

Article

A Composite Velocity Map Imaging Spectrometer for Ions and 1 keV Electrons at the Shanghai Soft X-ray Free-Electron Laser

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Abstract: Velocity map imaging (VMI) spectrometry is widely used to measure the momentum distribution of charged particles with the kinetic energy of a few tens of electronVolts. With the progress of femtosecond laser and X-ray free-electron laser, it becomes increasingly important to extend the electron kinetic energy to 1 keV. Here, we report on a recently built composite VMI spectrometer at the Shanghai soft X-ray free-electron laser, which can measure ion images and high-energy electron images simultaneously. In the SIMION simulation, we extended the electron kinetic energy to 1 keV with a resolution <2% while measuring the ions with the kinetic energy of 20 eV. The experimental performance is tested by measuring Ar 2p photoelectron spectra at Shanghai Synchrotron Radiation Facility, and O⁺ kinetic energy spectrum from dissociative ionization of O₂ by 800 nm femtosecond laser. We reached a resolution of 1.5% at the electron kinetic energy of 500 eV. When the electron arm is set for 100 eV, a resolution of 4% is reached at the ion kinetic energy of 5.6 eV. This composite VMI spectrometer will support the experiment, such as X-ray multi-photon excitation/ionization, Auger electrons emission, attosecond streaking.

Keywords: velocity map imaging; Synchrotron Radiation; femtosecond laser; free-electron laser



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1. Introduction

The velocity map imaging (VMI) spectrometer is a powerful tool to obtain a high-resolution projected two-dimensional (2D) image of the three-dimensional (3D) momentum distribution of charged particles, which was widely used in atomic and molecular physics. The standard VMI spectrometer was originally designed by Eppink and Parker in 1997 [1], which consists of a repeller, an open extractor, and a flight tube. The charged particles are created at the crossing region where the light beam meets the molecular beam. Then they are driven towards a two-dimensional position-sensitive detector. With the well-designed electric field, the radius that the charged particle impact on the detector depends only on its initial radial momentum and is almost independent of the source position.

Recently, with the development of the femtosecond laser [2], high-order harmonic generation [3], and the free-electron lasers [4,5], scientists find it increasingly necessary to build a VMI spectrometer that is capable of imaging electrons with kinetic energies up to keV or even higher. The keV electrons can be created in the laser-induced electron diffraction experiment [2,6], the Auger electrons emission, or photoemission by multi-X-ray photons absorption [7], and the X-ray photoelectron diffraction (XPD) experiments [8,9].

Several other types of spectrometers can detect keV electrons but have their limitations: the angular resolved electron time-of-flight ARTOF spectrometer [10] and the hemispherical electron energy analyzer [11] are characterized by their small acceptance angle and

ultrahigh resolution, e.g., 0.5% of 4π and $\Delta E/E = 1000$ at 300 eV energy. Although the magnetic bottle spectrometer has large acceptance angles, it can barely provide angular distributions [12,13]. Because multi-photons are often involved, the electron's angular distribution can no longer be described using a beta parameter. Jens Viehhaus developed a “cookiebox” at beamline P04 at synchrotron PETRA III. With 16 electron time-of-flight spectrometers, “cookiebox” can measure simultaneously high-resolution electron energy spectra at 16 angles. Later, it is used to commission EXFEL [14]. A full-angle measurement is preferred to elucidate the reaction process. As a result, despite its relatively low energy resolution, a VMI spectrometer is often selected instead of an ARTOF spectrometer.

Usually, the VMI spectrometer is used to measure low kinetic energy electrons up to a few tens eV [15–19]. To increase the kinetic energy limit of the VMI spectrometer, several modifications of the standard Eppink-Parker setup have been conducted. Ablikim et al. [20] reported a double-sided coincidence VMI spectrometer is reached a resolution of 4% at the electron kinetic energy of 100 eV. Garcia et al. [21] add an einzel lens to zoom the images of energetic electrons and ions. Skruszewicz et al. [22] and Kling et al. [23] proposed a thick lens achieved a high energy acceptance up to 500 eV and 1000 eV, respectively. Schomas et al. [24] experimentally reached a resolution of <4% at the electron kinetic energy of 65 eV using a 42 mm detector, and simulations suggested a resolution of <5% at the electron kinetic energy of 1200 eV for the detector with a diameter of 78 mm. Li et al. [25] have developed a co-axial thick lens VMI for both high-energy (above 100 eV) and low-energy (tens of eV) electrons. Here, we report on a composite velocity map imaging (CpVMI) spectrometer at Shanghai soft X-ray free-electron laser, capable of mapping the electrons with kinetic energies up to 1 keV and low energies ions simultaneously.

