

Research on the ultrafast laser in microscopy

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Abstract: Ultrafast laser technology has been making a huge difference in different areas, such as material processing, laser surgery, and military usage. In parallel with the development of ultrafast lasers, microscopy also greatly promotes modern science, even though there are still some shortcomings, and one of them is that it is hard to obtain information about dynamic samples. However, this shortcoming can be solved by using ultrafast laser technology. This review will introduce the concept of ultrafast laser technology and electron microscopy, while several limitations of conventional electron microscopy will be mentioned. This paper also introduces two advanced technology in ultrafast microscopy. Nowadays, by taking advantage of ultrafast laser technology, the dynamic sample can be observed with highly improved temporal resolutions, which are basically restricted by the duration of pulses, but the ultrafast laser technology makes the duration even reach the range of attoseconds. However, there are some limitations, such as aberration, still hamper the development of high spatial resolution in microscopy. It is hoped that the ultrafast microscopy will break through the bottleneck of traditional microscopy in the future.

Keywords: Ultrafast Laser, Electron Microscopy, SEM, TEM, Time Resolution.

1. Introduction

The technique of microscopy has been developing rapidly over centuries, but for pure optical microscope, and it is not possible to achieve a very high magnification because of the existence of diffraction, the light will scatter when the wavelength is close to the obstacle, and the closer the wavelength, the more obvious the phenomenon is, the blurrier the image is. Basically, the maximum magnification of light microscope is about $\times 1500$, which means it can let the image becomes 1500 times larger than the actual sample. In order to obtain a larger magnification factor, people invented electron microscope. In addition, there are also several main disadvantages for electron microscopy, such as the inability to analyze an live specimen.

Over the past few decades, the object we want to observe tend to be more and more dynamic. However, we cannot easily observe the phenomenon with previous standard electron microscope. Hence, scientist developed ultrafast electron microscopy by taking advantage of the high temporal resolution of ultrafast laser to trigger ultrafast electron pulses, and this new technique opens the door for people to detect the fast dynamic.

Ultrafast laser is an unique light with some special characteristics compared to ordinary light, such as a much shorter wavelength, much higher energy, monochromaticity, etc.

This review will first briefly introduce the history and characteristics of ultrafast laser and electron microscopy, particularly scanning electron microscopy (SEM) and transmission electron microscopy (TEM). This paper will present the limitations of these two methods. Next, this paper will introduce the principle and fundamental progress of two techniques by combining the ultrafast laser with electron microscopy to achieve a more precise way to probe and observe samples. Then the research will discuss the limitations that still exist in the realm and the future development of ultrafast microscopy.

2. Ultrafast laser

The generated pulses range of ultrafast laser is in picosecond and femtosecond, and it has been developing for about half of a century and evolving from complicated laboratory system with a single function to a user- friendly, less costly and more reliable instruments.

The first helium-neon (He-Ne) laser to generate a continuous wave of light, the wavelength can reach at 1.15 μm was made by Ali Javan, William Bennett Jr. and Donald Herriott of Bell Labs in 1960. After six years, the first ultrafast pulses were generated by De Maria and co-workers by using a passively mode-locked Nd: glass laser, and this pulse width was considered to be just picoseconds long [1].

The main way to achieve ultrafast laser is mode- locking technology, which is a way of performing coherence between various phases of different modes in the resonator. There are also two types of mode-locking: passive mode-locking and active mode-locking; the former can make a much shorter pulse by plugging saturated absorbers. Since Kerr lens mode-locking (a saturable absorber mechanism) was invented, the duration of every pulse can be generated from a picosecond to a femtosecond. Because of the ultrashort pulse duration, this kind of laser has a high temporal resolution, which is an essential character to make it becomes very powerful in ultrafast microscopy. Fast temporal resolution enables us to capture or “freeze” the motion with high speed or frequency. Such as the motion of atoms in a fast vibrating molecule can be easily frozen; the progress of dissociation dynamics of molecules and more intricate chemical reaction dynamics have been measured, and it can even resolve the tiny motion of electrons in an individual atom, which constructs the basis of quantum physics and chemistry. Undoubtedly, these achievements revolutionized our ability to detect molecular matter and were therefore rightly granted Nobel Prizes in Chemistry to Ahmed Zewail in 1999 and Eric Betzig, Stefan W. Hell, and William E. Moerner in 2014.

Nowadays, attosecond (10⁻¹⁸ second) laser can be even generated based on the theory of the mutual effect between the electromagnetic field and matter in atomic scale, and this will also play a key role in ultrafast microscopy in the future [2].

