Design of an Ultra-fast Electron Diffraction System*

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Abstract The designed electron diffraction system consists of an ultra-fast electron gun, a sample chamber, a readout system, a power supply system, and a vacuum system, and it bears such unique characteristics as high energy, high temporal resolution, and high detection capacity. The photocathode is of a 35 nm Ag film deposited on an MgF_2 glass disk, and it is sensitive to ultraviolet light with wavelength of 266 nm. A magnetic lens is used to focus the electrons. Two pairs of electric deflection plates are used in the X and Y directions respectively to control the movement of the electrons, and one pair of them will act as a scanning plate while measuring the pulse width of electrons. The sample chamber is made of stainless steel, and in the middle of the chamber there is a specimen holder, capable of shifting in three dimensions and turning around its axis. The diffraction pattern recording system has a very high detecting efficiency, and even a single electron could be detected. A cascade MCP detector is used to ensure an electron gain reached to 10^4. The electron gun is in a vacuum system of 10^{-4} Pa. The whole gun is shielded by a μ-metal sheath. The designed temporal resolution of the ultra-fast electron diffraction system (UED) is about 358 fs.

Keywords Ultra-fast electron diffraction; MCP; Imaging; Magnetic focusing; Temporal resolution

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0 Introduction

There are many physical, chemical, and biological processes that happenning in an extreme short time scale such as in picosecond (ps) or femtosecond (fs) range, and these processes happen as a result of the interaction between the atoms and molecules of the matter. The atomic motion of the matter will ultimately determine the evolution of new phases in solid, the chemical dynamic processes, and the biological functioning processes. The observing and recording of these processes will provide many important information of material. Time resolved x-ray diffraction and electron diffraction are two characteristic systems developed for these purposes, and they are very useful in some areas such as ultra-fast diagnosis. Time resolved x-ray diffraction is restricted to the measurement of a single rocking curve because of its relatively weak probe beam intensity, and it provides only limited information of atomic motions within the unit cell. In contrast, time-resolved electron diffraction can determine the transient structures completely in atomic level under similar intensity, since the atomic elastic scattering cross section of electrons is about five orders of magnitude greater than that of x-ray's[1].

The first experimental system was reported in 1982[2], in which the melting process of metal Al was studied. Some other experiments about metal Al have been carried out since then[1,3-5] by using the time-resolved electron diffraction system (especially the femtosecond electron diffraction system (FED)[1,4-5]). Several kinds of FED systems have been developed, and they all play an important role in the study of the structure dynamics of the matter. The time-resolved reflecting electron diffraction has been used in the study of the surface structure characteristic or the surface structure phase transition of germanium[6-7], sodium chloride crystal[8], plumbum[9-11], etc. Easy preparation of specimen and low working energy are two main advantages of reflection typed electron diffraction systems. In the near years, the time resolved transmission electron diffraction plays a main role because that the incidence angle of electrons is not restricted, and the diffraction pattern is more orderly and clearly. Using the transmission UED system, nickel, platinum[12], and Au[13] were investigated using a ps electron gun. Zewail A. H.[14-16] team

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studied the breakage of carbon-iodine bonds with their FED system: the nonconcerted elimination reaction of 1,2-diiodotetra- fluoroethane ($C_2F_4I_2$) to give tetrafluoroethene and iodine, and the ring opening of 1,3-cyclohexadiene(CHD) to form 1,3,5-hexatriene.

The spatial and temporal resolutions of the ultrafast electron device is serval hundred to one ps[2-3,6-12,17], and in 2003 Cao J.[1] recorded a very clear electron diffraction imaging of metal Al using their 400fs system, and Bradley J. Siwick[4] achieved a 600 fs resolution.

The Institute of Physics of the Chinese Academy of Sciecnes began the study of the fast electron diffraction in 1999, when they observed a well directional high energy electron beam from the interaction between a high power femtosecond laser beam and a solid target[17]. They developed an electron diffraction method by utilizing of the directional fast electrons[18], and they got a conclusion that after the femtosecond electron beams were accelerated and focused, a femtosecond level temporal resolution and 0.01nm spatial resolution could be achieved, and could be used as an ultra-short electron probe.

1 The UED system

The UED system developed by the present research group consists of similar components to the former ones demonstrated by other labs. Fig. 1 is the block diagram of our ultra-fast electron diffraction system, which consists of the photocathode, the anode, the focusing system, the deflection system, the sample chamber, the readout system, the vacuum system, and the power supply system and so on.