2. Structure of Spectrometer

The CpVMI spectrometer is schematically illustrated in Figure 1. The ultrahigh vacuum system consists of five chambers: (i) source chamber; (ii) the 1st differential pumping chamber; (iii) the 2nd differential pumping chamber; (iv) main chamber; (v) jet dump chamber. They are pumped by a 2300 L/s turbomolecular pump TMP, two 400 L/s TMPs, two 700 L/s TMPs backed by 100 L/s TMP, in addition a 2000 L/s SAES non-evaporable getter pump is installed to the main chamber. All TMPs are from Pfeiffer. After 5 days' bake-out at 120 °C, the base pressures reach 5×10^{-9} , 2×10^{-9} , 1×10^{-10} , 5×10^{-11} and 7×10^{-11} mbar, respectively. In the main chamber, the electron-optical lens system was shielded by a double-layer μ -metal barrel to minimize the influence of the external magnetic fields on the trajectories of the electrons. A supersonic molecular beam is created by adiabatic expansion through a 100 μ m pulsed valve (ACPV2-100 from Amsterdam, The Netherlands) in the source chamber. It passes through a Beamdynamics skimmer with a ϕ 0.3 mm hole, two plates with ϕ 1 mm holes, then enters the main chamber, and is finally caught by a dump chamber. With supersonic molecular beam turned on, the working pressures are 4×10^{-4} , 4×10^{-7} , 1.5×10^{-10} , 5×10^{-11} and 9×10^{-11} mbar, respectively. In case there is a problem with the molecular beam, or the photon beam is too weak, an effusive molecular beam could be ejected from a stainless steel hollow needle with 100 μ m inner diameter. The molecular beam enters the interaction region of the CpVMI spectrometer, where it meets the X-rays beam or the femtosecond laser beam.

The electron-optical lens projects charged particles from the interaction region onto two VID275 detectors from Photek, each is composed of two multi-channel plates (MCPs) (75 mm effective diameter) and a P46 phosphor screen. Two cameras (Andor Zyla 4.2 PLUS) were used to record the images of electrons and ions. The single-shot TOF signals are decoupled from the phosphor screen, fed in a fast amplifier (FAMP8c from Roentdek, Germany), and finally recorded by an oscilloscope (Tektronix MSO56).

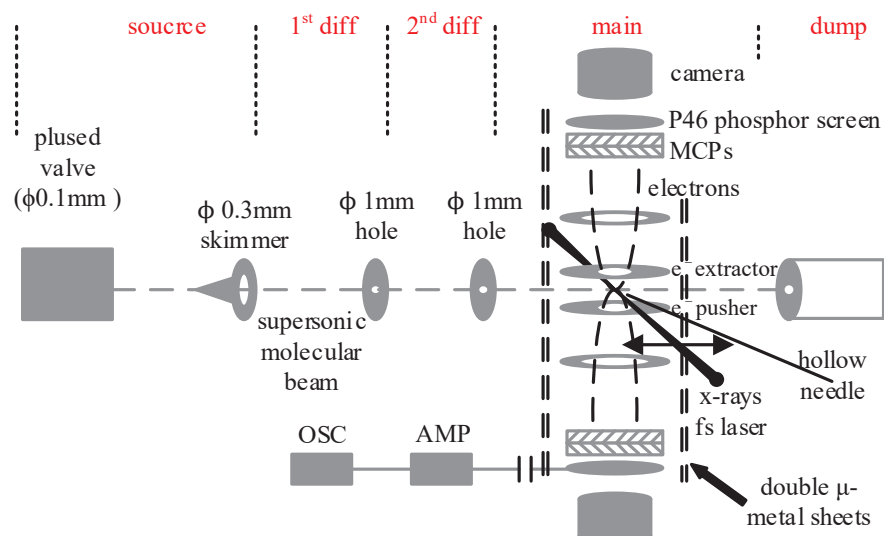


Figure 1. Schematic view of the CpVMI spectrometer. The gas sample can be prepared as either a supersonic molecular beam or an effusive molecular beam. Charged particles produced in the interaction region are projected onto the detector (stacked MCPs with a P46 phosphor screen), and the images are recorded by a camera. The ion TOF signals are recorded using an oscilloscope.

