Multihit two-dimensional charged-particle imaging system with real-time image processing at 1000 frames/s

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(Received 14 November 2008; accepted 10 December 2008; published online 30 January 2009)

A high-speed imaging system developed for two-dimensional counting of charged particles is presented. Microchannel plates coupled with a phosphor screen of a short emission lifetime (<1 μs) are used to visualize the two-dimensional positions of charged-particle impacts, and the image on the phosphor screen is captured with a 1 kHz complementary metal oxide semiconductor (CMOS) image sensor (512×512 pixels). A multistage image intensifier consisting of the first and second generation devices was used to compensate for the low sensitivity of CMOS. The centers of gravity (COG) of individual light spots in each image frame are calculated in real time by a field programmable gate array circuit. The performance of this system is tested by time-resolved photoelectron imaging (TR-PEI) of NO using (1+1') resonance enhanced multiphoton ionization via the A 2Σ⁺ state with a femtosecond laser operated at 1 kHz. The new system enabled COG detection for more than ten particles in each frame at 1 kHz and achieved an extremely high degree of accuracy in the measurement of photoelectron angular distributions in TR-PEI. © 2009 American Institute of Physics. [DOI: 10.1063/1.3062945]

I. INTRODUCTION

Imaging technology has enabled multiplex (simultaneous) detection of photons and particles in various research fields, such as astronomy and microscopy. Efficient multiplex detection assists in experimental challenges to capture events with extremely low count rates, setting a new paradigm of experimental investigations. In atomic and molecular physics, information on the velocities (speed and angle) of scattered particles in photodissociation, photoionization, and elastic/inelastic/reactive collisions is crucial for elucidation of their dynamics. Chandler and Houston showed that three-dimensional (3D) velocity distributions of scattered atoms and molecules can be efficiently measured by two-dimensional (2D) charged-particle imaging (CPI) coupled with resonance enhanced multiphoton ionization (REMPI). Since REMPI allows for the selection of a single quantum state of atoms and molecules, CPI enables measurements of differential cross sections of state-selected particles, which have hardly been achieved by conventional techniques (with a few exceptions). The fundamental principle of CPI is widely applicable to one-photon ionization, electron impact ionization, ion pair formation, charge transfers, etc.

Cold target recoil ion momentum spectroscopy (COLTRIMS) is analogous to CPI, however, it employs a delay-line anode to register both positions and timings of particle impacts on the detector. Therefore, COLTRIMS offers direct 3D imaging of charged-particle velocities. Yet, its disadvantage is that dead times (∼ns) of the time-to-digital converters (TDCs) restrict detection of simultaneous particle impacts (multihits) on the detector. To improve the detection capability of multihits, various efforts have been made so far: for instance, a charge coupled device (CCD) camera (256×256 pixels) was used as a complementary readout with the delay lines, and the numbers of delay lines were also increased to discriminate particle impacts with short intervals. Despite these efforts, detection of more than ten incident particles is not practical with delay-line detectors (DLDs) at the present time. The camera-based CPI techniques have a clear advantage in observation of multihits. It is also noted that a spatial resolution is generally higher in CPI using cameras than with DLDs. These features of CPI are essential for the time-resolved photoelectron imaging (TR-PEI) presented in this work.

Without the timing information of particle impacts, CPI only observes a 2D projection of a 3D velocity distribution. However, when the 3D object has axial symmetry, mathematical procedures such as inverse Abel transforms can reconstruct the 3D distribution uniquely from a single 2D projection image. Such mathematical procedures had been well established and employed in various fields such as plasma diagnostics. If mathematical inversion is not possible due to noncylindrical symmetry of the original 3D distribution or zooming of the 2D image, then slice imaging, a variant of CPI, can be employed.

