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**AEC Research and
Development Report**

**Knolls
Atomic Power
Laboratory**

A Three-Stage Research Mass Spectrometer

***F. A. White
F. M. Rourke
J. C. Sheffield
May 28, 1958***

**Operated for the
United States Atomic
Energy Commission by
GENERAL ELECTRIC**

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UC-37, Instrumentation
(T1D-4500, 13th ed)

General Electric Company
KNOLLS ATOMIC POWER LABORATORY
Schenectady, New York

A THREE-STAGE RESEARCH MASS SPECTROMETER

F. A. White
F. M. Rourke
J. C. Sheffield

May 28, 1958

Authorized Classifier

Date

Operated for the
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ABSTRACT

A three-stage mass spectrometer that utilizes two consecutive magnetic focusing lenses of 20-in. radii followed by a 20-in. radius-of-curvature electrostatic analyzer is described. An abundance sensitivity of 10^{10} was measured for this instrument in the low mass region. Application of this instrument to analytical and research problems in the field of mass spectrometry and the results obtained are discussed.

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A THREE-STAGE RESEARCH MASS-SPECTROMETER

F. A. White, F. M. Rourke, and J. C. Sheffield

INTRODUCTION

The design and construction of the instrument described herein were undertaken after a thorough appraisal of current and anticipated problems associated with isotopic ratio measurements. The limitations in isotopic ratio determinations fall into two broad categories: (1) those inherent in a specific experiment, such as the difficulty of producing a sufficiently copious number of ions from a metal with a high boiling point; and (2) fundamental restrictions that impose physical limits of observation and detection, i.e., boundary conditions such as the lower limit of detection of a singly charged particle of a given kinetic energy, or the resolution of various ions with very small differences in momenta. All "signal-to-noise" limitations could be included in this group. It is apparent that overlapping exists between the so-called practical restrictions of a particular experiment and limitations that are more basic and of broader scope.

A brief review of the latter group of problems is presented and the experimental results obtained with the three-stage research mass spectrometer are submitted as a progress report in the field of mass spectrometric instrumentation. Many photographs are included in response to the many inquiries about mass spectrometric instrumentation at this laboratory.

No brief is made for the mechanical design of the components of the instrument; some methods and techniques are simply an outgrowth of particular experience and prejudice in assembling laboratory-type spectrometers. On the other hand, economical construction and flexibility of adaptation for a wide variety of investigations were well-defined prerequisites.

ISOTOPIC PROBLEMS AND OBJECTIVES

The following aspects of isotopic abundance measurements were either actively explored or investigated as potential projects:

1. Nuclear research
 - a. Measurement of cross sections
 - b. Fission fields
 - c. Higher-order reactions

2. Reactor research
 - a. Fuel, structural material, and coolant
 - b. Irradiation damage and burnup measurements
 - c. Power distribution and energy spectrum
3. Solid state
 - a. Identification of semiconductor impurities
 - b. Quantitative "doping" of crystals
 - c. Use of p-n junctions or crystal counters for positive ion detection
4. Search for new naturally occurring isotopes
 - a. Identification and isotopic ratios of new nuclides
 - b. Establishment of new experimental upper limits
5. Studies of the penetration of heavy ions in matter
 - a. Balance between electron capture and loss for low-velocity ions
 - b. Excitation phenomena
 - c. Energy degradation measurements

It is clear that the above-mentioned represent only a fraction of the total number of investigations which rely, in part, on mass-spectrometric measurements. They have, however, affected the objectives of instrument development and, therefore, the specific objectives in designing the new machine were:

1. High abundance sensitivity
2. High isotopic sensitivity
3. Simultaneous detection of ion beams
4. Differentiation between atomic and complex ions with the same charge-to-mass ratio.

To a certain degree, a contribution has been made to the first three of these objectives, and work is in progress on the last and perhaps the most interesting facet of the work. The four objectives of instrument development are discussed below.