3. Lens Design

Figure 2a shows the dimensions of the electron-optical lens, which consists of 27 independently tunable electrodes. high voltages are supplied by Iseg EHS40200x power supplies. The interaction region is located between e^- pusher and e^- extractor. The right side is the electron arm, while the left side is the ion arm. The electrodes are made of aluminum plate with a thickness of either 5 mm or 1 mm. The gap between the electrodes is normally either 6 mm or 3 mm except the one between e^- pusher and e^- extractor, which is 18 mm so that the 800 nm femtosecond laser can be back-focused and the strayed X-ray photon would be less likely to see the electrodes.

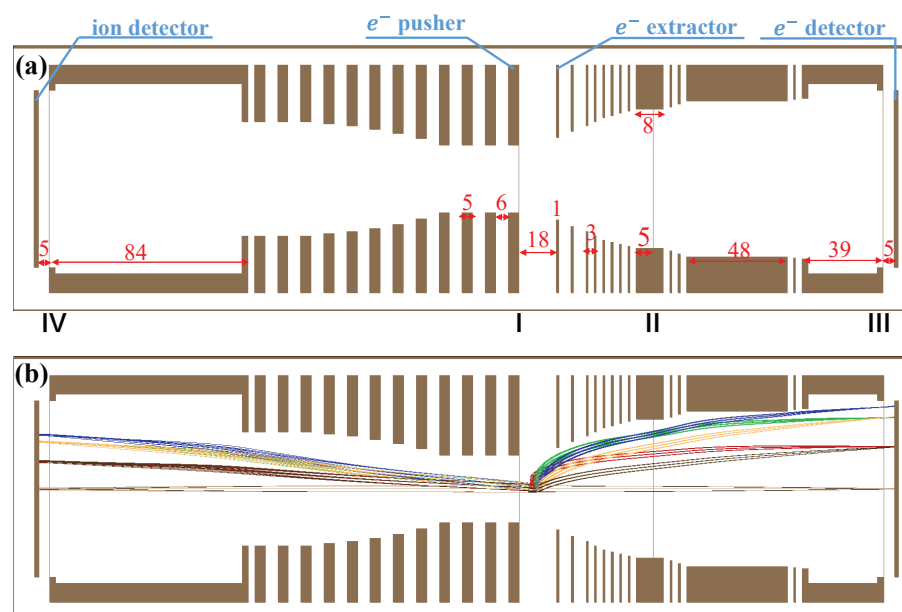


Figure 2. (a) The geometry of the electrostatic lens (length unit: mm). The electron arm and ion arm sit at the right and left sides, respectively. (b) SIMION simulated trajectories of the electrons with the kinetic energy of 1000 eV in the strong focusing mode and ions with the kinetic energy of 20 eV. The emission angles are 0° , 30° , 60° , 90° , 120° , 150° , and 180° with respect to the lens axis. The source region is defined as 9 particles emitted from a $1 \times 3 \text{ mm}^2$ rectangle.

In the lens there are four ultrafine stainless steel meshes with an 80% transparency. Mesh I and II separate the spectrometer into three parts, ion focusing region, reaction/1st electron focusing region, and 2nd electron focusing region. The electron arm can work in either strong or weak focusing mode. In the strong focusing mode, both the 1st and the 2nd regions are used for focusing. In the weak focusing mode, while the 1st region is set in a uniform electric field, only the 2nd region is used for focusing. As described before, there are two ways to introduce the gas into the main chamber: the supersonic molecular beam and the effusive molecular beam. Because the supersonic molecular beam does not affect the electric field, the electron arm can be set in the strong focusing mode. On the contrary, the effusive molecular beam will strongly distort the electric field if the interaction region is not set in a homogeneous field, so the electron arm needs to work in the weak focusing mode. Mesh III and IV are used to prevent the detector voltage from distorting the electric field in the lens. The distance between the reaction point and mesh II is half of that between mesh II and mesh III, as required by the Wiley-McLaren condition [26].

