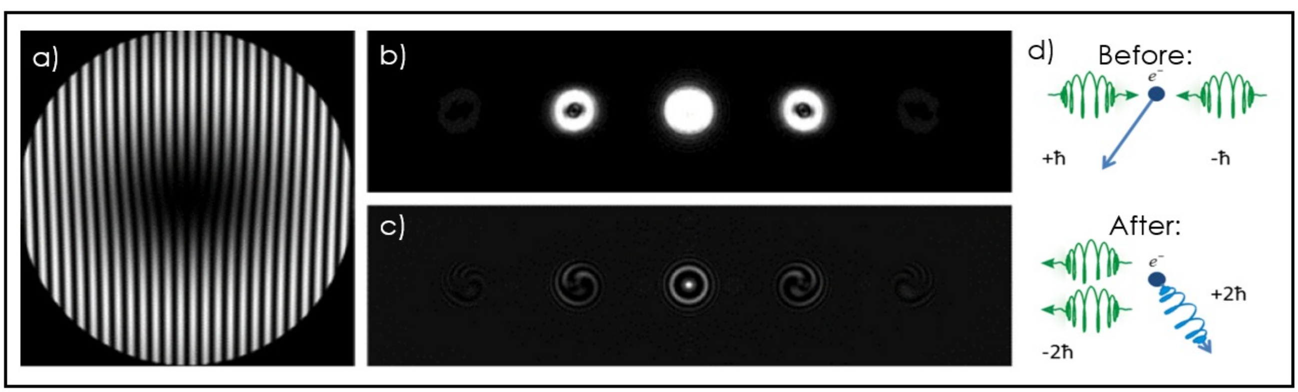

Figure 4. a-c) Example calculation results of electron beams carrying orbital angular momentum [13]. a) Optical intensity pattern from a standing wave created by overlapping two laser pulses (of l = -1 and l = +1, respectively). The circle is 10 microns in diameter. b) Electron diffraction pattern after interaction with the standing wave shown in a) created with optical vortex beams. c) The same electron diffraction pattern as b), after being interference with a plane electron wave, which is representative of the spiral phase. d) The two optical vortex beams – with opposite angular momentums – converge at the electron, transferring OAM to the electron, while still conserving OAM.

The Kapitza-Dirac effect was predicted in 1933 and proposed the use of standing waves to act as a grating for electrons, in a manner similar to a physical diffraction grating[24]. Nearly 70 years after first being predicted the effect was experimentally demonstrated[25]. More recently the KD effect has been proposed as a way to transfer orbital angular momentum from photons to electrons[12], see Fig. 4d. This is done by substituting the laser beams that create the standing wave with beams carrying OAM. This results in an optical phase grating for electrons with the appropriate holographic pattern that has a higher periodicity above its center than it does below, similar to the 'fork' grating material holograms that have already been shown to produce OAM electron beams[26]. The diffracted electron beams carry integer amounts of OAM, which varies with the diffracted order, Fig. 4b. The 'donut' pattern of the electron beams is indicative of beams carrying OAM, Fig. 4b, and if the electron beams are interfered with a plane electron wave the phase of different diffracted orders can be determined, Fig. 4c.

CONCLUSIONS AND FUTURE PERSPECTIVES

As shown above, the capability of low energy UEM's can be extended into the few femtosecond regime by using an all optical compression technique which will open up a variety of dynamical experiments that are out of reach using current UEM's. By using a single frequency of light to create the standing waves that are used for compression, the experimental complexity is reduced, while still delivering few femtosecond electron pulses. In addition, the same experimental apparatus and optical standing waves can be used to diffract electron beams using the KD effect. By using OAM optical beams to create the standing wave, the transfer of quantized amounts of OAM from photons to electrons can be studied. These low energy UEM advancements will allow dynamics to be studied with temporal resolutions approaching one fs, and open up experimental capabilities utilizing femtosecond electron pulses carrying OAM.

REFERENCES

[1] Zewail, A. H., "Four-Dimensional Electron Microscopy," Science 328(187), 187-193 (2010).

[2] Lobastov, V. A., Srinivasan, R. and Zewail, A. H., "Four-dimensional ultrafast electron microscopy," Proc. Nat. Academy Sci. 102(20), 7069-7073 (2005).

[3] Barwick, B., Flannigan, D. J. and Zewail, A. H., "Photon-induced near-field electron microscopy," Nature 462(17), 902-906 (2009).

[4] Park, S. T., Lin, M. and Zewail, A. H., "Photon-induced near-field electron microscopy (PINEM): theoretical and experimental," New J. Physics 12, 1-57 (2010).

