Chapter

Introductory Chapter: 4D Imaging

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1. Introduction

The study of ultrafast phenomena, including structural dynamics and molecular reactions, is of great interest for physics, chemistry, biology, and materials science. There are numerous examples of phase transitions in condensed materials and chemical reactions in free molecules proceeding on nanosecond, picosecond, and even femtosecond time scales. To study processes or reactions on such intricate scales, more sophisticated apparatus would be needed. It is well known that electron microscopy is a powerful imaging technique and is applied to a wide research field. The progress of electron microscopy has shown that three-dimensional (3D) material structures can be observed with an atomic spatial resolution. However, the conventional electron microscopy does not allow studying ultrafast processes because of the limitation of the speed of video camera.

The study of ultrafast structural dynamics or molecular reactions requires the use of probes ensuring not only high spatial but also high temporal resolutions. For this purpose, the new development of ultrafast electron microscopy (UEM), by combining temporal resolution into conventional electron microscopy, has been begun in the world. UEM uses a short pulsed electron beam replacing the continuous electron beam in the conventional electron microscopy to image the atomic motion by time-resolved recording in real time. By introducing temporal resolution into 3D electron microscopy, UEM allows us to observe the four fundamental dimension structures of matter: three spatial and one temporal, which is called 4D imaging.

Recent developments in UEM have shown that spatial and temporal information of matter can be obtained simultaneously on very small and fast scales. The first UEM was proposed to observe fast processes using a modified 120-keV electron microscope by Ahmed H. Zewail, Nobel Prize winner in Chemistry 1999, in the California Institute of Technology [1, 2]. He and his colleagues succeeded to observe the laser-photon-induced picosecond structural phase transition in vanadium dioxide film using a stroboscopic method with "single" electron pulses [3]. Later, a hybrid 200-keV apparatus was developed. A spatial-temporal resolution of 3.4 Å and 250 fs has been achieved. Recently, there are many research activities focused on improving the electron source and electron optics inside the microscope to achieve better temporal and spatial resolutions [4–9]. However, in the current UEM, the samples must be pumped 10^7 times or more by the laser. The process being studied must be perfectly reversible. To study the irreversible processes, it is necessary to record images with a larger number of electrons per pulse possible.

In this chapter, we introduce a novel UEM method with relativistic-energy electron pulses. In this relativistic UEM, an advanced radio-frequency (rf) acceleration technology is used to generate relativistic femtosecond electron pulses containing a large number of electrons in pulse and to achieve single-shot femtosecond imaging for the study of ultrafast irreversible structural processes.

2. UEM with relativistic femtosecond electron pulses

The relativistic UEM [10–14] is constructed with three principal components: a rf acceleration-based electron gun, a condenser system, and an imaging system. **Figure 1** shows a photo of the relativistic UEM, which is 3.5 m in height and 0.8 m in diameter. The rf electron gun is driven by a high power of rf to generate a high-peak rf electric field of 100 MV/m, which is 10 times higher than that of direct current gun in the conventional electron microscopy. The electrons emitted from photocathode are then quickly accelerated by the rf electric field into the relativistic energy region to reduce the effect of space charge, yielding ultrashort pulses containing a large number of electrons in pulse. The details of the rf electron gun and the generation of femsecond electron pulses are described in Chapter 2 [15].

Next, the electrons pass through a series of condenser lenses, which use magnetic field to precisely control the intensity of the beam, and its illumination angle on the sample. A relativistic-energy electron imaging system, including an objective lens, an intermediate lens and two projector lenses, is used to magnify the microscopic images. Finally, the images are recorded with a viewing screen (scintillator)