Abundance Sensitivity

Abundance sensitivity is defined here as the reciprocal of the fractional number of positive ions of mass M which are detected one mass unit distance (i.e. at the mass $M \pm 1$ position in the detector focal plane). Thus defined, abundance sensitivity is usually mass dependent, increasing with decreasing mass, but it has only a secondary relationship to resolution as it is customarily defined. It is, rather, an over-all figure of merit that indicates the lower limit of contribution to "noise" from an adjacent isotope.

In applications where it is desirable to search for isotopes whose relative abundance is small, high abundance sensitivity is important. Nearly all of the early isotopic abundance work has been limited to measurements in which the ratio of the more abundant to less abundant isotopes is $\sim 10^4$ or less. The advent of current research problems makes an improvement imperative. With the assumption of good ion optics, there are four important parameters that limit the measurement of ratios greater than 10^4 :

1. Dispersion
2. Finite source width
3. Energy spread of ions in source region
4. Small-angle elastic scattering of positive ions with molecules in the spectrometer tube,

In some instances, both the magnitude of the ion beam current and the range of detector sensitivity may also present a practical limit to the order of magnitude of the ratio that can be measured.

It has been shown, in work performed at KAPL, that scattering is one of the most important single limitations in abundance sensitivity. If F is the probability that an ion of mass M will be elastically scattered between the analyzing magnet and the detector slit, to a first approximation, this probability can be expressed as:¹

$$F = \frac{dx}{2\lambda} \left[\left(\log \frac{2S}{x} - \frac{2}{\pi} \right) \frac{\alpha S}{L} - \frac{1}{18} \left(\frac{\alpha S}{L} \right)^3 + \dots \right]$$

where L = distance between center of analyzer and exit slit

x = dispersion (for one mass unit)

$2S$ = exit slit height

λ = mean free path

dx = slit width

α = ratio of incident ion mass to scattering molecule mass.

Clearly, the scattering probability is only a slowly varying function of the dispersion. This immediately rules out as a solution the construction of a single magnetic analyzer with a large radius of curvature.

Consider, however, two magnets which are used as shown in Figure 1. Here, a single mass only is allowed to pass through an intermediate slit, (i.e., this exit slit serves as a virtual source for the second magnetic analyzer). In such a case, the probability that an ion of mass M will be scattered into the mass $M \pm 1$ position will be equal to the product of the two independent probabilities, F_1 and F_2 .

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Figure 1. Three-Stage Mass Spectrometer

In the case of two analyzers in tandem, F_1F_2 rather than F is the limitation on the abundance sensitivity that results from scattering. If the appropriate numbers are inserted in the above analytical expression (for usual operating pressures, etc.), a value of 2×10^{-4} is obtained for F for masses in the uranium region.

It is pointed out, however, that such a two-stage analyzer will yield a high abundance sensitivity only under several assumptions. The scattering must be elastic and the total energy spread of the ion beam must be small. That is to say, if there is a large distribution in energy of the positive ions as they leave the source, the directional focusing of the two magnetic analyzers alone cannot, of course, provide a sharp spectrum at the detector.

The advantages of two stages of directional focusing are, however, clearly apparent. Table 1 shows a few elements where upper limits on isotopic abundance have been measured. These measurements² have never been obtained in a single magnetic analyzer.

TABLE 1. Upper Limits of Isotopic Abundance

<u>Element</u>	<u>Mass No.</u>	<u>Upper Limit, % Abundance</u>
K	36	0.000008
	37	0.000008
	38	0.000008
	42	0.000004
	43	0.000002
In	109	0.000002
	110	0.000002
	111	0.000002
	112	0.000002
	114	0.000002
	116	0.000001
Cs	130	0.000002
	131	0.000001
	134	0.000005
	136	0.000003

Isotopic Sensitivity

High isotopic sensitivity demands high abundance sensitivity as prerequisite. However, the isotopic sensitivity is also a function of (1) the fractional number of atoms that can be ionized in the source, (2) the transmission of the spectrometer, and (3) the range and sensitivity of the

detector. For those metals which can be analyzed with a thermal ionization type source, the source efficiency is given by:

$$\frac{N^+}{N_0} = \exp \left[\frac{F(\phi - V_i)}{RT} \right]$$

where $\frac{N^+}{N_0}$ = ratio of positively charged to neutral particles evaporated

ϕ = work function of hot ionization filament

V_i = ionization potential sample metal.