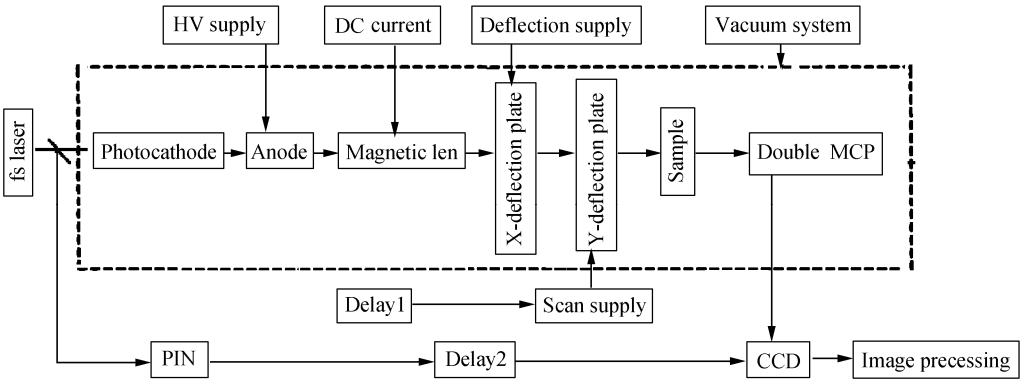

Fig. 1 The block diagram of the UED system

The inner part of the dashed line should be kept to high vacuum state, and this is achieved by a dry vacuum pump and a molecular pump. The pressure is measured by a vacuometer.

The main parts of the UED system are shown in Fig. 2, and the testing experiment is going along now. The photocathode is of a 35 nm Ag film plated on an MgF_2 disk. The accelerating voltage between the photocathode and extraction mesh is 50 kV, and the distance between the photocathode and extraction mesh is 5 mm, resulting in an electric field as high as 10 kV/mm. The high electric field is used to decrease the chromatic-spherical aberration of the focusing system, and to increase the temporal resolution of the UED system. There is a 100 μm pinhole on the anode to reshaped the electron beams and let the electron beams to go through. The working function of Ag (ϕ = 4.26 eV) is less than the energy of photoelectrons (ϕ = 4.67 eV) of the ultraviolet light (λ = 266 nm) in the present system. A frequency-tripled diode-pumped Ti-sapphire laser system or a frequency-quadruple Nd : YAG laser system can be used to offer a 266 nm laser light source to the UED system.

Fig. 2 The main parts of the UED system

Both electrostatic and magnetic electron lens can be used to focus the electron beams, and each of them has its own advantages. Magnetic lens can effectively focus high-energy electron beams without changing the kinetic energy of electrons. A magnetic lens is used in the present UED system because of the demand of the high temporal resolution.

The axial magnetic flux density (continuous

line is the calculated result with finite difference method, and the line with little circles is the measured result) is shown in Fig. 3. The Full

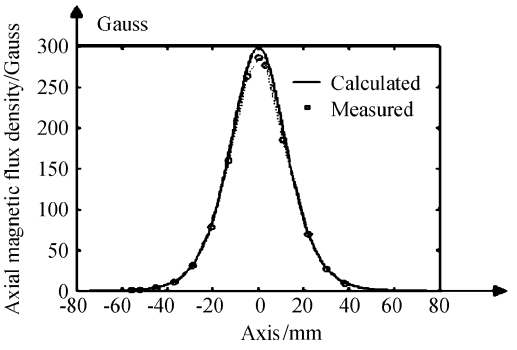

Fig. 3 The axial magnetic flux density

width at Half Maximum(FWHM) of the calculated result is about 34 mm, which is very near to the measured result 33 mm.

In the designed electron gun, there are two pairs of meander-shaped deflection plate (X direction, Y direction), which control the transverse displacement of electron beams in two dimensions. The function of the deflection system is just like a prism. That high deflection sensitivity, high deflection linearity, and small deflection dispersion is the performance which a well system should bears. One pair of the meander-typed deflection plate is used simultaneously as a scanner when measuring the pulse width of electron beam.