The original CPI by Chandler and Houston is based on the Wiley–McLaren time-of-flight mass spectrometer, in which the arrival position of a particle on the detector surface depends on both an ionization position and particle velocities perpendicular to the flight axis. However, a slight modification of CPI with an immersion lens (velocity mapping) by Eppink and Parker has almost completely removed the effect of an ionization position to make the image primarily depend on the velocity focusing in charged-particle optics.
After this improvement in charged-particle optics, it became clear that an imaging system [microchannel plates (MCPs), a camera, and image processing] limits speed resolution in CPI to approximately 2%-3%; a typical problem is that the diameter of a light spot on the phosphor screen is much larger than the pore size of a MCP. Therefore, center of gravity (COG) calculation\(^{19,20}\) is necessary (sometimes down to the subpixel\(^{21-24}\)) on each light spot in an image frame for enhancing the resolution. Aberration in the charged-particle optics becomes problematic in the much higher resolution range of 0.1%-0.2%.

It is noted that COG calculations fail if the light spots overlap each other. Therefore, the frame rate of imaging should be comparable with or exceeding the repetition rate of a pulsed laser that induces ionization events. Most CPI experiments so far have used pulsed lasers at repetition rates less than a few tens of hertz, therefore, both readout and real-time COG calculations could be performed on the shot-to-shot basis by a standard CCD camera and a personal computer (or with a dedicated frame grabber).\(^{19,20}\) It was also feasible to store the video-rate images (25 or 30 Hz) in hard disks or memory banks to apply COG analysis afterwards (offline). On- and offline methods were employed in our laboratory, and the same results were obtained.\(^{25}\) On the other hand, in TR-PEI using a 1 kHz femtosecond laser system, a CCD-based system is no longer applicable due to its low frame rate. It is also inconvenient to carry out offline analysis, as the data size of a 1 kHz video movie is quite large to store or transfer, particularly when we record a large number of movies in different experimental conditions. It is desirable to perform real-time image processing at 1 kHz to avoid data storage. In the present work, we have constructed an imaging system that enables readout and image processing at 1 kHz using a complementary metal oxide semiconductor (CMOS) image sensor with a multistage image intensifier and a field programmable gate array (FPGA) circuit.

The motivation behind the construction of a 1 kHz CMOS/FPGA system is not only the improvement of resolution; high fidelity is another important goal for us. Notice that MCPs, an essential constituent in CPI, convert the impacts of primary particles (electrons, ions, fast neutrals, etc.) into pulses of amplified secondary electrons. These pulse heights (intensities) are determined by stochastic processes in the microchannels of MCPs and are never identical to each other, which causes variations in brightness and sizes of light spots on the phosphor screen. Therefore, thresholding and binning in the brightness of light spots are necessary for counting the particle hits rather than recording the image directly. Such real-time image processing is necessary for achieving high-fidelity measurements even if the resolution is sufficiently high without COG calculations.

II. IMAGING SYSTEM

A schematic view of the 2D CPI system constructed in the present work is shown in Fig. 1. It consists of a dual MCP (Burle, 3075PM) backed by a P47 (Y\(_2\)SiO\(_5\) : Ce) phosphor screen, a multistage image intensifier, CMOS image sensor, and FPGA circuit. Both MCPs of the Chevron assembly are 10 \(\mu\)m \(\phi\) in pore size, 12 \(\mu\)m in pitch length (pore-to-pore spacing), 60:1 in aspect ratio (channel length: pore size), and 75 mm \(\phi\) in effective diameter. The bias angle of an individual microchannel with respect to the surface normal is 8°. The pore size and the bias angle of MCP affect uniformity of the sensitivity of MCP, as we discuss in Sec. V.

In the present work, we observe photoelectrons. The gain of the MCP assembly is set over 10\(^9\) (\(\sim\)880 V/a plate), for which a quasi-Gaussian pulse height distribution (PHD)
is anticipated. The output electron pulses are accelerated to excite a P47 phosphor screen with an emission decay time $\tau$ down to 10% in intensity of less than 1 $\mu$s. The short lifetime is essential in avoiding double counting of a single impact in the consecutive camera frames in measurements at 1 kHz. The image on the P47 phosphor screen is transferred through an optical fiber bundle to a quartz view port (80 mm diameter) for observation from the atmospheric side.

