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International Journal of Mass Spectrometry 238 (2004) 33–46

International Journal of Mass Spectrometry

www.elsevier.com/locate/ijms

Review

Recent advances in detector-array technology for mass spectrometry

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Received 24 February 2004; accepted 13 August 2004
Available online 13 September 2004

Abstract

Array detectors capable of determining the spatial distribution of particles, whether photons, neutrons, atoms, or others, have long existed. More recently, this technology has been applied to mass spectrometry for the simultaneous detection of multiple ions of differing mass-to-charge (*m*/*z*) values. When simultaneous detection is utilized in mass spectrometry, benefits such as improved detection limits and precision, reduced sample consumption and analysis time, and the elimination of correlated noise sources are realized. Presented here is a review of some of the relevant technologies that have been or could be applied to mass spectrometric ion detection. The array detector technologies are classified according to detection method; the figures of merit of each type of array are discussed.

Keywords: Array detectors; Multiple detectors; Mass spectrometry; Simultaneous detection

Contents

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doi:10.1016/j.ijms.2004.08.004

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1. Introduction

Simultaneous detection in mass spectrometry offers several key benefits. With simultaneous detection, the duty cycle of the measurement increases, since there is no longer a need to scan the instrument to obtain a complete mass spectrum. Since all m/z values are continuously monitored, unlike with scanning instruments, the sample size required for complete analysis goes down. This benefit leads to better absolute detection limits. An additional benefit of simultaneous detection is the ability to eliminate correlated noise sources through ratioing techniques. Since correlated noise affects all signals identically, one can ratio two m/z signals to remove it, provided they are recorded simultaneously. This process can raise the signal-to-noise ratio of a measurement, resulting in improved detection limits and measurement precision. A final benefit of simultaneous detection is the ability to accurately record transient signals, such as those produced from chromatographic separations, laser ablation, or flow injection. Because rapidly changing analyte concentrations can generate transient signals, the finite length of time required to move between m/z values in a scanning instrument introduces errors into concentration measurements, known as spectral skew. With simultaneous detection, all m/z values are continuously monitored, eliminating this source of error.

In order to obtain truly simultaneous multichannel detection, a spatially dispersive mass spectrometer is required. Of the available instrument geometries, only magnetic-sector instruments (also know as sector-field mass spectrometers, SFMS) are dispersive. Several other types of instruments can approximate simultaneous detection. Time-of-flight (TOF-MS) and ion trap (ITMS) instruments extract a representative population of a sample and then sequentially record a mass spectrum. With simultaneous extraction, spectral skew and correlated ionization-source noise are eliminated. Additionally, the duty cycle of the measurement increases slightly. Unfortunately, though, correlated noise sources affecting the mass analyzer and detection electronics cannot be eliminated. Fourier transform ion cyclotron resonance (FT-ICR) instruments can simultaneously record a mass spectrum, and therefore achieve most of the benefits associated with simultaneous detection. One drawback of FT-ICR is the time required to record a mass spectrum, leading to limited duty cycle.

There is one notable alternative to SFMS for simultaneous detection: the use of parallel mass spectrometers. Studies have been performed with parallel QMS systems to improve isotope-ratio precision [1–3], but with such an apparatus, the measurement of many m/z ratios becomes prohibitively expensive. An array of nine parallel miniature QMS analyzers has been fabricated [4,5]. Although this system was used to monitor only a single m/z ratio, it would be possible to modify it to determine a different m/z ratio in each analyzer. There are several reports of arrays of miniaturized ITMS, with each ITMS tuned to a specific m/z ratio [6–8]. This method has the best prospect as an alternative to SFMS for simultaneous detection, since one set of controlling electronics can be used for the entire array.

Presented below is a review of the available detector technology for performing simultaneous multichannel detection. The vast majority of research into improving spatially resolved ion detection does not stem from mass spectrometry research, but rather from fields such as high-energy particle physics and astronomy. A strict adherence to mass spectrometry detector arrays will not be made, but instead this review will survey the technologies that can be applied to ion detection in mass spectrometry.

2. Characteristics of an ideal detector array

As a target, it is useful to set down the characteristics that would be desired in an ideal multichannel detector. The array should be sensitive to a single ion while still being capable of handling ion fluxes upwards of 10^{10} counts per second (cps). With this feature, both bulk and trace analyses can be handled simultaneously. In order to accommodate a wide range of ion fluxes, the array must be linear over at least 10 orders of magnitude, have a high gain, and very little read noise. The array must also give the same response regardless of the ion or ion energy incident upon it. Each individual detector in the array must respond identically to every other detector in the array or, as a minimum, each detector's response must be highly reproducible, so the detector-dependent response can be normalized. The array should be free of dark counts and thermionic noise, problems that plague electron-multiplying detectors. Another problem affecting electron-multiplying detectors is that the response is governed by dynode statistics. The probability of obtaining a given detector response for a particular ion should resemble a delta function, not a Gaussian distribution. Direct integration of the ion signal is desired, since each conversion step in the detection process will introduce noise into the measurement.

Table 1
Characteristics of ion-sensitive detector arrays[a]

Detector type	Resolution (μm)	Simultaneous detection/integration	Speed (μs/acquisition)	Cost	Uniform response	Dynamic range	Limit of detection (# of ions)
Chemical	<1	Y/Y	10^6–10^8	+++	−−−	10^2	10^4
Multiple collector	10[b] 2000[b]	Y/Y	<1	−−−	+	10^{10}	1
Microchannel plate arrays							
Photon detection	100	Y/Y	10^3	−−	−−	10^3	10
Charge division							
Resistive anode	100	N/N	<1	+	−−	10^6	1
Delay line	25	N/N		+	−	10^7	1
Shaped anode	20	N/N	<1	0	−	10^6	1
Capacitively coupled	100	N/N	1	0	−	10^6	10
Discrete anode	10	Y/Y	<1	0	++	10^7	1
Integrated array							
Tracy array	10	Y/N	10^3	−	++	10^5	10^2
Active pixel sensor	10	Y/Y	10	−	++	10^4	10^2
MFCA	150	Y/N	10^4 current 10^3 proposed	−−	+++	10^7	10^4
FPC	175 current 10 designed	Y/Y	10^3	−−	++	10^8	10

[a] The properties of each array has been classified from greatest advantageous (+++) to average (0) to greatest disadvantage (−−−).

[b] 10 μm for one detector, 2000 μm between detectors.

