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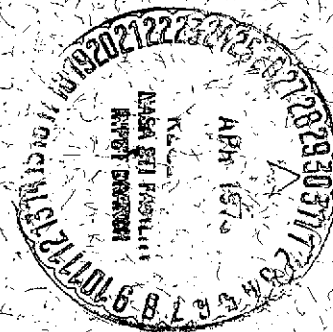
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A COMPOSITION ANALYZER FOR MICROPARTICLES

USING A SPARK ION SOURCE

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ABSTRACT

Iron microparticles were fired onto a capacitor-type microparticle detector which responded to an impact with a spark discharge. Ion currents were extracted from the spark and analyzed in a time-of-flight mass spectrometer. The mass spectra showed the elements of both detector and particle materials. The total extracted ion current was typically 10 A within a period of 100 ns, indicating very efficient vaporization of the particle and ionization of the vapor. Potential applications include research on cosmic dust, atmospheric aerosols and cloud droplets, particles ejected by rocket or jet engines, by machining processes, or by nuclear bomb explosions.

INTRODUCTION

Analyzing the chemical composition of cosmic dust particles is presently one of the most attractive goals of the physics of interplanetary and interstellar matter, particularly of cometary physics. A prior composition analyzer (Auer and Sitte, 1968; Auer, 1971; Friichtenicht et al., 1973; Dietzel et al., 1973) utilizes the plasma that is emitted when a dust particle impacts upon a solid surface. Ions are extracted from the plasma and analyzed in a time-of-flight mass spectrometer with respect to their charge-to-mass ratios. That analyzer has one major disadvantage resulting from its low overall efficiency: the proportions of ions observed in the mass spectrum are usually very much different from the proportions of elements in the original particle. For example, alkali metals are overrepresented whenever the impact velocities are below approximately 20 km/s, whereas an element having a high electron work function like carbon may not be observable at all even though the original particle consisted of pure carbon (Dietzel et al., 1973; Drapatz and Michel, 1974).

A new method is described in this paper which makes use of the fact that a capacitor-type microparticle detector very efficiently converts an impacting microparticle to vapor and the vapor molecules, in turn, to ions. The conversion occurs when the electric energy stored on the detector (typically of the order of 1000 erg) is discharged through a spark, the spark being triggered by the release of kinetic energy (typically of the order of 0.1 erg) of the impacting microparticle (Auer, 1974).

CONSTRUCTION AND SENSITIVITY OF A CAPACITOR-TYPE

MICROPARTICLE DETECTOR

Capacitor-type microparticle detectors have been used on many spacecraft such as Mariner IV, Pegasus I-III, and Explorer 46, to count particle impacts. A very reliable metal-oxide-silicon (MOS) version is described by Kassel (1973). Its construction is schematically shown in Figure 1. The detector is made from a 51 mm-diameter, 0.5 mm thick wafer of low resistivity, p-type (boron doped) silicon which forms the inner electrode of the capacitor. A 0.4μm or 1μm thick layer of silicon dioxide is grown on the wafer by thermal oxidation to form the dielectric of the capacitor. A 0.1μm thick aluminum coating is then vapor deposited on the SiO_2 to form the outer electrode. An electric field with a strength of the order of 10^6 V/cm is applied across the SiO_2 dielectric. An impacting particle triggers a spark between the electrodes, giving rise to a voltage drop across the capacitor that can readily be detected and counted. The spark is accompanied by an audible crack and a visible flash. According to Kassel (1973), the sensitivity of the MOS detector can be determined by the relationship d = $2.81\, v^{-2}$ ($m^3\, s^{-2}$), where d is the diameter of an iron particle and v is particle velocity (1.1 km/s $\leqslant$ v $\leqslant 2.0$ km/s). For example, an MOS detector with an SiO_2 thickness of 0.4μm will respond to a particle with a diameter of 0.7μm (mass of 1.3×10^{-12} grams) or larger and a velocity of 2 km/s. Brooks et al. (1974) have calibrated MOS detectors with relatively slow particles having velocities between approximately 12 m/s and 110 m/s, and also tested them

successfully as atmospheric dust particle counters on aircraft. They expressed the sensitivity by the relationship $m = 3.8 \times 10^{-8} v^{-1.85}$, where m is mass in gram and v is velocity in m/s. For example, a particle with a velocity of 12 m/s and a mass m of 1.3×10^{-9} g or greater would trigger a spark discharge on the most sensitive detector.