3. Electron based imaging

Electron microscopy is an advanced methodology to research the extremely tiny structure, such as crystal structure, the composition of molecule or even the structure of atoms in a scale of nanometer. Based on De Broglie wave theory, the wavelength of any moving particle equals to the Planck's constant (6.63×10^{-34}) divided by the momentum of the particle. Therefore, we can use an electron beam as a substitute by using the wave-like characteristics to magnify an object's image because the momentum of moving electrons is much smaller than that of a photon, so the wavelength is also smaller than that of light; it can be up to 100,000 times shorter than visible light. Comparing with traditional optical microscope, the source of illumination of electron microscopy is a bunch of electron beam with high speed. In 1932, theoretical studies on electron optics were firstly making by Walter Glaser, and his work made a primary foundation for the future research [3].

3.1. Electron Microscope (EM)

“Without microscope, there is no modern science.” Nobel Laureate Dr. Alan Finkel recently said. So we can see how important a advanced microscope is, but in 1900, the conventional visible light microscope arrived at the limitation with only magnification of 500x to 1000x. Scientists took approximately 30 years to conquer the difficulty [3]. Hence, a much more powerful microscope, which can even magnify things by 1 million or even more than the original image that we can see, was invented.

There are two types of electron microscopes: transmission electron microscopes and scanning electron microscopes, and they are both widely used nowadays, but some limitations or disadvantages should be seen.

3.1.1. TEM (transmission EM). In 1931, since the fact that by transforming the electron beam through samples can achieve a better resolution than the ordinary light was noticed by two German scientists, Ernst Ruska and Max Knoll [4]. Transmission electron microscope (TEM) was firstly demonstrated based on the theory. Two years later, the TEM was developed with a higher resolution. In 1939, the first commercial TEM was developed [3].

The TEM electron microscope is consisted with several basic components. The electron gun to emit electron beam, and usually the gun was bonded with a high voltage source to accelerate the electrons, so the initial kinetic energy of the electron beam can reach to a high standard. The column, coupled with a series of lenses, provides a place for the electrons to get through. The vacuum system maintains the state of vacuum in the column, a computer to control those components automatically with a few central parameters. The working progress is straightforward, the electron beam getting through the vacuum column and the sample, and it is ended up projected on the florescent screen, then the image can be observed.

The TEM technology is used across a lot of places. We use it if we want to know the inner structure of the sample, such as cell or even protein.

3.1.2. SEM (scanning EM). Scanning electron microscope (SEM) was first built by Von Ardenne in 1937. He added scan coil to TEM and by using voltage of (23kv) to make the magnification and the resolution reach at (8000x), (50-100nm) respectively. Multiple features of Ardenne's laboratory instrument gradually turn to a standard for SEM system afterwards. Nevertheless, an explosion happened in Ardenne's lab, and no commercial equipment was made based on his SEM experimental results. In 1942, a new methodology of SEM for the observation of thick specimens was given by Zworykin, Hillier and Snyder. Oatley corporate with his student McMullan successfully create the electrostatic lens SEM under the voltage of (40 kV) for the electron gun in 1953. Smith, who supplied extra principles to McMullan's contribution in his research. He found that signal processing could grow in micrographs, and the amplification of nonlinear signals was introduced. Besides, he invented double deflection scanning to modify the scanning system. All of them built a solid foundation for further work. In the same year, O. Wells contrived an updated stereoscopic pair in order to check the third dimension in the micrographs. Everhart, T., and Thornley, R., who are other research students, developed the secondary detector in electron conversion to light. Their work not only increased the collected signals but also ameliorated the signal-to-noise ratio. In 1963, the first SEM V system with triple magnetic lenses commercially sold under the name of "Stereoscan" Cambridge Scientific Instruments Mark 1 was built by Pease, and it was significant in the marketplace until 1965. Since so many achievements have been developed by pioneers such as stable electron beam sources which can also emit more electrons than before and better resolution has gained.

Basically the SEM electron microscope is consisted with several main components which is quite similar with TEM.

The electron gun which is the source of electron beam with high energy. The column where provides a place for the electrons getting through two or more electromagnetic lens. The same vacuum system as TEM to keep vacuum. The extra components comparing with TEM is a deflection system which is mainly consisted by scan coils. The chamber where to place the sample. The computer system with a display screen in order to receive the information of the electrons scattered by the specimen to display the scanned image [5].

SEM relies on the electromagnetic lens to control the track of the electron beam and the resolution of the image given by the size of beam is controlled by the condenser.

As the electron beam reaches the specimen, the characteristics of those backscattered electrons actually depend on the interaction in between electrons and the sample. The detector can pick up those electron and output an image according to this.

Comparing with TEM, SEM can provide 3d images, while only 2d images can be obtained by TEM. Also, the specimen should be extremely thin because we need the electron beam transmit through the specimen, so sometimes the thickness is even below 30nm, but there is no such demand for SEM. Due to the special requirement of TEM, so the time for preparation will also be long, but there is almost no complex preparation for SEM.