Certain geometric restraints must be placed on the detector array. The individual detectors must be small enough so they do not limit the resolving power of the instrument. A minimum of 5–10 detector elements should be used to detect each *m*/*z* value so that the peak shape can be known to a reasonable degree [9]. Often, due to non-flat-topped peaks, it is necessary to measure peak area instead of maximum to obtain the most accurate representation of a mass spectrum. Additionally, the array must be large enough to cover the entire focal plane of the instrument. Typically, this means covering 5–10% of the total desired mass window but, ideally, an instrument could be designed to cover the entire mass window of interest.

Often a detector array is placed in harsh environments such as ones with a corrosive atmosphere, extreme heat or cold, or high levels of radiation. Because of this, the detector must be robust and have a long, reliable lifetime. If the lifetime of the array is short, the detector needs to be inexpensive to replace. Additionally, if the array is to be used outside a laboratory setting, it should be a low-power device. Finally, the array operation and maintenance should be simple.

No existing detector array possesses all of these features. Therefore, it is necessary to analyze what the array will be used for, and determine the most desirable set of properties. The remainder of this text will discuss different types of detector arrays, some of which have existed for nearly a century while others are newly developing technologies. Much of the technology that has been applied to mass spectrometric detection was not originally designed with that propose in mind, but instead the detection of energetic particles, either charged, neutral or photon. With little adaptation, though, the transfer can be made. Therefore, a strict adherence to mass spectrometry will not be attempted and we will use examples from other areas of science. The properties of the detectors discussed below have been summarized in Table 1.

3. Chemical detector arrays

Mass spectrometry detector arrays are as old as the science of mass spectrometry. The initial detectors used by Thomson [10] and Aston [11,12] can be classified as chemical detector arrays (CDA). These arrays, more commonly known as photographic plates or emulsions, were the standard detector array for more than 60 years, from the beginning of the 20th century to the early 1960s. The CDA works simply by exposing the focal plane of a mass spectrometer to an ion-sensitive material. The effect of ions on photographic plates was first explored by Koenigsberger and Kutchewski in 1910 [13,14]. Since then, much of the technology initially developed by companies such as Ilford Ltd. and Kodak has been adapted for mass spectrometric use. These detectors have traditionally been used in spark-source MS, but have also been used with most other atomic mass spectrometries, as well as several applications to organic mass spectrometry [15].

There are several benefits to mass spectrometric detection with CDA. Each CDA has a virtually limitless number of detector elements, with the size of each element being equal to the size of a grain in the photographic emulsion. This property provides high-resolution measurements and a broad mass range. The ion current is directly integrated by the CDA and the CDA itself serves as the storage device for the mass spectrum. The technology for CDA detection is mature and well understood. The photographic plates are inexpensive and disposable. Additionally, multiple mass spectra can be acquired on each plate. Lastly, the size of the CDA can be adjusted to match the focal plane of the mass spectrometer.

Unfortunately, there are numerous drawbacks to CDA detection. The dynamic range is very limited, on the order of 10^1–10^3 [16]. In order to record a measurable image, a minimum of 10^3 ions is necessary [17]. They are also inherently non-linear, which complicates quantitative measurements. Calibration of CDA detectors is difficult and time consuming due to plate-to-plate variations. Moreover, processing and analysis of a photographic plate can take several hours, reducing the duty cycle for multiple experiments. Although each CDA is relatively inexpensive, for repeated analyses the cost of preparation and development can be high.

Little research has been performed to improve these shortcomings, due to the development of alternative detector arrays. Several of the more important advances should be noted, though. Devices have been developed to aid in the analysis of each CDA, such as computer-controlled position and density measurements, leading to quicker analyses and improved accuracy [16,18]. Additional work has been performed in the development of new emulsions and ion-sensitive materials [19,20]. With these materials, the dynamic range and linearity can be slightly improved. A further advance has been improved instrumentation for plate positioning and changing [21].

4. Multiple-collector detector arrays

The use of multiple single-channel detectors is one of the simplest methods for realizing the benefits of simultaneous detection. Much of the technology for single-channel detectors is well established, leaving little design work necessary for its implementation. In fact, the advances in multiple detectors have closely followed developments in single-channel detector technology. This method is very versatile, with several types of detectors available to suit the needs of an experiment. By employing several types of detectors, a dynamic range of 10^{10} is obtainable. Most SFMS instruments do not have a linear mass scale, but with this approach, detectors can be placed as needed to intercept the desired m/z ratios [22].

The use of multiple detectors does have some limitations. For multiple detectors, a multiplicity of controlling electronics is needed as well as a means of parallel data acquisition, although with advancing computer power, the latter restriction becomes less problematic. Additionally, in order to achieve comparable resolution to that obtainable with a CDA, either the detector elements must be on the order of micrometers wide or the peak spacing must be increased. Finally, it is often important to acquire tens to hundreds of m/z ratios, so the use of multiple detectors becomes quite costly.

Two types of detectors, Faraday cups and electron multipliers (EM), have been widely used in this method of simultaneous detection. Faraday cups are advantageous for several reasons. In its simplest form, a Faraday cup is an electrically isolated conductor, so a thin wire can be used. Wires are commonly fabricated with diameters on the order of single to tens of micrometers, which will permit high-resolution measurements. Additionally, Faraday cups and their measurement instrumentation require little power and with current charge-sensing technology, as few as 10^4 ions can be detected.

Electron multipliers can produce a detectable signal from a single ion due to their internal gain of 10^6–10^7. Much of the initial research on electron multipliers was performed by Allen [23,24], who used discrete beryllium-plated nickel dynodes to produce a gain of 10^5 for ions having an energy of 0.05–20 keV. With improvements in plating material and increasing the number of dynodes, the gain of this type of EM, also known as a discrete-dynode EM, can be increased to over 10^7 [25]. The other major type of EM is the continuous-dynode EM. This was initially conceived by Farnsworth and successfully implemented by Goodrich and Wiley [26,27]. With this type of EM, the discrete dynodes have been replaced with a semiconductor-coated insulating channel.

In both types of EM detectors, a potential difference is applied across the dynodes, such that the front of the detector is less positive than the back. This is accomplished in a discrete-dynode EM with a series of resistors placed between adjacent dynodes. In contrast, the inherent resistance of the surface coating in a continuous-dynode EM provides a gradual voltage drop across the channel. With the correct choice of low work-function plating material, an energetic particle striking a dynode causes several secondary electrons to be emitted. Although much of the information about the exact composition of a dynode plating is proprietary, many of these alloys are beryllium–copper based. Due to the potential difference across the EM, the secondary electrons are accelerated towards subsequent dynodes, creating an avalanche of electrons with each impact.