Because this paper proposes to utilize the spark discharge of the capacitor detector as a spark ion source, let us summarize the major advantages and disadvantages of this type of ion source (see, e.g., Inghram et al., 1954). The advantages are:

1. A spark ion source ionizes any element present in the sample, both volatile and involatile substances;

2. It has no blind spots, i.e., all elements ionize with equal efficiency within one order of magnitude;

3. There is little fractionation of elements.

A major disadvantage is the fact that large spreads of energy are always present and consequently, recourse to double focusing mass analyzers is necessary. Another disadvantage is the flow of high currents with short rise times in the spark which can result in conducted and radiated electromagnetic interference with other equipment. Both disadvantages can be met with carefully designed instrumentation.

UNFOCUSED ANALYZER

Experiments were carried out with iron microspheres from the hypervelocity dust accelerator at the Goddard Space Flight Center. Most particles had diameters of several μm and velocities of several km/s. An MOS capacitor-type micrometeoroid detector, shown schematically as part of Figure 1, and kindly provided by J. Alvarez and P. Kassel of the NASA Langley Research Center, was exposed to impacts of dust particles, one at a time. Many, but not all particles triggered a spark. As shown in Figure 1, the polarity of the bias voltage on the detector was such that ions were accelerated from the back to the front electrode. An additional electric field was applied by means of a grid in front of the detector in order to extract ions from the spark and to accelerate them further (total acceleration voltage U = 265 volts). Some ions hit this grid, giving rise to a short and intense current pulse (about 100 ns wide, 1 Ampere peak current) which provided the zero-time reference for a time-of-flight mass spectrum to be recorded on an oscilloscope. Other ions passed through the grid, were deflected in a transverse electric field, and detected at a distance l = 47 cm from the detector. The deflection was necessary in order to prevent neutral ejecta from hitting the ion collector. Note that this very simple arrangement has all essential parts of a time-of-flight mass spectrometer, ion source, accelerating field, drift space, and ion detector. Correspondingly, the current flowing onto the ion detector and being measured on an oscilloscope as a function of time represented a time-of-flight mass spectrum of the ions that were extracted from the spark.

A spectrum is shown in Figure 2a. A large peak can be seen around mass numbers 27/28, probably due to aluminum and/or silicon. Another large peak around mass number 56 is probably due to iron. Oxygen is only weakly indicated if at all. Notice the absence of alkali metal peaks which have always presented a severe problem, as stated earlier in this paper, in other composition analyzers. The first large signal should be disregarded. It was caused by crosstalk between adjacent pins on the common vacuum feedthrough and essentially disappeared when different pins were used.

The total charge of extracted ions can be estimated from the grid current to be of the order of $10 A \times 10^{-7}$ s $= 10^{-6}$ As or 6×10^{12} elementary charges. This is at least 6 orders of magnitude more than the total charge of ions which are typically produced in prior type composition analyzers.

In order to make sure that the pulses were due to real ion currents, the accelerating voltage U was varied from 160 to 330 volts. As anticipated, the spectral lines appeared at times t_i following the relation

$$t_i \sim U^{1/2}$$

FOCUSED ANALYZER

Since the mass spectrometric resolution of the first model was relatively poor, a double-focusing principle designed by Oetjen et al. (1974) was applied. Hereby, ions drift first on a linear, second on a circular, and third on another linear path. In the circular section, ion paths are turned around by 164.4 degrees

by means of an electric field between two cylindrical plates. Ions having an energy e (U + ΔU) drift through the linear sections in a shorter time than ions having an average energy eU. In the circular section, however, their flight path is increased due to the larger radius of curvature r $\sim$ U + ΔU. The gain of time in the linear sections can be made equal to the loss of time in the circular section by properly selecting the lengths of each section; the radius of curvature of the second section; the aperture dimensions; and the electric potentials.