The camera system consists of a standard lens (Nikon, Ai Nikkor 50 mm F1.2S), a fast-gated, proximity-focused image intensifier (Hamamatsu, C6653MOD-B) coupled with an image booster (Hamamatsu, C4412), a relay lens, a 512 x 512 pixels CMOS image sensor, and FPGA circuit. A fiber coupling is not used for transferring an image on the phosphor to the CMOS sensor because the difference in diameter between the phosphor screen and the CMOS sensor is quite large. The use of the image intensifiers is unavoidable, despite the fact that they degrade imaging resolution, because the CMOS image sensor is much less sensitive than CCD. The gated image intensifier consists of a multialkali photocathode plate, a single MCP (25 mm $\phi$ active area), and a P46 phosphor screen coupled with a fiber optic plate to transfer the image to the subsequent booster (a first-generation image intensifier). The image from the booster is finally focused on the CMOS sensor through a relay lens. The total luminance gain of the system was set to 100 lm m$^{-2}$ lux$^{-1}$, and a characteristic decay time $\tau$ of the emission was $\sim$50 $\mu$s at 10% decay, which was sufficiently short compared to the 1 ms interval in measurements at a 1 kHz repetition rate. Thus, double counting of photoelectrons in two consecutive frames can be avoided. The 25 mm $\phi$ single MCP in the image intensifier is triggered synchronously with a Q-switch in the Nd:YLF (yttrium lithium fluoride) laser in our femtosecond laser system with a delay controlled by a digital pulse generator (Stanford, DG535). The gate pulse width for the intensifier is 100–200 ns, corresponding to 0.01% of the duty cycle, which almost completely rejects dark counts of the MCP/phosphor (0.6 counts s$^{-1}$ cm$^{-2}$ at a gain of 10$^7$).

A flowchart of our CMOS+FPGA imaging system is shown in Fig. 2. The readout timing of a CMOS sensor (9 V lux$^{-1}$ s$^{-1}$, 512 x 512 pixels, 16 $\mu$m pixel pitch) is synchronized with the 1 kHz laser trigger described above. The image output is 8 bits in dynamic range. This image can be stored in a 256 MB buffer random access memory (RAM) in a “live monitor” mode that is used for adjusting experimental conditions such as lens focus, gate pulse for the image intensifier, threshold levels for digitization (described later), and laser intensity. We monitored the live image with a standard 14 in. B/W video monitor at a video rate of 30 Hz. A sequence of 1000 images can also be saved live. In “event counting” mode, the captured 8 bit image is digitized so that intensities for individual pixels become “1” or “0” according to a threshold value. We set the threshold value to be 26, which corresponds to 10% of the dynamic range (=8 bit). After this process, the positions of unit intensities are detected by scanning an 8 x 8 window through the 1 bit image. When a light spot is detected in the 8 x 8 block, the COG $(x_c, y_c)$ is calculated in a 1/8-subpixelized coordinate of 4096 x 4096.

This subpixelization capability was prepared to obtain an extremely high resolution ($\Delta u/v < 1\%$), while this article presents only the results from a standard 512 x 512 image. All of the above image processings are implemented on a FPGA circuit. The typical size of a single light spot ranges from 5 x 5 to 7 x 7 pixels in 512 x 512 resolution. The number of incoming photoelectrons was controlled either by
varying the photoionization laser intensity or the gas target density to avoid overlapping of light spots on the image sensor, which may otherwise cause miscounting in centroiding calculations. The determined COG coordinates \((x_0, y_0)\) are finally stored and accumulated in the 256 MB buffer RAM as an image with a 16 bit dynamic range. The accumulated images on the buffer memory are transferred to a personal computer by TCP/IP through an Ethernet 100Base-TX and updated until the final frame is processed.