Electron multipliers have several drawbacks for use as ion detectors. Often, photons or neutral particles can be energetic enough to liberate electrons from the dynode surface, creating a high background. Additionally, the number of electrons liberated per collision is dependent on the energy of the interaction. Therefore, large molecules with high degrees of vibrational freedom will absorb internally much of the collisional energy, resulting in a loss in production of secondary electrons and detection efficiency. For these detectors to perform efficiently they must be operated at reduced pressure, or the mean free path of the secondary electrons is not sufficient to gain the kinetic energy needed to produce a multiplicative effect. Finally, since the electron gain is dependent on the energy and number of collisions, these detectors generate dynode statistics and produce a Gaussian gain distribution.

Despite the aforementioned limitations, multiple-collector detectors have been successfully employed in mass spectrometry. The use of multiple single-channel detectors for simultaneous detection first appeared in the literature in 1947 [28]. On this particular device, two Faraday cup

Table 2
Commercially available multi-collector mass spectrometers

Manufacturer/instrument	Instrument type	Number of Faraday cups	Number of EM	Moveable detectors
Cameca NanoSIMS 50	SIMS	1	6	Y (4 EM, 1 F)
PDZ Europa 20-20	IRMS	3	0	N
ThermoFinnigan Neptune	ICPMS	9	8	Y (8 EM)
ThermoFinnigan Triton	TIMS	9	8	Y (8 EM)
GV Instruments IsoPrime	IRMS	10	0	N
GV Instruments IsoProbe-T	TIMS	9	7	Y
GV Instruments IsoProbe	ICPMS	9	7	Y
Nu Instruments NuPlasma HR	ICPMS	12	3	N
Nu Instruments Noblesse	Noble Gas MS	1	2	N

detectors were placed sequentially near the focal plane of the mass spectrometer. A slit in the foremost detector allowed one *m*/*z* ratio to pass through it to the second detector. The next generation of this instrument was modified to be field-portable and included an EM in addition to two Faraday cups [29]. Another notable implementation of multicollector mass spectrometry was by Spencer and Reber [30,31], whose instrument was placed aboard the Explorer 17 satellite. This particular device was equipped with seven fixed-position Faraday detectors for analysis of the gas composition of the upper atmosphere and outer space. Multiple collectors were first used in plasma-source MS (PSMS) in 1992 when Walder and Freedman [32,33] coupled an ICP to a SFMS with seven fixed-position Faraday cups for isotope-ratio analysis. This system was later commercialized and marketed as the VG Elemental (now ThermoElemental) Plasma54. Since then, arrays have been developed consisting of Faraday cups [34], EM detectors [35], or hybrid systems utilizing both detector technologies. All current PSMS instruments capable of simultaneous detection employ this type of technology. These instruments, as well as other commercially available simultaneous-detecting mass spectrometers, are summarized in Table 2.

Though the use of multiple collectors in mass spectrometry has reached maturity, several significant advances have been made over the last several years. Jochum, et al described an array of 20 continuous-dynode EM consisting of 2.3-mm thick detectors for use with SSMS [35]. A series of slit plates was placed in front of the detectors to vary the resolving power from 300 to 3000. One limitation of the system is its inability to monitor adjacent *m*/*z* ratios at high mass due to the size of the detectors. This deficiency has been overcome in several instruments through the use of a zoom lens capable of increasing the mass dispersion [34,36].

A problem when measuring adjacent *m*/*z* ratios of significantly different abundances is that the shoulders of the more intense peak often interfere with the less-intense peak. In order to increase abundance sensitivity, Merren [37] introduced a mass spectrometer with an electrostatic analyzer between the magnetic sector and one of the detectors to eliminate peak shoulders. This detector was used to monitor the less-abundant *m*/*z* ratio, since poor abundance sensitivity is most detrimental to weak *m*/*z* ratios.

A new type of L-shaped EM has been developed in which the detector body is placed at a right angle to the path of the ion beam [38]. From the focal plane of the instrument, the ion beam enters a narrow channel and is confined as it travels down the channel to a conversion dynode, after which the secondary electrons are focused onto the entrance of the EM. By varying the length of the confining channel, several of these L-shaped devices can be stacked together to cover a much narrower region of the focal plane than traditional EM detectors, as shown in Fig. 1. A variation of this design was developed by Brown et al. [39], who used the natural angle of the mass-spectrometer focal plane to place detectors sequentially such that the majority of the detector body was concealed by the previous detector. In this setup, the conversion dynode of each detector was exposed to the focal plane while the rest of the detector was out of the path of the ion beam.

Several other studies have been performed to improve the reproducibility and eliminate errors in isotopic measurements. Rehkamper and Mezger [40] used Tl as an internal standard to eliminate Pb mass-discrimination effects in a MC-ICPMS instrument. Waight et al. [41,42] discussed some of the problems associated with Sr isotope ratio-measurements by MC-ICPMS. The decay of ^{87}Rb to ^{87}Sr can be used as a marker for geochronology, but often interferences arise from Kr, doubly charged rare-earth elements, and calcium dimers and argides. Without spiking methods, isotope-ratio measurements similar to those obtainable with TIMS were obtained. A more detailed review of other advances in isotope-ratio mass spectrometry can be found in the literature [43].

5. Microchannel plate detector arrays

One of the most important and most often used devices for position-sensitive detection is the microchannel plate detector (MCP). Initially developed by the Army Night Vision Laboratories, much of this technology was classified until 1971. Almost immediately after this declassification, publications began to appear in the literature for MCP-based position-sensitive detectors. The array consists of upwards of 10^6 microscopic glass channels, each 5–50 μm in diameter, bound together and electrically connected. Each channel