Nowadays, SEMs are widely used in range of fields, such as identifying the detailed information of virus or bacteria, and we can also get the corresponding treatments to deal with various diseases caused by them.

3.2. *Limitations*

There are several main limitations that people have considered so far. Firstly, the electron will probably influence or even destroy the sample, especially the organic or biological one, because the electron beam is an actual particle beam, so it will exert more impact on the sample than the light. Second, the operation should be held in a vacuum space because the electron will be easily affected by other molecules in the air, so a live specimen will be hard to analysis. Next is the existence of spherical aberrations and chromatic aberrations which limits the resolution of conventional electron microscope, but series methods have been presented to compensate the aberration, such as by using high voltages or multipolar compensation approach [6]. The ultimate limitation of electron microscope is still diffraction, because the wavelength of electron beam cannot be infinite short, just as the case of light. However, if we combine ultrashort laser with electron microscopy, then we can detect smaller structures and get more clear and detailed images.

4. **Techniques of ultrafast microscopy**

In parallel with the rapidly development of ultrafast laser technology, the progress of electron microscopy has also been developing significantly. Therefore, scientists take advantage of ultrafast laser technology to develop ultrafast microscopy. The theory is that by combining the ultrafast laser with high temporal resolution with an electron wave with high spatial resolution, this is called ultrafast transmission electron microscopy (UTEM).

Ultrafast microscopy has already been considered an effective way to image the dynamic characters of material with high spatial resolution on the atomic timescale.

4.1. *Laser excited metal tips*

Laser excited metal tips is a way to improve the temporal resolution of electron pulse probes as an electron point source in a nanometer scale. It is feasible to make a projection of electron images with ~10 fs electron pulses to place the sample in a few distance of μm [7]. The resolution in electron probing can be greatly accomplished by tunneling electrons in a scanning tunneling microscope. In the recent years, by a THz pulse-actuated single electron tunneling process through an STM tip-molecule-metal junction, the sensational movies of a molecule vibrational motion were successfully created by Cocker et al. on the sub-picosecond time scale [8]. Then the electron probe method was taken into a quantum realm by targeting molecule within an individual tunneling junction and coupling of electron source.

4.2. *Time-resolved photoemission electron microscopy*

Time-resolved photoemission electron microscopy (TR-PEEM) is another option to ultrafast microscopy. TR-PEEM subdues the difficulties of pulsed electron beam probing [9]. The roles of electron imaging and light pulses was divided in this approach. The key that the approach relies on is the excitation of nonlinear photoemission from the specimen by a pair of pump and probe pulses for the temporal resolution, as well as the imaging of the transverse distribution target to the spatial resolution. Therefore, the only limitation for temporal resolution is the duration of laser pulses, which can even

reach to the domain into attosecond theoretically. Aberration is the general limitation for the spatial resolution in electron microscopy. The typical range of the photoelectron energy is within 1 eV, and the limit of diffraction of the photoemission electron microscopy can be less than 1 nm, but this kind of resolution has not been made currently. The spatial resolution which defined by the edge contrast is about 50 nm for a typical column in common cases, but it can be less than 10 nm by using more advanced magnetic lens-based instruments for the correction of aberration, and this kind of technology is normally designed as an electron microscope with low-energy (LEEM), and it makes the independent structural characterization of the specimen be possible [10]. Inside the LEEM, the field emission electron gun source emits a bunch of electron beam with high coherence, and the electron beam will get into the sample directly the being decelerated to a few electron voltage. We can get information about the surface structure and electronic excitations from the reflected electron. The information about the sample's structure can be obtained by imaging the reflected electrons to a real or reciprocal plan, and we can also analyse the information by comparing the energy and the momentum. The transverse resolution of this instrument can reach to 2 nm by ameliorate the spherical and chromatic resolution [11]. Besides, the contrast of single atomic step is enabled in the standard direction by the phase difference between the electron waves from atomically flat terrace. The continuous electron source is used by LEEM, so the temporal resolution is mainly limited by the acquisition rate of the CCD camera. Thus, it is a good tool for obtaining the detailed information of the sample structure when proceeding the extensional growth or thermal processing, but ultrafast temporal resolution is not made in the implementations currently [12].

5. Conclusion

This article focused on the usage of ultrafast lasers in microscopy and first introduced the ultrafast laser and its related characteristics, as well as two types of electron microscopes, TEM and SEM. In terms of conventional electron microscopes, the temporal resolution and the difficulties of pulsed electron beam probing can be improved and ameliorated by ultrafast laser technology; two applications are listed at the end. Even though the later application hasn't been totally accomplished, it is still an advanced direction for scientists to promote ultrafast microscopy. With the development of ultrafast laser technology, the pulse width can be shorter and shorter, and this paper believes this will be greatly beneficial to the development of microscopy.

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