III. EXPERIMENTAL SETUP FOR TESTING IMAGING SYSTEM

The performance of the imaging system has been tested by two-color pump and probe time-resolved photoelectron imaging of NO via the \(A^2\Sigma^+\) state. The descriptions of the molecular-beam apparatus and the laser system are only briefly described here. More detailed discussion on the photoionization dynamics of NO is presented elsewhere.\(^{28}\) The molecular-beam apparatus is identical to that used previously\(^ {29}\) except for two modifications to achieve a higher quality in photoelectron images. One modification is that a new buffer chamber was introduced between a molecular-beam source and an ionization chamber for differential pumping to prevent effusion of background thermal gas from the source into the ionization chamber. Two electroformed skimmers (2 mm \(\phi\) orifice diameter) were used for both collimation of a supersonic molecular beam and restriction of conductance between the chambers. In the second modification, the electrodes for the acceleration of photoelectrons and a field-free drift region were entirely shielded with a 1 mm thick Permalloy tube against external magnetic fields.

5% NO seeded in Ar was expanded at a stagnation pressure of 760 Torr into the source chamber through a pinhole (50 \(\mu\)m diameter) to produce a continuous molecular beam. The source, buffer, and ionization chambers were pumped by turbomolecular pumps of 2000, 210, and 500 l/s, respectively. Each of these turbopumps was backed by a 37 m3/h oil-free scroll pump. A typical pressure in the ionization chamber during operation (beam on) was \(1 \times 10^{-8}\) Torr.

The 1 kHz femtosecond laser system employed here has been described elsewhere.\(^ {26}\) The output of a diode-pumped Ti:sapphire oscillator was amplified by a Nd:YLF-pumped Ti:sapphire regenerative amplifier to produce a 1 kHz pulse train with the output of 2.8 mJ/pulse light centered at 802 nm. The amplified femtosecond pulse was split into two equal intensity beams to pump two optical parametric amplifiers (OPAs) to generate tunable visible or UV pulses for pump-probe experiments. A 226 nm pump pulse was obtained by sum-frequency generation between the fundamental light from the regenerative amplifier and a 314 nm UV pulse from one of the OPAs. The probe wavelength (269 nm) was optically delayed with respect to the pump pulse using a hollow corner cube on a computer-controlled translational stage. Both the pump and probe laser pulses were then introduced collinearly into the ionization chamber. Two polarization vectors of the laser beams were aligned parallel to each other. The 226 nm pump pulse excites jet-cooled NO molecules up to the vibrational ground level of the \(A^2\Sigma^+\) (3\(\sigma^Rydberg\)) state and the probe pulse ionizes them. Typical energies for the pump and probe pulses were \(-0.1\) and \(2 - 3\) \(\mu J/pulse\), respectively.

The photoelectrons are accelerated up to a kinetic energy of \(-1\) keV in an electric field (\(-90\) V/cm) parallel to the supersonic molecular beam and projected onto a position-sensitive 2D imaging detector described above. A stack of eight stainless steel rings with a 33 mm inner diameter was used to make the acceleration electric field for velocity mapping so that the photoelectron image only reflects the linear momentum of the electron parallel to the detector face.\(^ {18}\) The rings were spaced by 3 mm \(\phi\) rings and linked to each other by 2 M \(\Omega\) resistors placed inside the ionization chamber. The negative potential of approximately \(-1\) keV applied to the base plate is successively increased by these dividing resistors up to the ground potential at the top electrode. Trajectory simulations were performed using the SIMION 3D software package (Scientific Instruments Services) to confirm that the velocity focusing condition was achieved.

IV. PHOTOELECTRON IMAGING

In the \((1+1')\) REMPI with polarization vectors of the pump and probe beams parallel to each other, the photoelectron angular distribution in a laboratory frame (LF-PAD) has cylindrical symmetry around the polarization axis \(z\). Thus, LF-PADs are characterized as a function of the polar angle \(\theta_h\) with respect to the symmetry axis \(z\) as follows:\(^ {30}\)

\[
\frac{d\sigma}{d\Omega} = \sigma_0 \left[ 1 + \beta_\perp P_2(\cos \theta_h) + \beta_\parallel P_4(\cos \theta_h) \right],
\]

where \(\sigma_0, \beta_\perp\), and \(P_4(\cos \theta_h)\) are the integral ionization cross section, anisotropy parameters, and the Legendre polynomial, respectively. According to Eq. (1), observed photoelectron images (2D projections of the 3D distributions) are expected to be identical at each quadrant in the image frame; however, the four distributions are almost always different in actual measurements.