F = the Faraday number

T = absolute temperature.

It is noted that:

$$\ln \frac{N^+}{N_0} \propto T, V_i > \phi$$

$$\ln \frac{N^+}{N_0} \propto \frac{1}{T}, V_i < \phi.$$

Since, in general, $V_i > \phi$, it is necessary to use filaments capable of operating at high temperatures, so that a sufficiently large fraction of evaporated atoms would be ionized. The total ion beam current, of course, is proportional to the fractional number of ionized atoms emitted times the rate of evaporation of sample. Where very small samples of material are available (10^{-9} gm), a choice must be made of measuring larger beam currents for a limited time, as compared to detecting smaller ion currents over a longer period.

Two methods of approach have been applied to this problem. The first method was to develop filaments that allow some of the neutral evaporated ions to recontact the hot surface and thus increase the probability of ionization. Presumably, the ions preferentially leak out of a low-geometry source in the direction of the electric field gradient. Much work remains on this problem.

The second approach was to increase the efficiency of collection and detection of ions. More developmental effort has been applied to this than to ion production and, hence, the latter is described in detail. For isotopic ratio work, the conservation of available ions can be maximized by increasing the total number of detected ions. A desirable solution is, of course, an array of detectors, one for each isotope of the group to be

measured, with a one-to-one correspondence between incident ion and recorded output pulse. Long-range work toward this objective is now in process at this Laboratory but desired results are certainly not imminent. (A technique of simultaneous collection for just two isotopes is discussed in the following section.)

A definite improvement has been achieved, however, in the conservation of the ions made available from a given sample by reducing the number of unobserved and, hence, wasted ions. It is clear that if an isotopic sensitivity of 10^{-18} gm is ever to be achieved, even for samples that ionize easily with surface ionization sources, the detection of some 10^4 to 10^6 atoms must be very efficient. The usual "scanning" of the spectrum by magnetic sweeping fails to detect most ionized atoms. If the width at half maximum of the "ion peak" is small compared to the dispersion, this is especially true; i.e., the detector is collecting ions only a small fraction of the time even though the production of ions and, hence, destruction of the sample is proceeding at a constant rate. A modulation of source voltage, with suitable timing and scaling circuitry, has been demonstrated that is both flexible and practical, if magnetic stability is guaranteed.

Basically, the accelerating high voltage on the source is modulated with a step function such that voltages V_1 , V_2 , V_3 are sequentially added to a reference voltage V_0 . If, now, the full width of the ion peak is smaller than the detector slit, small voltage drifts will not alter the detector counting rate. An advantage of this scheme over a ratio recorder is primarily that it is applicable for very low currents, i.e. the ratio of two isotopes, both of whose beam currents are less than 10^{-18} amp. Usually, any normalizing preamplification requires a signal of 10^{-14} amp. It is also clear that, if the timing intervals for the step function voltages V_1 , V_2 , etc. are made sufficiently short, slow fluctuations of beam emission, which are especially troublesome in the surface ionization type of work, can be minimized. Thus, this method is applicable when greater precision is required in ratio measurements, even if the sample size is not a restriction.

Simultaneous Collection of Beam Currents

The fast modulation of acceleration source voltage is convenient, flexible, and greatly reduces the number of ions that are "lost" during an analysis. The simultaneous collection of all isotopes, even at the smallest beam currents, is, of course, the desired objective. If the dispersion is small, the use of photographic plates and possible semiconductor crystal detectors seems to be indicated. If ultimate sensitivity is imperative, the detection of each individual positive ion could be accomplished by microscopic examination of the photographic plates. This would require very high particle energies or postacceleration of charged particles after a spectrum has been resolved. The potentialities of semiconductor detectors are more practical and some work in this direction is being actively pursued.

The simultaneous collection of two isotopes has quite widespread utility since, in many applications, only a single isotopic ratio is required to follow a physical phenomenon.