[5] Flannigan, D. J., Barwick, B. and Zewail, A. H., "Biological imaging with 4D ultrafast electron microscopy," Proc. Nat. Academy Sci. 107(22), 9933-9937 (2010).

[6] Joy, D. C. and Maher, D. M., "Electron energy-loss spectroscopy," J. Physics E: Sci. Instruments 13(3), 260-270 (1980).

[7] García de Abajo, F. J. and Kociak, M., "Electron energy-gain spectroscopy," New J. Physics 10, 1-8 (2008).

[8] Barwick, B. and Zewail, A. H., "Photonics and Plasmonics in 4D Ultrafast Microscopy," ACS Photonics 2(10), 1491-1402 (2015).

[9] Piazza, L., Lummen, T. T. A., Quiñonez, E., Murooka, Y., Reed, B.W., Barwick, B. and Carbone, F., "Simultaneous observation of the quantization and the interference pattern of a plasmonic near-field," Nature Commun. 6(6407), 1-7 (2015).

[10] Feist, A., Echternkamp, K. E., Schauss, J., Yalunin, S. V., Schäfer, S., Ropers, C., "Quantum coherent optical phase modulation in an ultrafast transmission electron microscope," Nature 521(14), 200-212 (2015).

[11] Müller, M., Paarmann, A., Ernstorfer , R., "Femtosecond electrons probing currents and atomic structure in nanomaterials," Nature Communications 5, 2014.

[12] Handali, J., Shakya, P. and Barwick, B., "Creating electron vortex beams with light," Optics Express 23(4), 5236-5243 (2015).

[13] Quiñonez, E., Handali, J. and Barwick, B., "Femtosecond photoelectron point projection microscope," Rev. Sci. Instrum. 84, 103710 (2013).

[14] Gulde, M., Schweda, S., Storeck, G., Maiti, M., Yu, H. K., Wodtke, A. M., Schäfer, S. and Ropers, C., "Ultrafast low-energy electron diffraction in transmission resolves polymer/graphene superstructure dynamics," Science 345(6193), 200-204 (2014).

[15] Müller, A.-D., Müller, F., Hietschold, M., Demming, F., Jersch, J. and Dickmann, K., "Characterization of electrochemically etched tungsten tips for scanning tunneling microscopy," Rev. Sci. Instrum. 70(10), 3970-3972 (1999).

[16] Müller, M., Kravstov, V., Paarmann, A., Raschke, M.B., Ernstorfer , R., "A nanofocused plasmon-driven sub-10 femtosecond electron point source," ACS Photonics 3(4), 611-619 (2016).

[17] Hilbert, S. A., Uiterwaal, C., Barwick, B., Batelaan, H. and Zewail, A. H., "Temporal lenses for attosecond and femtosecond electron pulses," Proc. Nat. Academy Sci. 106(26), 10558-10563 (2009).

[18] Mancini, G. F., Mansart, B., Pagano, S., van der Geer, B., de Loos, M. and Carbone, F., "Design and implementation of a flexible beamline for fs electron diffraction experiments," 691, 113-122 (2012).
[19] Kealhofer, C., Schneider, W., Ehberger, D., Ryabov, A. Krauz, F. and Baum, P., "All-optical control and metrology of electron pulses," Science 352(6284), 429-433 (2016).
[20] Baum, P. and Zewail, A. H., "Breaking resolution limits in ultrafast electron diffraction and microscopy," Pro. Nat. Academy Sci. 103(44), 16105-16110 (2006).
[21] Maznev, A. A., Crimmins, T. F. and Nelson, K. A., "How to make femtosecond pulses overlap," Optics Letters 23(17), 1378-1380 (1998).
[22] Kozma, I. Z. and Hebling, J., "Comparative analysis of optical setups for excitation of dynamic gratings by ultrashort light pulses," Optics Commun. 199, 407-415 (2001).
[23] Batelaan, H., "Illuminating the Kapitza-Dirac effect with electron matter optics," Rev. Mod. Physics 79, 929-941 (2007).
[24] Kapitza, P. L. and Dirac, P. A. M., "The reflection of electrons from standing light waves," Math. Proc. Cambridge Phil. Soc. 02, 297-300 (1933).
[25] Freimund, D. L., Aflatooni, K. and Batelann, H., "Observation of the Kapitza-Dirac effect," Nature 413(6852), 142-143 (2001).
[26] McMorran, B. J., Agrawal, A., Anderson, I. M., Herzing, A. A., Lezec, J., McClelland, J. J. and Unguris, J., "Electron Vortex Beams with High Quanta of Orbital Angular Momentum," Science 331(6014), 192-195 (2011).