We examined the asymmetry observed in photoelectron imaging, and we discovered that the incident angles of photoelectrons with respect to the bias angle of the microchannel cause the asymmetry, especially when a large pore size is used. Figure 3(a) shows a typical photoelectron image obtained by integrating the image on a phosphor screen without any image processing. The directions of the polarization vectors \(\epsilon\) of the pump and probe beams are horizontal in the plane of the figure. Here, we used a 2D-position-sensitive detector composed of a P20 phosphor screen and a Z-stack MCP with a 25 \(\mu m\) \(\phi\) pore size, a 31 \(\mu m\) pitch length, 40:1 aspect ratio, 8° bias angle, and a 77 mm \(\phi\) effective area. The gain of the Z-stack MCP was set around 10\(^7\) in these measurements. A standard image-intensified CCD camera having a 1024\(\times\)1024 pixel resolution was used for these particular measurements. The signal intensity was accumulated for 25 s on the CCD chip in a single scan, and the accumulation was repeated 12 times to obtain intensity-integrated photoelectron images. It is known that a Z-stack MCP provides a much narrower PHD compared to a chevron MCP. However, Fig. 3(a) shows that the signal intensity in

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the left-hand side is much greater than that in the right-hand side. As understood from Eq. (1), this asymmetry is due to an experimental problem: the intensities for \( \theta_l = 0^\circ \) and \( 180^\circ \) should be equal to each other.

V. EFFECTS OF INCIDENCE ANGLE OF PHOTOELECTRONS

Figures 3(a)–3(c) show the photoelectron images of different sizes [65, 38, and 26 mm in diameter (D), respectively] observed by changing the acceleration electric fields. The signal intensities along the symmetry axis \( z \) (a dotted line) for the photoelectron images of Figs. 3(a)–3(c) are plotted in Figs. 3(d)–3(f), respectively. Two peaks indicated with open and full circles are backgrounds due to scattered light from the rim of the phosphor screen and the photoemission from the acceleration electrodes, respectively. The intensities in Figs. 3(d)–3(f) are scaled to have the highest intensity to be unity. Both images and plots indicate that the degree of asymmetry diminishes with the decrease in diameter. The relative peak intensities on the right-hand side corresponding to the photoelectron ejected at \( \theta_k = 180^\circ \) are enhanced from 0.60, 0.72, to 0.87 for \( D = 65, 38, \) and 26 mm, respectively. When we rotated the entire MCP assembly by \( 180^\circ \), the relative signal intensities appearing on the left-hand and right-hand sides were completely reversed. The findings clearly suggest that the asymmetry arises from characteristics of the gain of MCP that depends on the incident angle \( \theta_i \) of photoelectrons to the detector face. The incident angles of \( + \theta_i \) that we calculated for photoelectrons ejected at \( \theta_k = 0^\circ \) using SIMION are also indicated in Figs. 3(d)–3(f) [see Fig. 4(b) for the definition of \( \theta_i \)]. As the photoelectron image size decreases, \( \theta_i \) decreases from \( 4.0^\circ \) \((D=65 \text{ mm})\) to \( 1.6^\circ \) \((D=26 \text{ mm})\).

The impact-angle dependence of the detection sensitivity of MCP has been investigated for both ions and electrons. Galanti et al. observed the characteristics of MCP using a monochromatic electron beam at various kinetic energies and angles of incidence to the detector. Figure 4(a) shows the relative gain of a single MCP as a function of the incident angle with respect to the direction of an individual channel at 1 keV. This figure is adopted from Ref. 33. Galanti et al. used a single MCP of 40 \( \mu \text{m} \) \( \phi \) in pore size, 50 \( \mu \text{m} \) in pitch length, and 40:1 in aspect ratio. As seen in Fig. 4(a), the efficiency rapidly rises with the incident angle from \( 0^\circ \) to \( 10^\circ \). After the maximum around \( 13^\circ \), it shows a negative dependence on the incident angle. They also measured an efficiency curve as a function of electron kinetic energy at the incident angle of \( 13^\circ \) and found that it has a maximum around the electron kinetic energy of 1 keV.