In the instrument described in this report, the utilization of two 20-stage electron multipliers serves a dual function. One multiplier serves as a detector for the 2-stage magnetic analyzer; the second serves as a detector for the third stage. Together, they are used for the simultaneous detection and recording of two isotopes whose dispersions are within the physical limits of a rather fixed mechanical design. While certain precautions are required with this dual collection technique, and it is not routine, the feasibility of the method has been demonstrated.

Differentiation Between Molecular and Atomic Ions

In the measurement of reasonably large isotopic abundance ratios, it became increasingly evident that there are practical limitations in measuring isotopic ratios of many orders of magnitude. This is the result of the presence of complex ions, which usually cannot be identified, and, in some cases, isobaric interference. An instrument that should measure an isotopic abundance ratio of 10^8 to 1, in certain instances, cannot do so because of the presence of peaks which arise from chemical impurity in the sample. It is unreasonable to ask for clean filaments, for example, when chemical methods, in many instances, cannot measure the desired degree of purity. Also, in most practical problems, the mass spectrometer loses some of its usefulness if a large amount of prior purification must be performed on solid samples or gases to make them susceptible to various isotopic measurements. For these reasons, it seemed desirable to investigate the possibilities of differentiating between atomic and complex ions with the same mass number.

The basic design consideration was the premise that, if a mechanism could be obtained for distinguishing between atomic and molecular species, a machine of very large radius of curvature with high resolution would be unnecessary. This is true for the large class of problems that involve the isotopic abundance ratio measurement of the metals when metallic atomic ions are analyzed. The problem is then reduced to measuring ion beams a full mass unit removed.

The most desirable approach would seem to be the development of a detector to replace the usual electrometer or electron multiplier, which would have a differential response to the impingement of the particles of the same (e/m) ratio but with either an atomic or molecular structure. While it is true that, in isolated instances with low mass number particles, an over-all difference in pulse height has been detected with an electron multiplier³, this approach does not offer any immediate promise. The future possibility of new detector devices should not be excluded, however.

Discrimination of complex and atomic ions at the source seems, in general, unrealistic. Even for surface ionization sources where the presence of complex ions, due to the mechanism of production with a hot filament, is not as serious as in a gas source, complex ions are formed. These arise largely from impurities of the filament or chemical reagents indigenous to the sample. It is a fact that spectroscopically pure tungsten will yield "peaks" on the whole mass spectrum if a very sensitive detector is employed.

By elimination, therefore, it appeared more reasonable to consider the possibility of collecting both atomic and molecular species with a common (e/m) ratio and reanalyzing the atomic constituents of this group. The idea of placing a thin "window" at the focal point of a first magnetic analyzer which would dissociate the molecular species in flight was the result. The problem of dealing with the energy degradation, charge exchange, and scattering of the atomic species appeared difficult but physically interesting. Measurement of these phenomena as a function of velocity, mass, etc. would thus be amenable to control. In fact, since the construction of the subject instrument, the potential measurements of physical parameters have led to greater immediate interest than that in a general operational method for obtaining pure atomic spectra.

It is evident in Figure 2 that, if a sufficiently thin film is placed at S_2 , the focal point of the first magnet and the "source" point of the second, dissociation of the complex ions will indicate a large change in the mass-to-charge ratio of the emerging atoms. Hence, the second magnet serves to sweep the dissociated products out of the trajectory of the beam of the atomic ions. The general practise of utilizing a thin metallic or other film seemed to offer the only practical mechanism that would serve as a dissociating mechanism at a discrete point in the trajectory. The development of very thin metallic⁴ or organic films⁵ made this approach at least promising.

The arrangement of the subsequent magnet and electrostatic analyzer is unorthodox but has a valid excuse for its form. It is clear that the first magnetic analyzer needs only reasonable resolution - sufficient to cause a given (e/m) group to be incident on the thin window as S_2 .

It should be stated that it is impractical to pass an intense, completely unresolved beam through a very thin window at a source point. An intense beam would be one that is many orders of magnitude greater than those usually used for analysis, that is, $\sim 10^{-13}$ amp.