A cascade configuration of two MCPs is used to make the number of electrons multiplied. The diameter of the channel pore of the MCP is 10 μm. The two MCPs are chevron-type arranged, and the gain of the cascade MCPs is about 10^4, see Fig. 4. The voltage supply to MCP 1 and MCP 2 is about 0.8～1kV respectively. There is a phosphor screen (ϕ26 mm) which is plated on a fiber-optic faceplate served as a vacuum interface. Even a single electron can be detected because of the high gain of the cascade MCP.

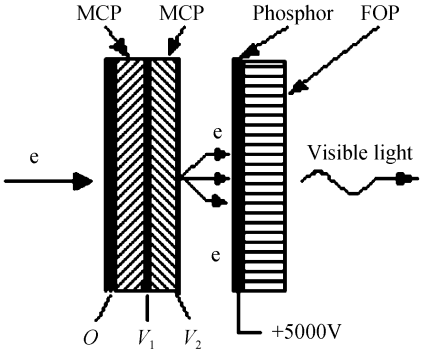

Fig. 4 Scheme of cascade MCP detector

The sample chamber is between the electron gun and the double MCP detector, which can be seen in Fig. 2. There are seven flanges on the chamber, one of which is used to emplace and to adjust the specimen holder, three flanges were used as watching windows, and the left three flanges linking-up to the vacuum chamber, the electron gun, and the cascade MCPs detector respectively. The specimen on the holder can be adjusted in three dimensions, it can be turned around, and the tilting angle can also be changed if necessary.

The electrical field and the magnetic flux distribution of the electron gun and magnetic lens are numerically calculated using finite-difference method (FDM). The emission area of the photocathode is assumed identical with that of the illuminating laser beam. That is to say, if the photocathode is illuminated by a laser beam with TEM_{00} mode, the initial position of emitting photoelectrons has a 2D Gaussian distribution.

We can see from the Law of Great Numbers that when sample number is high enough, the sampled distribution will be very close to the actual distribution. The initial energy distribution of electrons is believed to be follow the $\beta(n, m)$ distribution, while the values of the n and m are depending on many factors, such as the wavelength of the incidence laser, the material of the photocathode etc.. The initial position of the photoelectrons in our calculation is of Gaussian type, and its initial energy has a $\beta(1,4)$distribution with an 0.41 eV energy spreading. The azimuthal angle of the photoelectrons is evenly distributed within $0\sim2\pi$. The eleration angle distitution of the emitted electrons is of cosinoidal distitution in $0\sim\pi/2$ range. The trajectories of electrons were calculated by using the four order Runge-Kutta method.

In Fig. 5 the trajectories from the cathode to

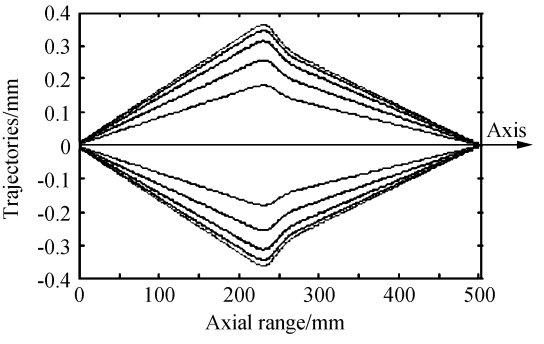

Fig. 5 The trajectories of eleven specified electrons

the sample plane (imaging plane) are shown. The initial angles of the electrons in the cathode plane are 0°, ±30°, ±45°, ±60°, ±72°, ±90° respectively. The axial range from $z=200$ to 250 mm is the location where the magnetic lens is placed, where the electrons change their moving direction. The electrons emitted near the axis are focused effectively, and part of the electrons off-axially emitted can also be focused. The FWHM of the beam spot size on the sample plane is about 15 μm.

Fig. 6 is the diagram of the calculated temporal distribution (i. e. the flight time distribution) of photoelectrons on the imaging plane, and the FWHM of this distribution is less than 300 fs. The dispersion of the time of flight of the electrons depend on the differences of emitting positions, emitting angles, and trajectories et al; the dispersion of the flight time is denoted as τ_{flight}.