Figure 2b shows the simulated trajectories of electrons with a kinetic energy of 1000 eV in the strong focusing mode and ions with a kinetic energy of 20 eV using SIMION software, the source region is defined as 9 particles emitted from a $1 \times 3 \text{ mm}^2$ rectangle. The optimization is separated into two steps. The first step is to optimize the electron arm. We set the e^- pusher grounded, then find the voltage setting of the electron arm in either weak or strong focusing mode. The maximum voltage in simulation is 17 kV when the electron arm can detect 1000 eV electrons with a 4π acceptance angle. 17 kV can cause discharge easily, so we shifted all voltages negatively. For experiments that produce low kinetic energy electrons, we reduce all voltages proportionally.

The second step is to find the voltage setting of the ion arm. Since the ion arm and the electron arm share the interaction region, the setting of the ion arm will be affected by that of the electron arm. Therefore, we fixed the voltage setting of the electron arm according to different experimental conditions, then found the voltage setting of the ion arm. It needs to be emphasized that although the electron's kinetic energy can be as high as 1000 eV, the ion's kinetic energy is probably below 20 eV in the molecular dissociative ionization process. If the ion arm has the same geometry as the electron arm but with the opposite voltage, we can detect electrons and ions with the same kinetic energies. But then the ions image will be too small, and the corresponding resolution will be too low [9]. Therefore, we need a new geometry design for the ion arm; not only to focus the ions but also to enlarge the ion image.

The simulated resolution was determined by evaluating the mean radius R of the 9 particles on the detector surface at the emittance angle of 90° and the associated error ΔR . The relative energy resolution is then calculated as $\Delta E/E \cong 2\Delta R/R$ based on the $E \propto R^2$ [23]. Figure 3a shows the relative energy resolution for electron energies ranging from 0 to 1000 eV in the strong focusing mode. The best energy resolution is achieved at the highest kinetic energy. Figure 3b compared the relative energy resolution for ion energies ranging from 0 to 20 eV when the electron arm is set for different energies. The corresponding resolution becomes worse gradually when the electron arm is set for higher kinetic energy. Since we need to find a compromise between ion energy resolution and the zoom factor of ion image, as the electron arm is set for higher kinetic energy, the zoom factor of the ion image increases, then the ion energy resolution decreases. As the focus surface is not planar [22], it is difficult to focus well for all radii. When the electron arm is set for 1000, 600, and 300 eV, the best ion focus is achieved at the outmost radius. When the electron arm is set at 100 eV, it is easy to make the focus surface more planar. Therefore, the relative ion energy resolution depends less on the ion kinetic energy.

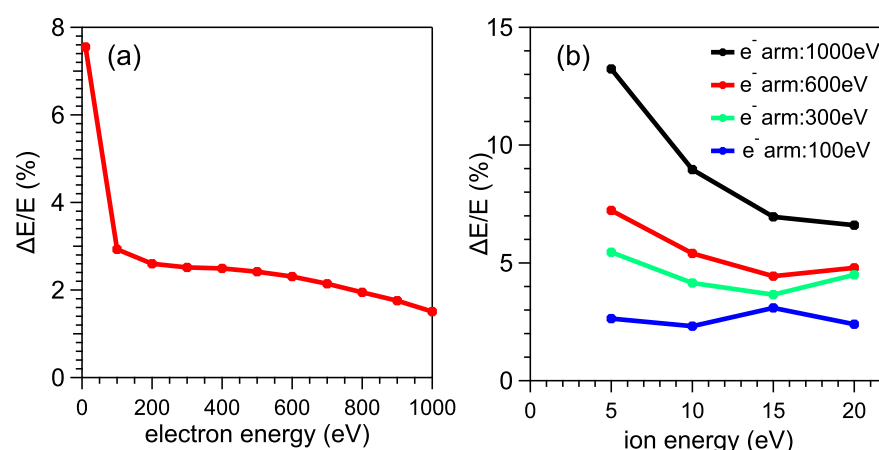


Figure 3. The simulated relative energy resolution $\Delta E/E$ of the CpVMI spectrometer with the electrons or ions emitted at 90° . (a) Relative kinetic energy resolution as a function of electron kinetic energy in the strong focusing mode. (b) Relative kinetic energy resolution as a function of ion kinetic energy with the electron arm is set for different kinetic energies.