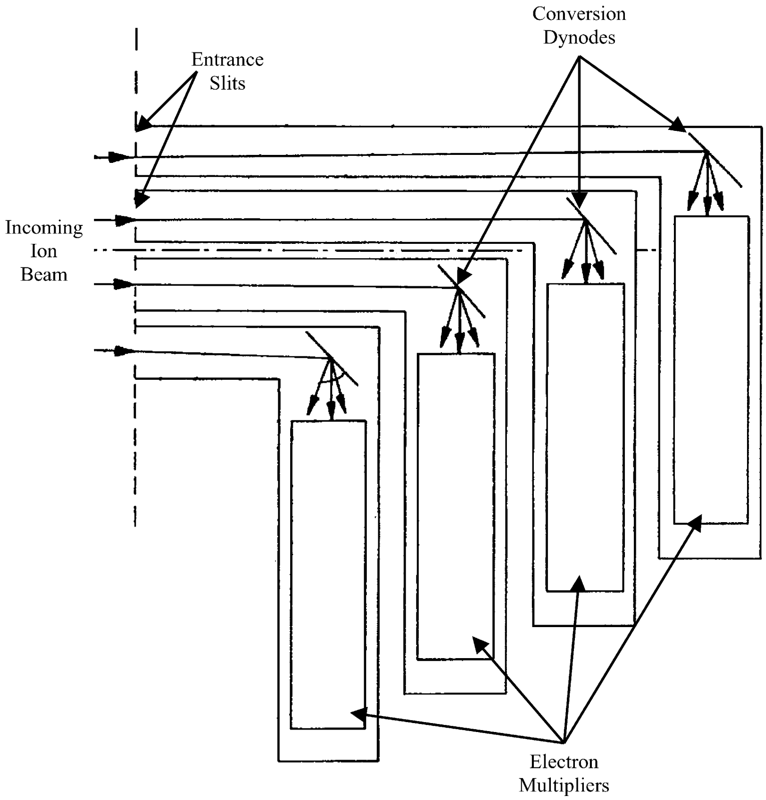

Fig. 1. The use of L-shaped electron multipliers allows adjacent detectors to be placed closer together. Reproduced from ref. [38].

operates as a continuous-dynode EM with a gain on the order of 10^4 [44]. With gains higher than this, atoms inside the channels can become ionized, producing “ion feedback” and instability. In order to increase the gain, two or three MCP detectors can be placed in sequence in configurations know as chevron and z-stacked, respectively [45]. Typical gains with these configurations range from 10^6 to 10^8. Since the secondary electrons are confined to the channel from which they originated, it is possible to obtain spatial resolution from the MCP, provided a means of recording the final electron-cloud distribution is available.

The MCP is a well characterized device, and numerous articles describe its operation. Studies have been performed on the effects of temperature [46,47], magnetic fields [48,49], and channel shape [50,51] and size. Additional research has been performed on the basic operating characteristics of the MCP, such as its lifetime [52], efficiency in detecting both charged particles and photons [53–58], the level of dark counts [59–61], and the effects of operating the devices at high count rates [62–64]. More recent studies have concentrated on improving the performance of the MCP. Deconihout et al. [65] placed a positively-biased grid in front of the MCP to insure that no secondary electrons escaped from the channels. Both Price and Fraser [66] and Tremsin and Siegmund [67] calculated the charge-cloud distribution exiting the MCP in order to better understand its effects on spatial resolution. Recently, a device called a microsphere plate detector (MSP) was developed as an alternative to the MCP [68–70]. In the MSP, glass beads with diameters of 20–100 μm are sintered together to form an array of irregularly shaped channels. Due to this irregularity, positive-ion feedback is effectively eliminated, leading to single MSP gains of over 10^6 [68]. A problem of the MSP is the lack of confinement of secondary electrons, leading to degraded spatial resolution.

Although a MCP can provide a spatially encoded cloud of electrons, a means of detecting the electrons is necessary that can retain the spatial information. Much of the driving force for developing and improving this type of technology has been for high-energy photon and particle physics. Since several reviews exist covering these types of detectors [71–74], an exhaustive coverage will be omitted here. MCP-based position-sensitive detectors can be divided into four main categories: photon detectors, charge-division detectors, capacitively coupled detectors, and integrated circuit arrays.

5.1. Photon detectors

Photon detectors operate by converting the cloud of electrons produced by a MCP into a photon signal by means of materials such phosphor screens [75] or scintillating crystals [76]. The photons can be readily detected by a conventional photon array detector such as a vidicon [75], CCD [77], photodiode array (PDA) [78] or even a photoplate. A schematic diagram of this class of detector, more commonly known as an electro-optical imaging detector (EOID), is shown in Fig. 2.

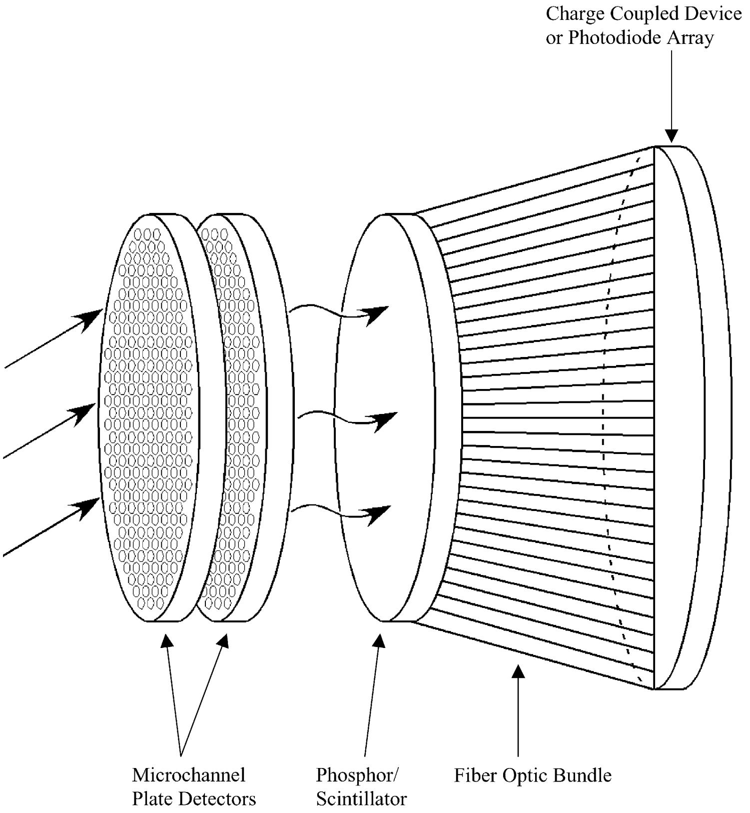

Fig. 2. Schematic diagram of the electro-optical imaging detector (EOID).