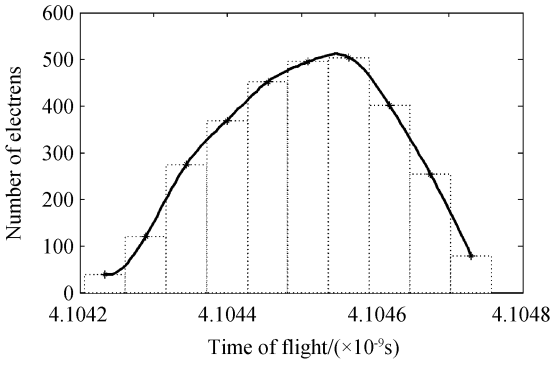

Fig. 6 Temporal distribution of the photo electrons

The overall temporal resolution τ_{total} of the UED system can be estimated as

$$\tau_{\text{total}}^2=\tau_{\text{laser}}^2+\tau_{\text{ied}}^2+\tau_{\text{flight}}^2 \tag{1}$$

where τ_{laser} is determined by the laser pulse width, and τ_{ied} is the the temporal dispersion of the photoelectrons in the cathode-anode area and it is determined by the initial energy distribution of electrons and the electric field between the photocathode and anode

$$\tau_{\text{ied}}=\frac{1}{\sqrt{2\eta}}\frac{\sqrt{\Delta\varepsilon}}{E} \tag{2}$$

and τ_{flight} denotes the dispersion of the time of flight in the anode - sample area. In Eq. (2) η is the charge-mass ratio of electrons, $\Delta\varepsilon$ denotes the initial energy spreading of the emitting electrons, and E denotes the electric field intensity between the photocathode and the anode. It is easy to find that smaller initial energy spreading and higher electric field intensity between the photocathode and the anode should result in a smaller τ_{ied}. For 10 kV/mm electric field intensity and 0.41 eV initial energy spreading, τ_{ied} is about 168 fs.

Therefore, for a 100 fs laser pulse and 300 fs temporal distribution, the overall temporal resolution τ_{total} is about 358 fs.

The actual temporal resolution may be larger than the calculated one because of the space charge effect, which will be checked up later.

2 The experimental result

The primary experiment was finished, and two parameter of the system were measured: the beam size and the deflection sensitivity. Three filaments was stickup to the surface of the screen, each with a diameter of 50 μm. The diameter of the beam size was got by comparing the scale of the filaments and that of the beam spot, which was about 83 μm, see Fig. 7. The electron beam size at the entrance of the first MCP should be measured, but we only measured the optical beam size after the phosphor screen and the optical-fiber faceplate, and the measured result may be a little larger than the actual electron beam size. The measured deflection sensitivity of the system is about 13 mm/kV, which is very near to the calculated result: 12.67 mm/kV.

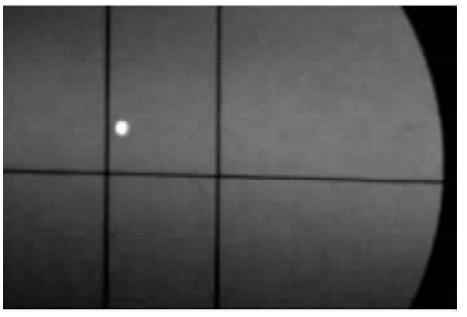

Fig. 7 The measurement of the beam size

3 Conclusion

An ultra-fast electron diffraction system is presented. The trajectories of 3000 electrons were calculated, and the calculated temporal resolution of the system is about 325 fs. All the electron optical characteristics of the system including the beam spot size, the spatial modulation transfer function, the spatial resolution, the temporal modulation transfer function, and the temporal resolution are calculated numerically, and a detailed dynamic experimental result of the system will be reported later.

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飞秒电子衍射系统的设计

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摘 要 研发的超快电子衍射系统由超快电子枪、样品室、超快读出系统、电源系统,以及真空系统等组成,该超快电子衍射系统具有较高的时间分辨能力和较强的探测能力.光电阴极是蒸镀于 $MgFB_2$ 窗上的 35 nm 的银膜,该阴极对 266 nm 的紫外光比较敏感,有较高的量子效率,又具有很好的化学稳定性.用短磁聚焦系统来实现对光电子的聚焦,有两对偏转板,其中的一对在测量时间脉宽时用作扫描板.用双 MCP 探测器来增强电子图像的强度,其增益在 10^4 以上,具有单电子探测能力.系统的总时间脉宽设计为 358 fs.

关键词 超快电子衍射;微通道板;磁聚焦;时间分辨

Wu Jianjun was born in 1970, Gansu . He earned M. Sc. at applied physics department from Northwest Polytechnical University, and now he is pursue for his master′s degree at Xi′an Institute of Optics and Precision Mechanics of CAS.