4. Experimental Results

To test the CpVMI spectrometer, we have performed (1) photoionization of Ar 2p at the beamline BL02B at the Shanghai Synchrotron Radiation Facility (SSRF), and (2) dissociative ionization of O₂ by 800 nm femtosecond laser. They showed experimentally resolution for the electron arm and ion arm, respectively.

4.1. Electron Arm

To test the performance of the electron arm, we shipped the CpVMI spectrometer to the bending-magnet beamline BL02B [27] at the SSRF. This bending-magnet beamline delivers soft X-rays with photon flux around 1×10^{11} photons/s at $E/\Delta E = 3700$ and a tightly focused beam spot size ($\sim 200 \times 75 \mu\text{m}^2$). Due to the space limitation, we installed the interaction region of the CpVMI spectrometer at 0.8 m away from the focal point of the beamline, where the size of the photon beam is about $2 \times 3 \text{ mm}^2$. Since the light intensity from a bending-magnet beamline is four orders lower than that from an undulator beamline, we use the effusive molecular beam instead of the supersonic molecular beam to gain the gas intensity by three orders. At the same time, we set the electron arm in the weak focusing mode.

The test is done by measuring the photoelectrons image of Ar 2p photoionization with the photon energy tuned from 350 eV to 750 eV. Since the highest kinetic energy is 500 eV, we reduce the voltages to half of those in the simulation. Figure 4a shows a raw electrons image recorded at a photon energy of 750 eV, in which there are three distinct circles corresponding to 2p, 2s photoelectrons, and Auger electrons, respectively. However, the image was dominated by the strayed electrons at the center. Since our chamber was located 0.8 m away from the X-ray focus, it is very likely that the photon beam may touch somewhere on the chamber surface, and a huge number of photoelectrons and Auger electrons are created. Some of them may enter the lens and become focused at the center of the detector. These electrons persisted when the Ar gas was switched off. Fortunately, since they are at the center, the analysis of photoelectrons at the border of the image is not affected. Figure 4b shows a slice through the momentum distribution at $p_z = 0$ after applying an inverse Abel transformation with maximum entropy approach [28] of the image in Figure 4a. To make the assignment more robust, we repeat the measurement at several photon energies. The corresponding photoelectrons spectrum was shown in Figure 4c. We plotted the $\Delta E/E$ and ΔE as a function of photoelectrons kinetic energy in Figure 4d. We reached a resolution of 1.5% at the electron kinetic energy of 500 eV, and the ΔE barely depends on the kinetic energy of the electrons. It agrees well with simulation

in Figure 3a. We listed the performance of several VMI spectrometers that aimed for high kinetic energy in Table 1. Among them, we are the one tested with the highest electron kinetic energy.