The first EOID-like detector was developed by Siegbahn et al. [79] for use in electron spectroscopy. This device consisted of a 167 × 32 array of miniature electron multipliers covering a range of 25 mm in the long axis. The electron output of the array was converted to photons with a phosphor screen and recorded with a vidicon camera. A year later, Riegler and More [78] developed the first MCP-based EOID for use as a UV and X-ray position-sensitive detector. The EOID was first applied to mass spectrometry by Giffin et al. [75] who used a Mattauch-Herzog geometry mass spectrograph and a MCP/phosphor/vidicon array with a series of laminar fiber-optic bundles to transfer the mass spectrum to a smaller footprint on the vidicon. Other research groups soon followed, producing similar devices for mass spectrometry [80,81]. A notable achievement for the EOID was the inclusion of a conducting phosphor into the detector [82]. This simplifies the acceleration of electrons onto the phosphor, which leads to better spatial-resolution integrity and ensures a high electron-to-photon conversion rate.

There are several advantages to the EOID for ion detection. It is a completely electronic detection system, eliminating the need for lengthy data interpretation and analysis. The spatial resolving power of the detector is a function of the MCP pore size and the pixel size of the photon detector. With small enough dimensions, this can be as low as 50–100 μm [83]. Problems do exist with the EOID, though. The conversion of ions to electrons, photons, and finally electrons again introduces noise into the measurement, which compromises quantification. The EOID, as with all MCP-based detectors, suffers from a non-uniform response across the array due to variations in the MCP and the phosphor.

5.2. *Charge division detectors*

Charge-division-based array detectors are the most common and easily employed of the MCP position-sensitive detectors. They function by employing an anode structure to intercept the electron cloud from the MCP and divide it among several sensors. Depending on the signal arriving at the sensors, mathematical interpretation can be performed to determine the centroid of the charge cloud. This type of detection is beneficial for several reasons. As mentioned before, it is a simple technology that is easy to implement. Spatial resolution of tens to hundreds of micrometers is typical and is often limited by the MCP and controlling electronics [74]. Therefore, with smaller-pore MCP detectors and faster electronics, spatial resolution can be improved. Although most of these devices rely on detecting single ion events, some are capable of measuring simultaneous events or ion fluxes of over 10^6 cps [84]. The limitations of these devices will be discussed with each type of anode structure. The types of anode structures include resistive division, delay line, and shaped-anode division.

5.2.1. *Resistive anode array detectors*

A resistive anode device consists of a layer of uniform-resistance semiconductor material with resistances typically of several $\mathrm{k}\Omega\,\mathrm{cm}^{-1}$ [85]. These devices have a multiplicity of sensing electrodes to measure the voltage that arises from an impinging electron cloud. Since the anode is resistive, the voltage created by the electron cloud will be divided amongst the sensors in proportion to the distance from the sensor. Therefore, the ratio of the voltage at any two sensors is proportional to the location of the electron-cloud impact between the two sensors. A linear-resistive anode detector was first applied to mass spectrometry by Carrico et al. [86]. Several years later, two-dimensional resistive-anode detectors were developed for particle imaging [87] and mass spectrometry [88].

The major limitation of the resistive-anode detector is its inability to detect simultaneous events. Since the position resolution depends on the ratio of voltages at a series of sensors, the detector has the ability to determine only the centroid of the voltage. Therefore, with simultaneous events, the average location of the detector events will be determined, not the location of each event. The maximum count rate is limited by the RC time constant of the anode and, with current technology, this limits ion beam fluxes to below 10^6 cps [89]. Another major limitation of this device is temperature fluctuations caused by resistive heating of the anode. Localized heating of the anode takes place with spatially-varying ion fluxes, such as those from a SFMS. The temperature fluctuation causes resistance changes in the anode, leading to errors in centroid location. This can be overcome to some extent by use of a Peltier cooler [85].

5.2.2. *Delay-line array detectors*

A type of detector that has overcome some of the problems of the resistive-anode detector is the delay-line detection method. This detector in its most basic form consists of a wire whose ends trigger the start and stop of a high speed timer. The wire is typically zig-zag or helical-shaped and meanders back and forth across a large planar footprint. When a cloud of electrons hits a portion of the wire, a current pulse propagates down the wire in both directions at typical speeds of 100–500 $\mu\mathrm{m\,ns}^{-1}$. A high speed timer can be used to measure the difference in arrival times of the current pulses at each end of the wire. The delay between pulses is then proportional to the location of the electron cloud along the wire. Typically, the time resolution is not sufficient to determine the exact location of a MCP event in two dimensions. Therefore, to obtain two-dimensional information, orthogonal delay lines can be used to pinpoint the exact location. Position resolution of as little as 25 μm has been reported [90].

The delay-line detector eliminates the problems of anode heating associated with the resistive anode detector. Additionally, since the rate of current propagation governs the speed of this detector, and not an RC time constant, the delay line detector can handle higher count rates, typically an order of magnitude greater than the resistive anode. Still, though, simultaneous events cannot be accurately recorded with one or two delay lines. Recently, Jagutzki et al. [84] described using at least three delay lines and mathematical algorithms to determine the position of particles arriving simultaneously at a MCP.

5.2.3. *Shaped-anode array detectors*

A method to provide high spatial resolution with a minimal number of detector elements is through the use of detector anodes that exhibit a known spatial variation. This type of array, originally known as a wedge-and-strip anode, was developed by Anger [91] and modified into its current form by Siegmund et al. [92]. The initial device consisted of two sets of matched electrodes, as shown in Fig. 3a. For determining position in the y-direction, rectangular electrodes were divided diagonally into two sets of isolated electrodes. The ratio of charge collected by each electrode is proportional to the location of the charge cloud in the y-direction. Interdigitiated between these electrodes, another set of complementary rectangular electrodes was placed. These electrodes varied in width across the array, such that the ratio of the two gave a value proportional to the x-displacement of the charge cloud. The Siegmund device eliminated one of these electrodes (see Fig. 3b). The first electrode was a comb-shaped electrode with rectangular fingers that increased in width across the array. The second electrode was interdigitated between the first with uniform triangular-shaped electrodes. Since these two electrodes do not match each other exactly, a third electrode occupied the space in between. The ratio of the signal on the rectangular electrode to the total signal (sum of the three electrodes) gives the x-location of the charge cloud, while the ratio of the signal on the triangular electrode to the total signal gives the y-location.

One of the benefits of these detectors is their relative simplicity. Only three sensors are needed to determine the spatial location of a detector event. Measurement can be made with spatial resolution better than 20 μm [93,94]. As with the aforementioned detectors, simultaneous detector events are, however, problematic.

