Ultrafast Electron Diffraction Technology for Watching Dancing Atoms

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1. Introduction

If we can directly observe atomic or molecular movements in a material of which phenomena have not yet been elucidated, we can obtain valuable clues to understand its secrets. We have already investigated atomic and molecular structures and their changes using X-ray or high-energy electron beam which have smaller wavelength than the size of the atom or molecule. Recently, techniques for directly observing structural changes of atoms and molecules with a temporal accuracy of less than 100 fs have been established. The most advanced one is X-ray free-electron laser (X-FEL), which is the 4th-generation synchrotron radiation [1,2]. On the other hand, ultrafast electron diffraction (UED) is a technology that can achieve similar performance with a much smaller and inexpensive device [3-6]. Both techniques can directly measure structural changes of atoms or molecules with time-resolved diffraction.

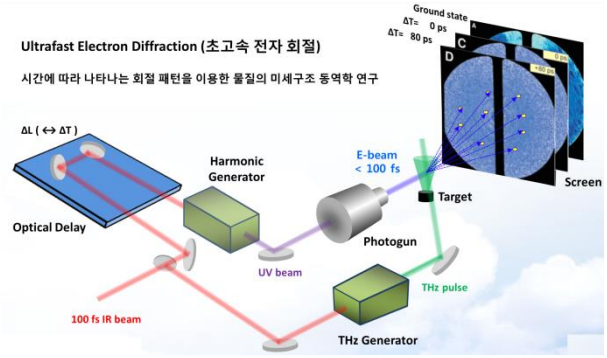

Fig. 1. Schematics explaining the operation principle of UED. Here, terahertz pulses generated by infrared 100-fs laser pulses are pumping source to excite a sample in the target and the electron bunches which are also generated by the same laser pulses are probe source to get diffraction images which contain the changes of atomic and molecular structure of the sample.

The important difference between both technologies is that the coherence length of the high-energy electron beam is just several to several tens of nm, which is much shorter than that of the X-ray. That is, the UED is not suitable for observing molecules larger than the coherence length, such as proteins. However, UED can be used for various applications in the field of physics, chemistry, and material science, except for bio science. It is very difficult for many users to use the X-FEL because it is not technically and economically easy to increase the number of beam lines, unlike the 3rd-generation synchrotron radiation. Therefore, it is necessary and also very important that UED has a similar or better performance compared to the X-FEL for the available target area.

The basic performance of camera is the spatial and temporal resolution. In a broad sense, X-FEL and UED are also cameras that observe atoms or molecules. The most important factor in determining the spatial resolution of a camera is wavelength of the wave used for observation. A few MeV-energy electrons have a wavelength shorter than pm, which is much shorter than that of X-ray. So, in terms of spatial resolution, UED is superior in principle.

Efforts to observe atomic and molecular movements in a shorter time have been actively conducted. Recently, US X-FEL, Linear Coherent Light Source (LCLS), reported that the structural change of the material has been observed with a time accuracy of 30 fs [7]. Some UEDs also exhibit temporal performance of about 100 fs or less and efforts are underway to further improve it.

Our efforts are focused on developing an UED having a better temporal resolution [8,9]. In conventional UED, a sample is usually placed as close as possible to electron gun in order to minimize beam expansion caused by space charge and energy spread of the beam, and the trajectory of the electron beam is straight. We employed two 90-degree-bending structures for the UED beam lines that satisfy achromatic and isochronous condition to compress electron bunch, and to reduce the arriving-time fluctuation relative to pumping laser pulse. With the introduction of this new structure, electron-bunch duration can be shortened to 30 fs, and temporal fluctuation between the electron bunch and pumping laser pulse can be reduced to 10 fs.

In this presentation, we will show numerical and experimental results of the KAERI UED that we have developed to improve temporal performance. We show the measured electron beam parameters of the UED beam line and the spatial resolution of our UED from the electron diffraction images of reference samples.

Finally, we will conclude with a brief description of the application researches we are underway.

2. UED with 90-degree-bends

UEDs can be classified into the use of electrostatic photogun and the use of RF photogun. In the case of electrostatic-photogun UED, the energy of the electron beam is 100 keV or less. Although this device is simple, the gradient of the electrostatic acceleration is low, which limits to shortening the electron-bunch duration and increasing the number of electrons in the bunch. Also, since the electron velocity is greatly different from the light velocity of pumping source, the time accuracy is deteriorated when the volume of the sample is large as in the gas sample. Because of these problems, UEDs using RF photoguns with large acceleration gradients have been developed. Generally, the electron energy of the RF-photogun UED is approximately 3 MeV. It has the advantage of producing a bunch duration of 100 fs while accelerating about one million electrons that can perform single-shot diffraction measurement. The RF-photogun UEDs developed so far have a structure in which the photogun and a sample are arranged in a straight line. No structure is used to further reduce the bunch duration of the electron beam generated from the photogun.