Figure 4(b) shows a schematic, cross-sectional view of a Z-stack MCP with a bias angle of \( \theta_B \) of the first plate and photoelectrons with incident angles of \( \pm \theta_i \) with respect to the surface normal. The bias direction of channels is also
shown with an arrow. In general, \( \theta_B \) in Chevron and Z-stack MCPs are optimized to around 8° by taking into account the efficiency and the ellipticity of light spots from a phosphor screen. At the largest image efficiency and the ellipticity of light spots from a phosphor MCPs are optimized to around 8° by taking into account the ejected at 

When an electron enters the microchannel almost parallel to its axis, deep penetration into the channel occurs before a primary interaction takes place, which reduces the gain in subsequent secondary electron amplification. This effect can occur for the observed results shown in Fig. 3.

Penetration into the microchannels is expected to decrease when a MCP with smaller pore size is used. We replaced a MCP with a 25 \( \mu \)m \( \phi \) pore size with a 10 \( \mu \)m \( \phi \) one and found that the asymmetry is considerably reduced. Even if we employed a new CMOS/FPGA system, a large variation in the gain profile of a MCP could never be completely compensated for, as the gain profile was not taken into account in our COG calculations. It was highly important to make the gain of the MCP/phosphor assembly as uniform as possible.

VI. PERFORMANCE OF THE PRESENT IMAGING SYSTEM

Under the experimental conditions described in Sec. III, 10–14 primary electron impacts were observed on the CMOS image sensor at each laser shot. Although our CMOS+FPGA imaging system has been designed to handle more than 50 multiths, the overlapping of individual light spots, which causes mismasurement of number of events, should be avoided for high-precision image measurements. A representative example of a raw photoelectron image (2D projection of 3D distribution) recorded with shot-to-shot event counting at 1 kHz is shown in Fig. 5(a). The directions of the polarization vectors \( \mathbf{e} \) of the pump and probe beams are parallel to each other and vertical in the plane of the figure. This raw image was accumulated for 600 000 laser shots (=frames), which corresponds to a 10 min integration time.

The number of electrons produced by each laser shot is maintained low to avoid a space-charge effect. Therefore, an appropriate integration time should be set to reach a sufficient number of electrons.

Figure 5(b) shows the photoelectron kinetic energy (PKE) distribution obtained from an inverse Abel transform of Fig. 5(a); a weak background image was subtracted from Fig. 5(a) prior to the analysis. In Fig. 5(b), an intense, single peak can be seen at PKE of 0.82 eV. Figure 5(c) displays an expanded view of Fig. 5(b) in the PKE region from 0.7 to 1.0 eV with the least-squares fit using a single Gaussian with full width at half maximum (FWHM) of 39 ± 1 meV. This corresponds to the energy resolution \( \Delta E_k/E_k \) of 4.8%. The typical bandwidths of our femtosecond pulses are 11 meV for 226 nm pump pulse and 32 meV for 269 nm probe pulse. Therefore, the energy uncertainty (FWHM) in our imaging setup is estimated to be approximately \( \sqrt{(11)^2 + (32)^2} \approx 34 \) meV in the two-photon ionization process. As can be seen in Fig. 5(c), this value is close to the observed FWHM (39 meV). This indicates that the energy resolution in the present imaging setup is almost limited by femtosecond laser bandwidth.