Although our instrumentation was mainly made from surplus materials costing less than 25 dollars, and far from being carefully designed, a relatively good resolution was achieved while keeping electromagnetic interference at a minimum. Figure 3 shows the arrangement used. In order to identify the dust particles, we placed a velocity vector analyzer, described elsewhere (Auer, 1975), in front of the composition analyzer. The first and third drift sections of the composition analyzer were field-free regions, each approximately 200 mm long. The third section was shielded by a grounded metal cylinder in order to prevent charged or uncharged particles originating in the spark ion source from interfering either directly, or through secondary particle emission with the ion detector. The second drift section was made of two concentric, conductive hemispheres mounted on, but electrically isolated from, an aluminum flange. The mean radius of curvature was r_0 = 95 mm. The separation between the hemispheres was d = 31 mm. The two apertures at the beginning and the end of the

circular section were round holes, 8 mm in diameter and covered with a metallic grid. According to Oetjen et al., the ion paths should be turned around by 164. 4° in the circular section. Preliminarily and for simplicity, we selected 180°, thereby sacrificing optimum focusing properties for simple and inexpensive construction. The ion detector was an aluminum plate fitted with aluminum honeycombs on top to reduce secondary emission effects, and placed in a grounded metal shield.

In order to properly adjust the potentials, a heated tungsten wire was placed at the position of the spark ion source, to emit a constant current of electrons with energy eU toward the first aperture of the hemispherical deflector. The outer hemisphere was connected to a negative voltage and the inner hemisphere to a positive voltage. The electron current from the ion detector was measured with an electrometer. The voltages were adjusted for maximum electron transmission. Then the polarities of all bias voltages were reversed and the heated tungsten wire was used as an ion emitter. The voltages needed no major readjustment for maximum ion transmission. This ensured us that the earth's magnetic field had no significant effect on the ion focusing properties of our configuration. Typical voltages were: for acceleration U_a = +329 V, on outer hemisphere U_{oh} = +170 V, on inner hemisphere U_{ih} = -152 V. No effort was made to focus the electrons or ions over the first linear drift section into the aperture of the hemispherical deflector. Most of the ions hit the aluminum flange so that only

a small fraction could enter the analyzer system. The heated tungsten wire was then replaced by a capacitor-type detector as a spark ion source. For the following experiments, we used a different capacitor-type detector. Unfortunately, the gold-wire contacts of all previously used detectors were damaged due to the unexpectedly high currents involved. While the previously used MOS detectors were manufactured from extremely pure materials under well-controlled environmental conditions, the presently used emergency detectors were crudely fabricated 12 years earlier for an OGO project. During that period they were exposed to non-environmentally-controlled conditions and contaminants. Their electrical contacts, however, had been made by a technique which made them resistant against damage from high currents. Such a detector was reconditioned and used in connection with the arrangement of Figure 3. Figure 2b shows a mass spectrum obtained from the impact of an iron microparticle. A large number of overlapping lines can be seen at atomic mass numbers below about 26 amu. Such lines have not been observed in spectra obtained from the MOS detector used earlier. We believe that these lines represent mainly impurities and contaminants such as hydrocarbons from oil layers on the surface of the detector. In addition, two narrow lines can be seen: one around 28 amu (probably Si^+) and one around 56 amu (probably Fe^+). The resolution appears adequate to separate nickel (59 amu) from iron (56 amu).

DISCUSSION

It is certainly possible to further improve the mass resolution of this analyzer, by optimizing the instrumental dimensions and by selecting a deflection angle

of 164. 4° instead of 180°. Oetjen and Poschenrieder (1974) have actually re-
solved the isotopes of xenon (128-136 amu) from an ion current with an energy
spread from 475 to 525 eV, using an instrument that was twice as large and op-
timally focused and dimensioned. By means of a spiratron (Bakker, 1971), an
extremely high resolution ($m/\Delta m = 630$) should be achievable.

Note that the electric fields employed are static; thus, the focusing charac-
teristics are independent of ion flight times, and light elements are focused as
well as heavy elements. Simultaneous focusing of all elements is not possible
with conventional time-lag focusing time-of-flight mass spectrometers (Dietzel
et al., 1973).