Another implementation of the shaped-anode array has become known as the coded anode coincidence array (CODACON) [95]. In this type of array, the detector elements are divided in such a way that the output of the device is a binary number corresponding to a location on the array. A series of electrodes, as shown in Fig. 4, termed “charge spreaders,” intercept the electron cloud produced by the MCP. These electrodes are capacitively coupled to the underlying coded anodes. With the CODACON, N electrode pairs will provide 2^N resolution elements. For example, if a 10-mm device consisted of eight coded anode pairs, there would be a total of 256 resolution elements, or a theoretical spatial resolution of approximately 40 μm. Each electrode pair is monitored by a comparator circuit. Depending on the charge on each electrode, the comparator will output a “1” or “0”. The series of comparator outputs can be read as a binary number whose value is the address of a particular resolution element.

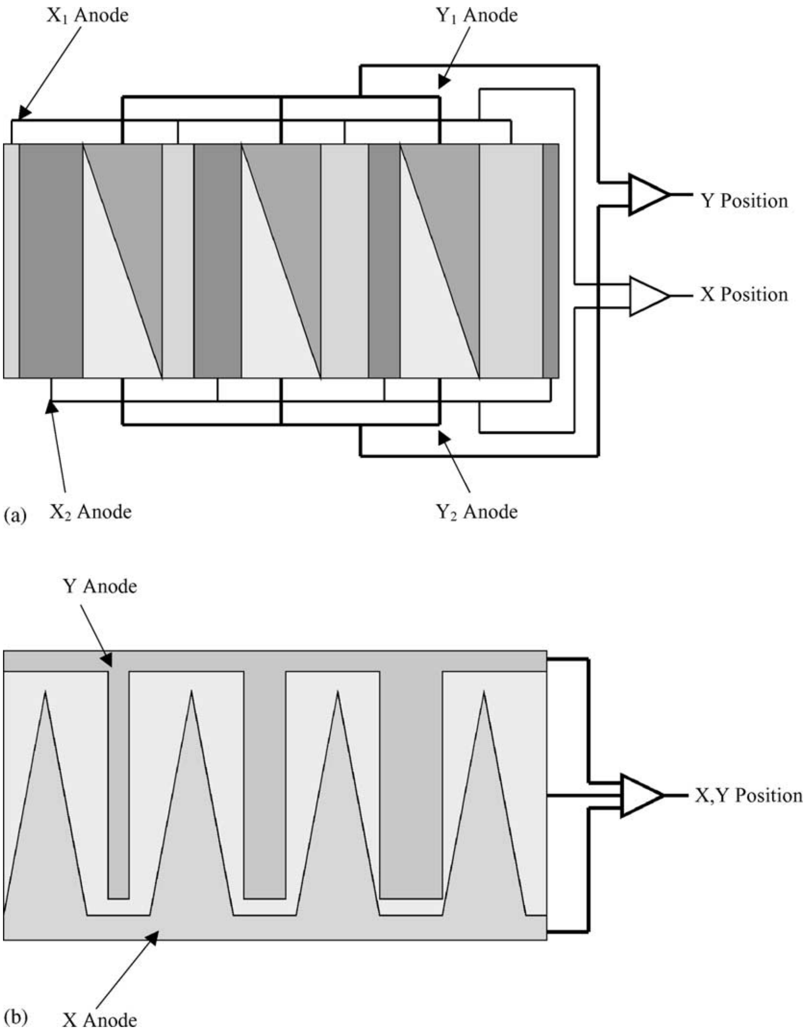

Fig. 3. Shaped anode array detector geometries. The detector surface can be divided into either a four-electrode system (a) or a three-electrode system (b).

The greatest benefit of the CODACON is that its resolving power is not dependent on the speed or precision of the readout electronics. The anodes, themselves, interpret the position of the charge cloud. This type of device is more complicated than the previously described ones and requires more electrical components for its implementation. Additionally, the resolving power will be limited by the ability to fabricate the small features needed for the electrodes.

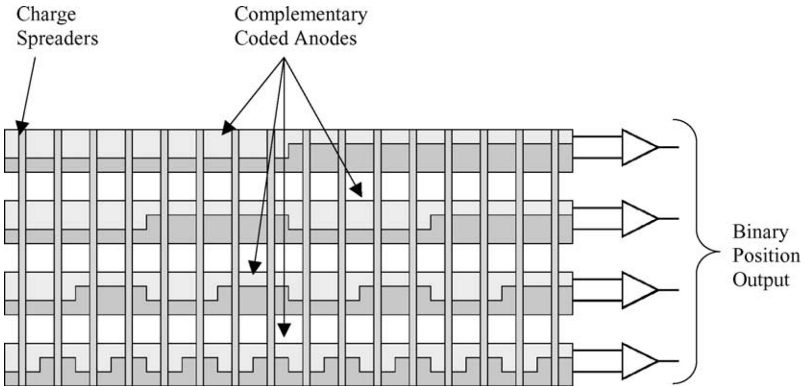

Fig. 4. The CODACON shaped anode array detector utilizes electrode pairs to convert the spatial location of an ion into a binary number.

5.3. Capacitively coupled array detectors

Capacitive detection devices are hybrid arrangements that can overcome some of the problems of charge-division detectors by indirectly measuring the effects of a MCP electron cloud [96,97]. For this type of detection, typically a resistive anode is used to intercept the charge cloud from the MCP. Situated a short distance behind the resistive anode is another position-sensitive detector such as a wedge-and-strip or delay-line device. Due to the dielectric of the medium between the two detectors, the arrival of a charge cloud at the resistive anode can induce a current in the second detector. By this means, the position-sensitive detector can be electrically or optically isolated from the measurement device.

The isolation of the detector from the instrument has several advantages. The detector can be placed outside the vacuum system, allowing for a wider selection of fabrication materials and isolation from sometimes harsh instrument environments. Often, detectors are at high electrical potentials, necessitating operation of the readout electronics at high voltage. This introduces safety hazards as well as high-voltage instability and noise sources. Since this type of detector is electrically isolated from the rest of the instrument, it can be operated with conventional low-voltage circuitry. One of the main limitations of the capacitive detector is that the additional conversion step slightly reduces the spatial resolution of the measurement. Moreover, this detector will suffer from the limitations of both the primary and capacitively coupled detectors.

5.4. Discrete-anode array detector

The final type of detection system commonly used with MCP detectors employs an array of individual anodes [98]. This method is the MCP-array equivalent of the multiple-detector method described earlier. An array of anodes, each with its own charge-sensing electronics, is used to locate the position of the electron cloud produced by the MCP. Much of the initial research with discrete anodes involved using macroscopic-current measurement equipment in order to record the signal on each detector element. Recently, arrays have been fabricated with all controlling electronics included in an integrated package [99–103]. An additional method for implementing this technology is to directly deposit the anodes on the back of the MCP [104].