It should be noticed that the laser polarization direction (axis of symmetry) or the laser propagation direction should be precisely known for accurate analysis of images. As shown in Fig. 6(a), if the camera frame defined by the horizontal (column) and vertical (row) axes is tilted by the angle of \( \alpha \) with respect to the laser propagation direction \( k \), the observed image should be rotated prior to analysis. Normally, when a camera sensor is not set completely aligned to the laser polarization, this problem always occurs. We solved this problem by spatial mapping of \( \text{NO}^+ \) ions produced by photoionization of NO. By adjusting the voltages of acceleration electric fields, one can make the image dependent on the initial ionization point. The ions are formed along the laser polarization, this problem always occurs. We solved this problem by spatial mapping of \( \text{NO}^+ \) ions produced by photoionization of NO. By adjusting the voltages of acceleration electric fields, one can make the image dependent on the initial ionization point. The ions are formed along the laser polarization direction so that the line of ions will indicate the laser polarization direction. There is a small recoil velocity of \( \text{NO}^+ \) against an electron (9.7 m/s, 14.6 \( \mu \)eV), but it can be neglected for the present purpose. Figure 6(b)
shows a typical example of a NO$^+$ ion image recorded in a spatial mapping mode to trace the spatial region where the laser beams cross the supersonic molecular beam. The image shown in Fig. 6 is an expanded view of the original 512x512 full frame image. In our experimental setup, two irises are set in front of the entrance and exit quartz windows for the alignment of laser beams. Since the irises are separated by approximately 600 mm, the optical passes of both the pump and probe beams can be easily aligned within ±1 mm diameter, which corresponds to a precision of 0.1°. Thus, once the laser propagation is found experimentally, we can accurately recover its direction. From both ends (188 and 358 columns) of the line-shaped NO$^+$ image, we determined the rotational angle $\alpha$ to be 0.45° in the present case.

The axially symmetric photoelectron distribution creates identical images in four quadrants. Therefore, comparison of the four quadrants serves critical tests of the performance of our system. Figure 7 shows photoelectron angular distributions extracted from the four quadrants of Fig. 5(a), shown in the inset. These angular distributions were obtained by integrating the radial part of the Abel-transformed image (the details are found in Ref. 28). The open circles are experimental values and the solid lines are the least-squares fits of Eq. (1). For each quadrant, the sets of anisotropy parameters $(\beta_2, \beta_4)$ determined from the fits are (1.66, 0.11), (1.66, 0.12), (1.65, 0.09), and (1.66, 0.11), which are also listed in Fig. 7. The standard deviations $\sigma$ of anisotropy parameters of $\beta_2$ and $\beta_4$ are as small as 0.005 for $\sigma(\beta_2)$ and 0.016 for $\sigma(\beta_4)$, demonstrating the unprecedented accuracies obtainable with our new system. It should be noted that such small values of $\sigma$ are obtained only when the raw image is rotated by $\alpha$. In fact, we searched the rotational angle $\alpha$ to find the minimum values of $\sigma(\beta_2)$ and $\sigma(\beta_4)$ and confirmed that the estimated $\alpha$ coincides with the value determined from the NO$^+$ image.

**FIG. 6.** (Color online) (a) Schematic diagram showing the camera frame defined by column and row axes and the laser $k$ vector. Here, the camera frame is tilted by an angle of $\alpha$ with respect to the $k$ vector. (b) An expanded view of the original 512x512 full frame image for NO$^+$ ions in space-mapping mode to trace the spatial region where the laser beams cross the supersonic molecular beam.
In conclusion, a high-speed CMOS+FPGA camera has been developed for multi-hit, 2D counting of charged particles. The imaging system can capture images at 1000 frames/s in 512×512 pixel resolution, can calculate the COG coordinates for more than 50 multi-hit events, and can provide efficient and precise measurements of 3D photoelectron/photoion scattering distributions. We also investigated the inhomogeneous detection sensitivity of MCP and found that a deep penetration of incident electrons into microchannels before a primary interaction at the input MCP causes a reduction in the gain, and it can be effectively improved by using a MCP with a small pore size. The present development leads to an exact determination of molecular-frame photoelectron angular distributions from highly precise LF-PAD measurements for the NO $^2\Sigma^+$ state ionization, which is one of the benchmark systems in molecular photoionization dynamics.

ACKNOWLEDGMENTS

We thank Dr. Masaaki Tsubouchi for his contributions in the early stages of this work. T.H. acknowledges the Special Postdoctoral Researcher Program at RIKEN. This work was supported in part by the Research Project “Hydration dynamics and molecular processes” at RIKEN.