The major difficulty that needs additional research is the presence of ions
from the capacitor detector material in the spectrum. It must be concluded that
a detector should not be made from silicon and silicon dioxide if the measure-
ment of the abundance of silicon in an unknown particle (such as a micrometeoroid)
is a major objective. Further tests with different materials have to be done in
order to find adequate conductors (e.g., gold) and dielectrics which are neither
abundant nor important in the composition of the particle to be investigated. For
similar reasons, impurities in the material must be kept to the lowest possible
concentration, and the detector must be held in a clean and oil-free environment.

Further work must also include an investigation of the ionization efficiency
as a function of particle velocity, mass, and composition, and of the composition

and thickness of the detector. We expect, however, that this investigation is not particularly difficult because of the established high efficiency of spark ion sources for all elements (Inghram et al., 1954).

POTENTIAL APPLICATIONS

The ideal detector for dust particles should measure the velocity vector, mass, charge, size, shape, and chemical and isotopic composition of each individual particle. We believe that an instrument based on the development work described above will come very close to this goal, particularly if combined with a velocity vector analyzer described elsewhere (Auer, 1975). The high sensitivity of a capacitor-type microparticle detector should make it possible to extend the measurements to particle masses and velocities substantially below those attained hitherto.

The low velocity threshold becomes particularly important on a comet rendezvous mission, where particle encounter velocities can be as low as 10-100 m/s. Particles in that velocity range and with masses of 10^{-9} g or greater have sufficient energy to trigger a spark discharge.

Since the instrument measures composition instantaneously, ices or other volatiles—most interesting on a cometary probe, but also on a planetary entry probe—cannot escape detection. Similarly, occluded gases can be detected and analyzed. Similar detection and analysis would be difficult, if feasible, by means of a particle collector and an electron microprobe.

The high ionization efficiency of a spark ion source is particularly useful on a cometary probe, since many elements such as carbon (which may be abundant in comets), are not detectable with prior composition analyzers (Dietzel et al., 1973). For similar reasons, the method should be most efficient in measuring interplanetary and interstellar (graphite?) dust particles.

The successful application of capacitor-type detectors on aircraft for aerosol particle and cloud droplet counting has been mentioned earlier. The detection principle can be extended to planetary atmospheres. In order to not only detect such particles but to analyze their composition as well, it will be necessary to employ a thin-film, self-supporting capacitor-type detector. Detectors of this type were used on the NASA RMS and MTS satellite meteoroid experiments in near-earth orbits. A detector in this application will have to serve not only as a spark ion source but also as a gas barrier to maintain a differential pressure between the atmosphere and the vacuum system of the mass spectrometer. A $0.3\mu m$ thin film has already been used successfully as a gas barrier in a micrometeoroid experiment in the upper atmosphere by Auer et al. (1970).

Solid particles polluting our air can be analyzed by pumping air through an apparatus such as the one used by Brooks et al. (1974) to calibrate MOS detectors. By means of an air filtering system, Sittkus (1964) has found and analyzed a single microparticle from a cloud of fission products produced in the explosion of a nuclear bomb. From the radioactivity and the time constants of the radioactive

decay, he was able to determine the type, strength, and date of the explosion. Utilizing a composition analyzer should provide even more information on similar particles and not be limited to the investigation of highly radioactive particles. Indeed, various other types of particles could be analyzed and traced back to their respective sources, such as volcano eruptions or industrial processes. While Sittkus' findings about one single particle were the result of several years of research, a similar investigation could feasibly be performed automatically and with a high degree of efficiency and reliability by means of a composition analyzer.

A composition analyzer placed near the exhaust of a rocket or jet engine should yield the composition of ejected solid particles. In a similar way it should be possible to analyze solid particles emitted during machining processes.

We can estimate the method's sensitivity for a single ion species by making the following assumptions:

- The mass spectroscopic resolution is adequate to separate and group all ions of interest according to their e/m ratios.

- Groups of ion species of lesser abundance will be individually extracted from the focused beam and injected into electron multipliers (Damoth, 1964) for high amplification or for counting of individual ions of a species.