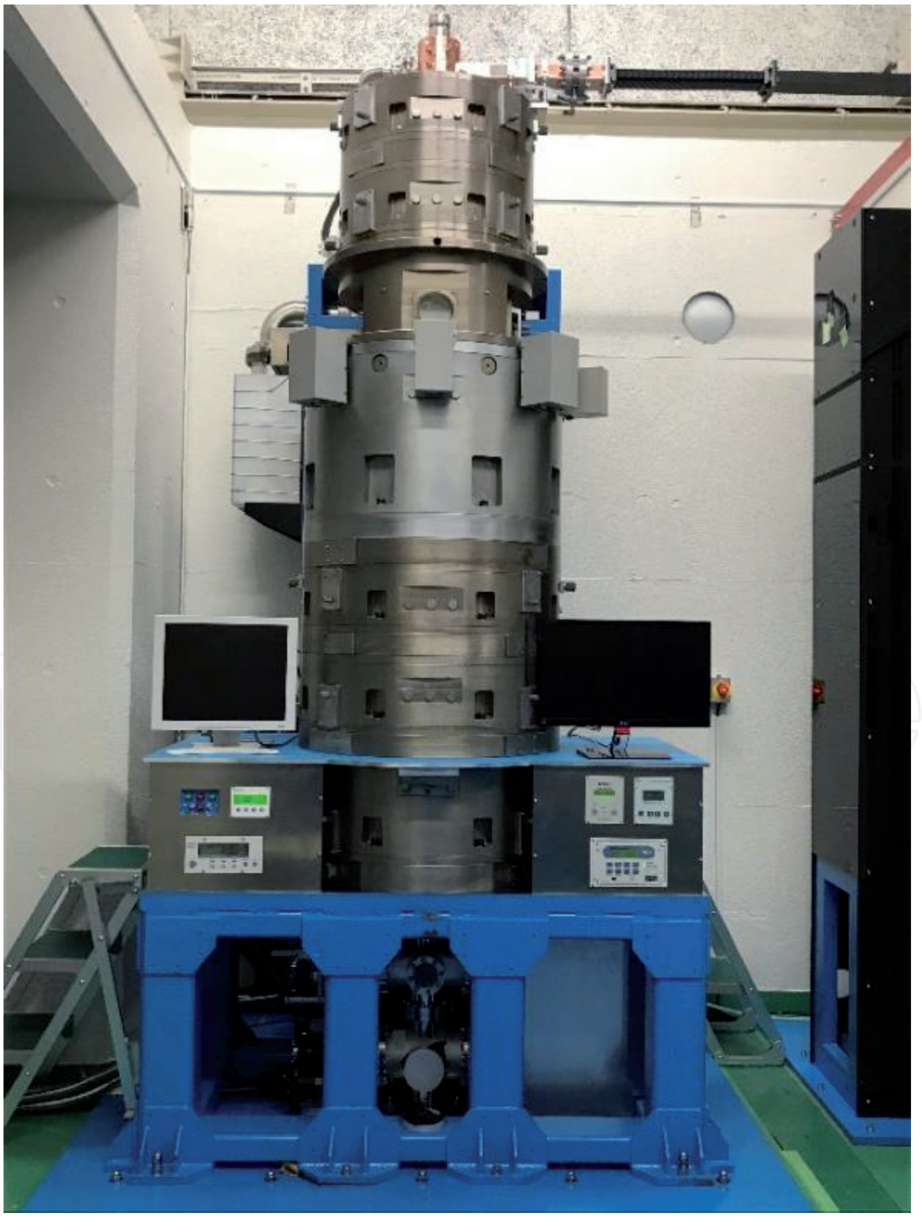

Figure 1.
Photo of UEM with relativistic-energy femtosecond electron pulses constructed at Osaka University [13, 14].

via a charge-coupled device camera [16]. The relativistic UEM is also an ultra-high voltage transimission electron microscopy (TEM). It exhibits many significant advantages over nonrelativistic-energy UEMs:

1. High temporal resolution of 100 fs or less is achievable, because the ultrashort electron pulses of <100 fs can be produced by the rf gun. The transit-time broadening due to the relative energy spread is reduced using the relativistic-energy electrons.

2. The relativistic UEM enables to observe the irreversible processes in materials by single-shot imaging with high-intensity femtosecond electron pulses.

3. The high-energy electrons significantly increase the extinction distance of elastic scattering. Our previous studies [17, 18] indicate that the kinematic theory with the assumption of single elastic scattering events can be applied in the relativistic UEM. This enables one to easily explain structural dynamics from the experimental results.

4. A thick sample can be used for measurement, thus obviating the requirement to prepare suitable thin samples.

5. The relativistic UEM is suitable for in situ observations. A large pole piece of the objective lens can be applied for installing various specimens.

The structural dynamics is observed in UEM with a pump-and-probe method, as shown in **Figure 2**. The femtosecond laser pulse is used as a pump pulse to excite the sample, while the electron pulse is used to record the time evolution of image of the structure by changing the time interval between the electron pulse and the laser pump pulse. The time resolution of UEM is determined mainly by the pulse duration s of the probe electrons and the pump laser. A high temporal resolution can be achieved with the ultrashort electron pulse and the ultrashort laser pump pulse. In this UEM, many demonstrations have been carried out and summarize as the followings:

1. A 100-fs-long pulsed beam containing 10^6–10^7 electrons at an energy of 3 MeV has been generated using the rf gun [10–12].