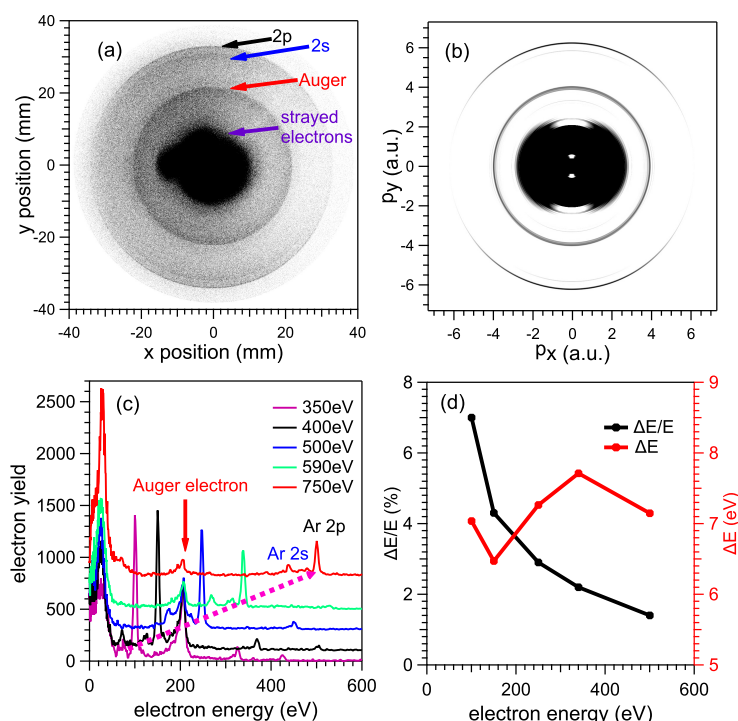


Figure 4. (a) The raw electron image of Ar ionized at the photon energy of 750 eV. (b) The slice at $p_z = 0$ of the 3D-momentum distribution after inverse Abel transformation. (c) The photoelectron spectra were obtained by angular integration of the complete 3D momentum distribution at different photon energies from 350 to 750 eV. (d) Electron kinetic energy resolution $\Delta E/E$ and ΔE as a function of electron kinetic energy.

Table 1. List of the performance of VMI spectrometers tested by measurements.

Author	e KE [eV]	$\Delta E/E$	Year	Reference
Schomas	65	3.00%	2017	[24]
Ablikim	100	3.80%	2019	[20]
Kling	60	1.00%	2014	[23]
Ghafur	100	1.80%	2009	[19]
Skruszweicz	300	4.00%	2014	[22]
This work	500	1.50%	2021	

4.2. Ion Arm

Since there is a distinct peak in the O^+ kinetic energy spectrum when O_2 is exposed to femtosecond laser [29], we tested the momentum imaging capabilities of the ion arm with a Coherent femtosecond laser (800 nm, 25 fs, 1 kHz). The 11 mm diameter laser beam was back-focused onto the supersonic molecular beam by a mirror of 100 mm focal length. Our measurements are record at $1.5 \times 10^{14} \text{ W/cm}^2$.

As described earlier, we set the working condition of the electron arm first, then find the voltage setting of the ion arm to focus the ions and enlarge the ions image in SIMION simulation. For the test of the ion arm, we repeat this measurement at different voltage settings of the electron arm. Figure 5 shows the raw images of O^+ with the kinetic energy of 5.6 eV at different settings of the electron arm: (a): 100 eV, (b): 300 eV, (c): 600 eV, respectively. The radius of the ions image decreases gradually from (a) 18 mm, (b) 12.5 mm,

to (c) 10 mm. In this way, It is possible to monitor the alignment of molecule while taking the electron image in the future.

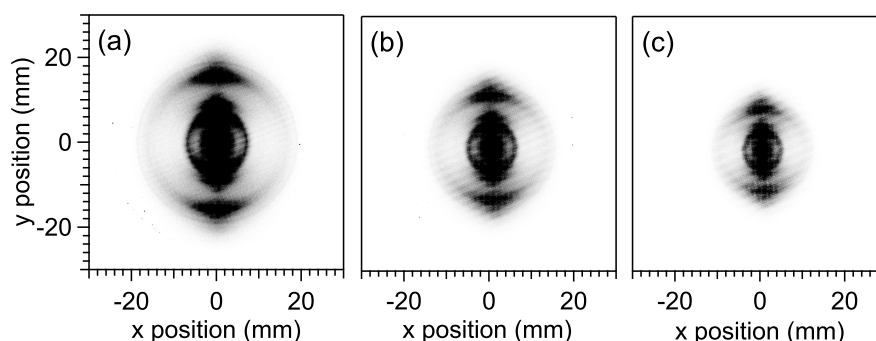


Figure 5. The raw images of O^+ ions by dissociative ionization of O_2 with a 800 nm femtosecond laser with an intensity of $1.5 \times 10^{14} \text{ W/cm}^2$ at different sets of the electron arm: (a) 100 eV, (b) 300 eV, (c) 600 eV, respectively.