- The ionization efficiency of the spark discharge is approximately equal for all elements.

- The total number of ions produced in a spark is 6×10^{12}, as mentioned earlier.

- At least 1% of all ions, or 6×10^{10} ions, are from the projectile material, while 99% or less are from the detector material.

- By properly focusing the ions through the mass spectrometer, the overall transmission will be nearly 100%. Focusing can be achieved, for example, by employing a curved detector (Auer, 1971) and a large aperture hemispherical electrostatic analyzer (Chase, 1973).

The resulting maximum possible sensitivity will be approximately 1 atom of a single species in 6×10^{10} atoms of the dust particle.

Such a high sensitivity should make it possible to measure extremely small concentrations of elements or isotopes in extremely small dust particles. Let us consider, for example, the heavy noble gases in an average dust grain on the lunar surface. A grain with a diameter of less than $5 \mu m$ contains only 4.0×10^{-7} ccm STP/g of Kr^{84} and 1.3×10^{-7} ccm STP/g of Xe^{132}, according to Hintenberger et al. (1974). The concentration can be expressed as $1 Kr^{84}$ atom or $1 Xe^{132}$ atom per 1.9×10^9 or per 5.8×10^9 atoms, respectively; in terms

of total number of atoms, a dust particle having a diameter of $4\,\mu m$, for example, contains approximately $1100\,Kr^{84}$ atoms and $350\,Xe^{132}$ atoms. Both concentration and number of atoms of Krypton and Xenon lie well above the estimated sensitivity threshold of the method.