2. In the imaging experiments using these femtosecond pulses, we successfully observed contrast TEM images of 200-nm-diameter gold nanoparticles and other materials. At a low-magnification observation, single-shot imaging with the 3 MeV fs electron pulse is achievable [11, 12].

3. In the electron diffraction measurement, we successfully detected high-contrast electron diffraction images of single crystalline, polycrystalline, and amorphous materials. An excellent spatial resolution of diffraction images was obtained as 0.027 $Å^{-1}$ [19, 20].

4. In the pump-and-probe experiments using the relativistic femtosecond pulses, a laser-induced ultrafast melting dynamics in crystalline gold [17, 18] and a laser-excited ultrafast electronically driven phase transition in single-crystalline silicon [19, 20] were observed. The best temporal resolution of 100 fs has been achieved [20].

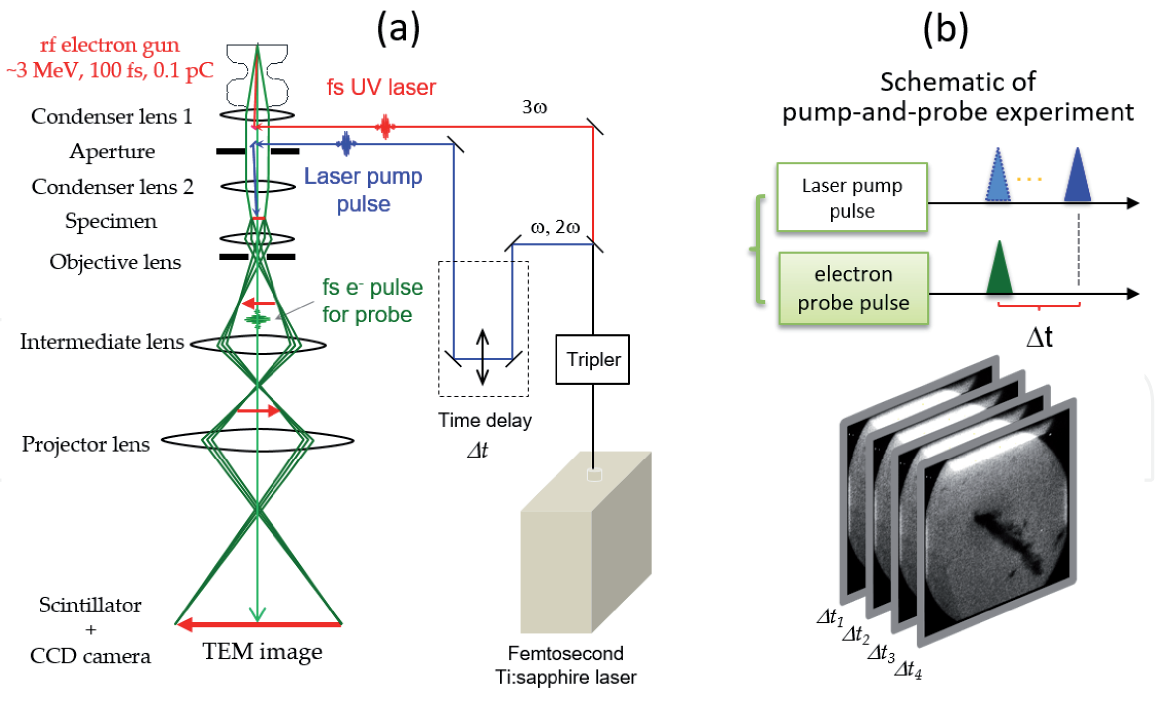

Figure 2.
(a) General schematic of UEM using relativistic femtosecond electron pulse and (b) pump-and-probe method for the observation of structural dynamics [13].

The details of the above experiments have been reported in the related references. The results demonstrate the advantages of relativistic UEM, including access to high-order Bragg reflections, single-shot imaging with the relativistic femtosecond electron pulse, and the feasibility of time-resolved imaging to study ultrafast structural dynamics.

3. Conclusion

Ultrafast electron microscopy with relativistic femtosecond electron pulses is a very promising 4D imaging technique for scientists wishing to study ultrafast structural dynamics in materials. It is an unprecedented innovative technology that enables femtosecond atomic-scale imaging using single-shot measurement and paves the way for the study of irreversible processes in physics, chemistry, biology, and materials science.

The relativistic UEM is also a very compact, ultra-high voltage electron microscopy. It can be used in a variety of settings such as general research institutions and laboratories. Furthermore, by providing a femtosecond temporal resolution, the relativistic UEM will constitute the next generation of electron microscopes. It will allow the study of structural dynamics to be broken into unprecedented timeframes, further encouraging the discovery of new knowledge.

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