Figure 6a shows the inverted momentum images (slice at $p_z = 0$) of O^+ ions obtained by applying an inverse Abel transformation with the Hansenlaw method [30] to the data in Figure 5a. The corresponding energy spectrum is obtained by integrating the momentum distribution over the polar angle between 60° and 120° with respect to the laser polarization, as shown in Figure 6b. Finally, the relative energy resolution of 4% is reached at the ion kinetic energy of 5.6 eV when the electron arm is set for 100 eV. In comparison, Ablikim et al. reached $\Delta E/E = 5\%$ at $E_{ion} = 10 \text{ eV}$ [20].

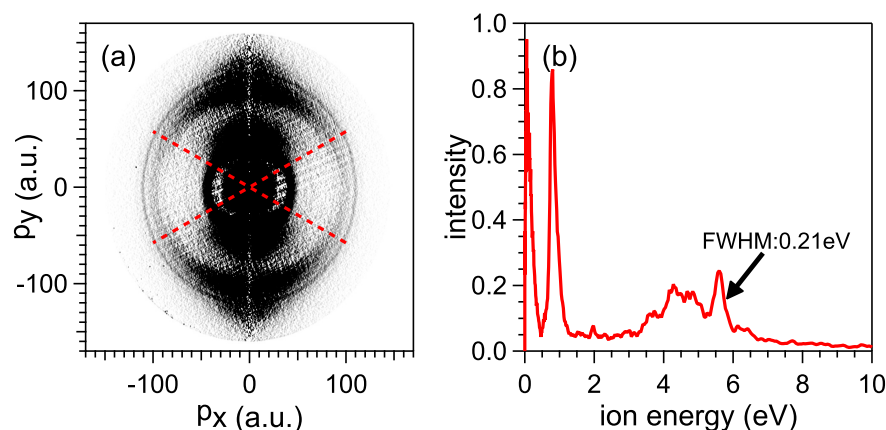


Figure 6. (a) The slice of 2D momentum (at $p_z = 0$) through 3D momentum distribution after a Abel inversion from the raw image of Figure 5a. (b) The kinetic energy spectrum of O^+ obtained by integration of the momentum distribution over the polar angle ranges between 60° and 120° with respect to the laser polarization.

5. Conclusions

We have presented the design and performance of a composite VMI spectrometer (CpVMI) optimized to measure both ions image and high-energy electrons image simultaneously at Shanghai soft X-ray free-electron laser. With the well-designed electric field, the spectrometer can detect the electrons with kinetic energies up to 1 keV and the ions with the kinetic energy of 20 eV simultaneously. With the experimental conditions investigated here, we reached a resolution of 1.5% at the electron kinetic energy of 500 eV. We also tested the momentum imaging capabilities of the ion arm at different voltage settings of the electron arm, reached a resolution of 4% at the ion kinetic energy of 5.6 eV when the electron arm is set for 100 eV. In a future upgrade, we plan to enable coincidence [20]

or covariance [17] detection of the high-energy electrons and the fragments ions using two novel timestamping fast optical cameras, Tpx3Cam [31,32], which can simultaneously measure the position and time of all charged particles with excellent spatial resolution and temporal resolution.

Author Contributions: Conceptualization: X.L. (Xiaojing Liu); Formal analysis: B.D., W.X., R.W., Y.F., L.T., Z.L. and X.L. (Xiaohong Li); Data curation: B.D., W.X., R.W., Y.F., L.T., X.L. (Xiaojing Liu), J.H. and X.L. (Xiaojing Liu); Investigation: B.D., W.X., R.W., Y.F., L.T. and X.L. (Xiaohong Li); Funding acquisition: X.L. (Xiaohong Li); Project administration: W.X. and X.L. (Xiaohong Li); Supervision, Z.L. and X.L. (Xiaohong Li); Writing, original draft: B.D., W.X. and X.L. (Xiaohong Li); Writing, review and editing: B.D., W.X., R.W., Y.F., L.T., X.L. (Xiaohong Li), J.H., Z.L. and X.L. (Xiaohong Li). All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest. The founding sponsors had no role in the design of the study; in the collection, analyses or interpretation of data; in the writing of the manuscript; nor in the decision to publish the results.

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