ACKNOWLEDGMENT

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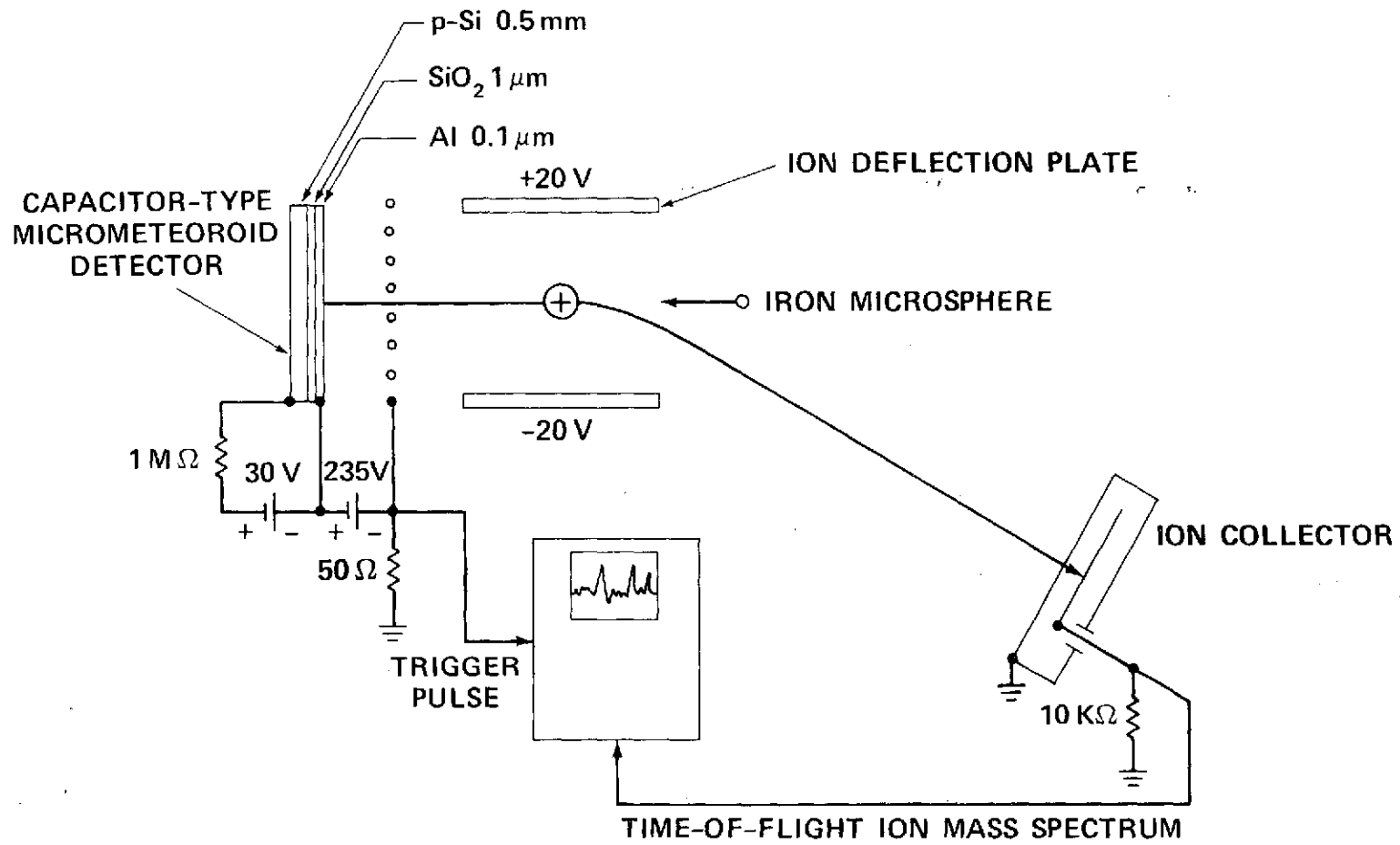
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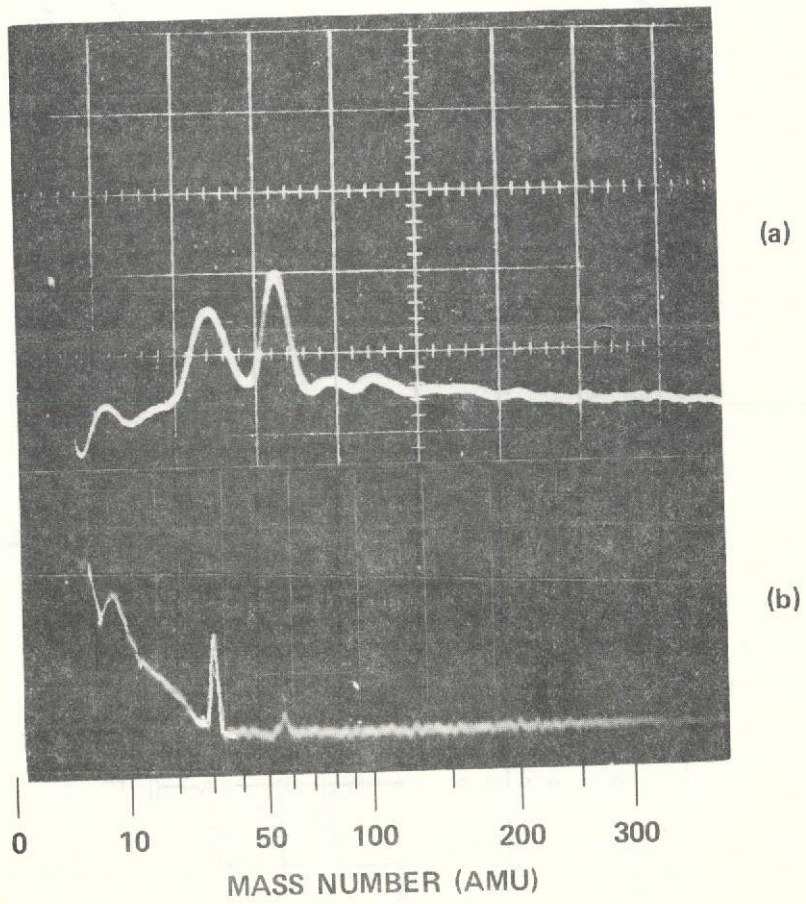
FIGURE CAPTIONS

Figure 1. Schematic Diagram of Unfocused Instrument

Figure 2. Ion Time-of-Flight Mass Spectra Taken with Unfocused (a) and

Focused Instrument (b), respectively.

Figure 3. Schematic Diagram of Focused Instrument.

(a)

(b)

0   10   50   100   200   300

MASS NUMBER (AMU)