The total temporal resolution of UED experiment, τ, can be expressed as,

$$\tau = \sqrt{\tau_{pump}^2 + \tau_{electron}^2 + \tau_{jitter}^2 + \tau_{vm}^2},$$

where τ_{pump} and $\tau_{electron}$ are the temporal durations of optical pumping pulse and electron bunch, τ_{jitter} is the jitter between them at the sample, and τ_{vm} is the time interval between the two pulses caused by velocity difference across the sample. In most cases, this value, τ_{vm} , is negligible for RF-photogun UEDs. As a pumping light source, Ti: sapphire regenerative amplifier is mainly used, and the pulse duration is usually 100 fs. We are also using the same specification of the laser, and we plan to introduce a 30-fs laser to further improve the overall time resolution. Overall, the bunch duration of the electron beam and the temporal fluctuation, jitter, between the electron beam and the laser pulse contribute to the overall time resolution.

We focused on the development of technologies to reduce the electron beam bunch width and the laser-electron beam, ie the pump-probe time swing, which is the most technical difficulty to achieve with time resolution.

We focused on the development of a new UED which can reduce the bunch duration of the electron beam and the timing jitter between laser and electron beams, pump-probe timing jitter, which is the most technical difficulty to achieve with time resolution. The new structure we proposed is a UED with two 90-degre-bending structures which is shown in the upper part of Fig. 1. The synchronization between the pumping laser oscillator and a master oscillator of the RF system for the RF photogun was successfully performed with the extremely low timing fluctuation of ~10 fs during 24-hour operation [10]. I will explain the detailed operation principle and characteristics of the UED during my presentation. The electron beam characteristics obtained from numerical analysis of the UED are shown in Table I.

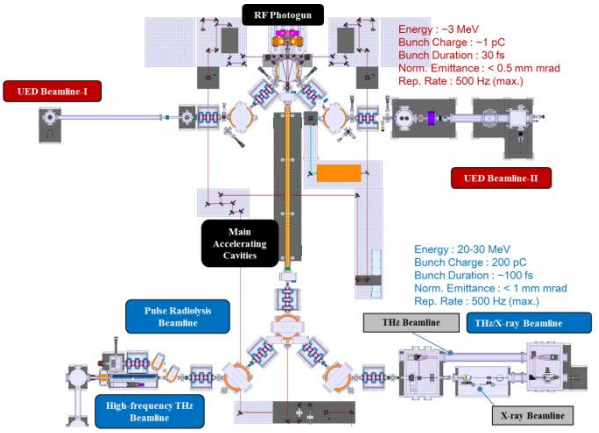

Fig. 1. Schematics of the KAERI Quantum-Beam-based Radiation Facility. The upper two beam lines are for UED experiments. UED beam line in the left part is for solid-state samples and the other beam line in the right part is for gas samples.

Fig. 2. Foreground of the KAERI Quantum-Beam-based Radiation Facility.

Table I: Simulated Electron Beam parameters of the KAERI UED

Beam parameters	Desired	Simulation	Units
Number of electrons	$> 10^6$	6.25×10^6	electrons
Beam kinetic energy	~ 3	3	MeV
Energy spread (rms)	< 0.1	0.17	%
Normalized emittance	< 0.3	0.29	mm mrad
Coherence length	> 2	1.8	nm
Bunch length (rms)	< 100	32	fs
Timing jitter	< 30	12	fs

3. UED Characteristics & Applications

The measured electron-beam parameters of the UED are well agreed with those in the Table I. Especially we could obtain high-charge (~3 pC) electron bunches having a relatively low emittance of approximately 0.4 mm·mrad. Measuring electron bunch duration and jitter between laser-electron pulses with an accuracy of ~10 fs is very challenging. We are conducting experiments to measure the electron bunch duration and jitter with THz spectral decoding by using coherent transition radiation from the electron bunches and pumping infrared laser pulses.

As shown in Figure 3(a) and 3(b), we recorded the diffraction patterns of single crystal gold with a single-shot measurement and a poly-crystal gold sample with 240 shots, respectively. We are conducting three independent application experiments with superconducting and strongly-correlated materials and gas samples for ultrafast molecular dynamics.

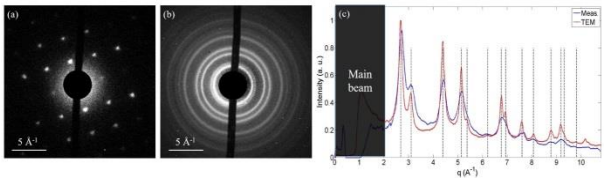

Fig. 3. Diffraction patterns of (a) single-crystal gold and (b) poly-crystal gold recorded in the UED beam line.

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