These devices have many advantages. The detector elements can be fabricated with widths of tens of micrometers, leading to high spatial resolution [99,101]. Additionally, they can be mass produced inexpensively, with a high level of reproducibility. Since the detector elements are interrogated independently of each other, simultaneous detector events can be recorded accurately without any loss of spatial resolution. Typically, these devices are limited by the maximum count rate of the MCP. A limiting factor in the widespread use of this type of detector is the multiplicity of detector components needed. Each anode needs its own current-sensing circuit, which can add cost.

With all of the aforementioned MCP-based position-sensitive detection schemes, the user has a wide selection of available technologies to suit the need of an experiment. Spatial resolution as small as 10 μm is possible with several of the arrays. With the MCP, the detection of single events is possible when a chevron or z-stack configuration is used and, as with the discrete anode arrays, simultaneous events can be monitored, leading to a broad dynamic range. These arrays can be decoupled from the instrument so the controlling electronics can be operated under conditions different from the instrument. The most severe limitation placed on these arrays arises from the use of an MCP to convert the particles of interest into electrons. The use of a single MCP, and to a greater extent, chevron and z-stack MCP arrays, adds uncertainty into the detected position of the particle. Although the majority of secondary electrons produced when a particle strikes the MCP stay in the channel from which they originated, many leave the channel and enter adjacent channels. Similarly, when the electron cloud leaves the first MCP in a chevron stack, it spreads out over several channels in the second MCP. It is possible to use mathematical corrections to minimize the uncertainty this process introduces. Nevertheless, position errors such as this can only be eliminated by removing the MCP from the detector array.

6. Integrated array systems

Recently, several new detector arrays have been developed that eliminate the need for electron multipliers or MCP arrays [105–108]. All of these detectors rely on electronic circuitry capable of sensing extremely low current levels. As early as 1984, it was speculated that the advancing semiconductor technology would allow a detector array to be fabricated with all the necessary electronics combined into a chip-type package [109]. This feat can be accomplished by employing circuitry that measures the voltage produced by the charging of a capacitor. With the inverse relationship between capacitance and the voltage a charge will produce, coupled with the ability to fabricate extremely low-valued capacitors on silicon, the technology exists to detect extremely low numbers of charges.

Several advantages derive from removing the multiplying stage from the detector. The cost will drop since the multiplier is often the most expensive component in a position-sensitive detector. Additionally, the need for high voltages for the multiplier is eliminated, leading to a safer, more stable detector. Finally, there will not be strict reduced-pressure requirements as with electron multipliers. Four of these detectors will be highlighted below: a Faraday cup array with either MOSFET or CCD readout, an active pixel sensor (APS) device based on CMOS technology, and two arrays of Faraday cups with capacitive trans-impedance amplification.

6.1. *Tracy detector array*

The detector array developed by Tracy et al. [105] at the Perkin-Elmer Corporation is one of the first examples of forgoing a multiplier stage and relying on the advances in semiconductor technology to directly measure the current produced by an ion beam. This detector contains of an array of conductive metal Faraday cups electrically isolated from each other and a silicon substrate. Each Faraday cup is connected to a MOSFET circuit, whose purpose is to maintain a predetermined voltage on the Faraday cup. When a charge arrives at the Faraday cup, the voltage is changed, causing the MOSFET circuit to react. This procedure could also be performed by connecting each Faraday cup to a pixel on a CCD. Although no specific details are given on the operation of the detector array, this signifies a change in thinking for position-sensitive measurements.

6.2. *Active pixel sensor (APS) array*

The term APS is derived from the fact that each pixel in the detector array has its own accompanying amplifier, as opposed to passive devices such as CCDs, which have a single amplifier for many pixels. Active pixel sensor arrays were developed primarily for use in photon detection, although ion detection can be performed just as simply. In a conventional photon APS, a photosensitive material is connected to the input gate of a FET. The arrival of a photon at the pixel creates an electron-hole pair, which is subsequently detected by the FET. To modify this device for charge detection, the photosensitive material is replaced with a conductive surface (i.e. Faraday cup), such that the charge of any arriving ions is directly transmitted to the FET gate. This type of detector responds to changes in the gate voltage, which is inversely proportional to the capacitance between the Faraday cup and ground. It is possible to have capacitances in the femtofarad regime, leading to microvolt changes for the arrival of a single charge.

Several such APS arrays have been developed for mass spectrometric detection at the Jet Propulsion Laboratory [110]. The one of particular interest to SFMS is a linear array of 128 Faraday cups, each 16 μm wide, 1600 μm long and on 20 μm centers. This array was mounted on a miniature Mattauch-Herzog mass spectrograph and used for the detection of atmospheric gases. The read noise for a single data acquisition was on the order of 400 e^-, while averaging several acquisitions brought the noise below 100 e^-. This detector was also successfully operated at elevated pressures. Moreover, a dynamic range of 10^4 was reported. One disadvantage of the array is that the detector elements are interrogated in a serial manner, rather than simultaneously.

6.3. *Micromachined Faraday cup array*

The micromachined Faraday cup array (MFCA) is an assembly of well-shaped Faraday cups connected to a multiplexer and interrogated by an integrating operational amplifier. A portable Mattauch-Herzog mass spectrometer is available commercially with this type of detector array [111]. Initial development of the MFCA was reported in 2002, with the construction of two arrays, one consisting of a linear array of 256 detector elements, each 0.7 mm × 5 mm, while the other was a two-dimensional array of 64 circular detectors, each 2.03 mm in diameter [112]. The subsequent generation of this device consists of a linear array of between 64 and 256 Faraday cups with widths of 150 or 250 μm [113]. The later generation has been fabricated using deep reactive ion etching (DRIE), which allows for smaller detector elements and highly vertical cup walls.

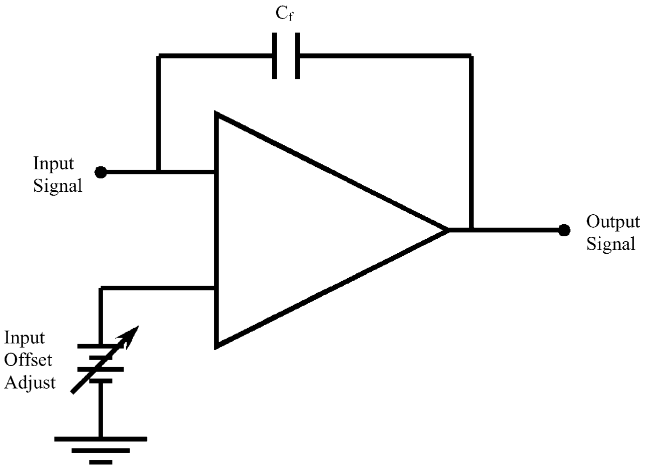

Fig. 5. Schematic diagram of the integrator circuit used in the micromachined Faraday cup array (MFCA) detector.

With all of these arrays, the ion signal captured by the Faraday cups was transferred to an integrating circuit through a multiplexer. In this geometry, each detector element is read out individually with the same circuitry, a benefit that offers a high degree of linearity. The integrating circuit, as shown in Fig. 5, has been implemented with operational amplifier technology, and with this particular geometry, the output voltage (V_o) is inversely proportional to the size of the feedback capacitor (C_f). Therefore, the gain of the device can be adjusted simply by modifying the capacitance. The circuitry reported for the MFCA used a 100 pF capacitor, leading to a several nanovolt change in the integrator output for every charge arriving at a Faraday cup. This corresponds to an ability to detect ion beam currents as low as single picoamps.

The MFCA has numerous attractive features. Detection of simultaneous events is possible, since the Faraday cups serve to hold the ion signal until integration. As mentioned previously, only one integrating circuit is used, so the array has a high degree of linearity. Another benefit of employing only one integrator circuit is the limited number of vacuum feed-throughs that are needed. With DRIE fabrication, the individual detector elements can be made on the order of tens of microns, which offers high mass-spectral resolution. Moreover, the well-shaped collectors produced by DRIE are extremely efficient at capturing ions and preventing cross-talk between adjacent detector elements. Finally, this device does not need high voltage or power.

Several limitations of the MFCA do exist. Although it can operate with unit duty cycle, it is a scanned system. The pixels are interrogated sequentially; therefore, any correlated noise sources affecting the multiplexer, integrator, or signal processing electronics cannot be removed through ratioing. An additional limitation is the inability to detect single ion events, which will hinder the use of the detector for trace analyses.

6.4. *Focal plane camera*

The focal plane camera (FPC) was developed in a collaboration among Pacific Northwest National Laboratory, the University of Arizona, and Indiana University [108]. As shown in Fig. 6, the detector consists of an array of gold Faraday strips, each 145 μm × 5 mm, with each cup electrically isolated from the others and connected to its own integrating circuit. In between each of the Faraday strip is a 30 μm space that incorporates a 10 μm grounded wire. The array of Faraday strip/integrators are processed with a multiplexer and the signal is recorded with a personal computer. The detector array allows for either destructive or non-destructive readout. With non-destructive readout, the integrated signal can be recorded multiple time to reduce read noise. The gold Faraday strips were formed with conventional lithographic techniques on a glass substrate and wire-bonded to integrating circuits similar to that pictured in Fig. 5. In this particular integrator, termed a charge-transimpediance amplifier (CTIA), originally intended for low-noise IR astronomy use, the feedback capacitor is 36 fF, leading to an output voltage of 4.4 μV per ion. In order to reduce read noise, the detector is cooled to 233 K. The first generation of the FPC contained 32 Faraday cup/integrator units, though only 31 were used for experimentation. Subsequent generations of the FPC will have a much greater number of Faraday cup/integrator units, as well as narrower cups and faster readout electronics.

A recent study demonstrated the utility of the FPC by coupling it to a Mattauch-Herzog mass spectrometer with both glow discharge and inductively coupled plasma ionization sources [114,115]. Limits of detection were comparable to those obtainable with an electron multiplier on the same instrument. A dynamic range of 10^6 was demonstrated for glow discharge, though this was limited in linearity by the availability of standards, while over 10^7 was demonstrated with an ICP source.

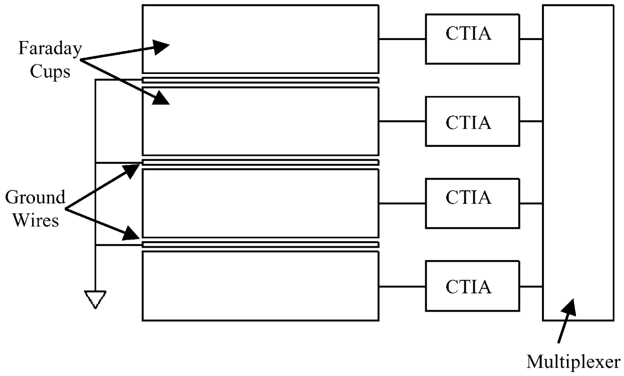

Fig. 6. Schematic diagram of the focal plane camera.

The FPC has also been recently used with an atmospheric-pressure ion mobility spectrometer (IMS) for the detection of explosives and chemical-warfare agents [116]. This coupling demonstrated the ability of the FPC to increase the measurement sensitivity of IMS by 10^3 over conventional Faraday-plate detection. This performance should be further enhanced with subsequent generations of the FPC designed to operate at elevated temperatures and atmospheric pressure.

The FPC is of great utility due to several attractive features. This technology can detect tens of ions in its current form, and with subsequent generations, this will be improved to single-ion detection [108]. Additionally, current lithographic technology will allow structures as small as single micrometers to be patterned on a substrate, expanding high-resolution applications. The integrating electronics of the device interrogate all detector elements simultaneously, which can remove correlated noise. The FPC will also benefit from all of the advantages of Faraday-cup detection, such as robustness, uniform response, and no or low-voltage requirements. Finally, the FPC is completely computer-controlled and easy to operate. Two of the current limitations of the FPC performance are its readout rate, currently only 1 kHz, and its detector element size. Both of these issues will be addressed in future generations of the detector.

7. Conclusions

The technology available for simultaneous particle detection has made great advances over the past 90 years. From its very rudimentary beginnings, the detector array has advanced from a time-intensive, low sensitivity, qualitative device to a computer-controlled, high speed tool capable of detecting single ions or photons. The ever-advancing fabrication technology used to make these devices has allowed them to be produced smaller, less expensively and with a higher degree of precision. Though many of the current position-sensitive detectors incorporate MCP detectors to provide high sensitivity, there is a trend towards utilizing devices with integrated detector elements and controlling electronics due to their simplicity and ability to provide comparable results.

Acknowledgments

Support to the authors has been provided by the U.S. Department of Energy, Office of Nonproliferation Research and Engineering. Pacific Northwest National Laboratory is operated by Battelle Memorial Institute for the Department of Energy under contract DE-AC06-76RLO-1830.

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