The large unresolved beams caused the film to burn up, while resolved beams have been intermittently incident on films for periods of many months.

Figure 2. Trajectory of Two Ion Beams Traversing Magnetic Lenses
M-1 and M-2 and 90-Degree Electrostatic Lens

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GENERAL DESIGN

The arrangement shown in Figure 1 is justified as a research instrument on the basis of flexibility. This arrangement also seems to be the best compromise for obtaining the first three objectives and carrying out fundamental studies toward the fourth objective. Two 90-degree symmetric 20-in. radius-of-curvature magnetic analyzers are followed by a symmetric 20-in. radius-of-curvature electrostatic analyzer. The use of 90-degree magnets with a 20-in. radius allows (1) analysis of high mass ions at 50 kv, (2) sufficient dispersions, and (3) economical fabrication on standard heavy shop machinery. The radius was also about the largest at which the Pyrex spectrometer tubes could be molded in an oven available at the Laboratory.

The physical arrangement has advantages and disadvantages but it seemed to be the best possibility for exploring the objectives. There are four focal points: S_1 , S_2 , S_3 , and S_4 ; sources may be placed at S_1 or S_2 . With a source at S_1 , beam currents can be detected by a vibrating reed electrometer at S_2 or by electron multipliers at S_3 and S_4 . The use of the two magnets alone yields an integral tandem arrangement spectrometer of very high abundance sensitivity. The use of a source at S_2 gives a conventional double-focusing machine "in reverse". It has the advantage that large unresolved ion beam currents do not strike the electrostatic lens surface with a possible charge buildup. The placement of the electrostatic analyzer following the magnet, however, definitely limits the mass range that can be accepted into the electrostatic analyzer. This limitation is sometimes objectionable. For a restricted mass range, however, this arrangement serves as a double-focusing scheme where the sign of the velocity vector in the differential equations of motion has simply been reversed. For both high abundance and high resolution work, all three analyzing elements are used, i.e. source at S_1 and detector at S_4 . With the two magnetic fields held invariant, scanning of the spectrum is achieved by varying the electrostatic analyzer voltage that serves as the reference voltage for the accelerating high voltage at S_1 . For simultaneous collection work, S_2 is opened to allow the two desired masses to pass to S_3 . The heavier mass is focused and deflected through the electrostatic analyzer and detected at S_4 . The lighter isotope is detected by the first electron multiplier at S_3 . Thus, simultaneous collection of two very low current beams is achieved.

As a tool for molecular dissociation work, it functions as noted: using S_1 as a source, a single group of ions (i.e. having a common (e/m) ratio) are focused at S_2 . The magnetic field, B_1 , is held constant as is also the voltage of S_1 . The second magnet is now powered and regulated from a separate current supply. The atomic ions formed from the dissociation of a molecule can be observed at S_3 by magnetic scanning for these lighter particles. The amount of energy degradation of a beam of known mass atomic ions is also measured in this fashion.

For positive identification of a given group, the beam is allowed to pass to S_4 by adjusting the electrostatic field voltage, E . A measurement of the magnetic field, B_2 , and the electrostatic field, E , thus gives an accurate knowledge of the momentum and energy of the particles formed by the dissociation of the incident molecule. Thus the identification of the dissociated particle group is made positive. It should be emphasized that, for molecular dissociation studies, all three analytical components are necessary: B_1 for selecting the single (e/m) group, B_2 for measuring the momentum of any products formed by dissociation, and E for their kinetic-energy determination.

ION OPTICS

As shown in Figure 2, the complete spectrometer consists of magnetic lenses M-1 and M-2 and the 90-degree electrostatic lens. Magnet M-2 and the 90-degree electrostatic analyzer, regarded as an integral system, result in a symmetric double-focusing mass spectrometer of 20-in. radius of curvature. The conditions for double focusing have been discussed in earlier reports.^{6,7} In all previously constructed double-focusing instruments, the direction-focusing magnetic lens followed the electrostatic lens but the conditions for double focusing are satisfied independently of the order of the two fields. The present arrangement was chosen in order to utilize (1) two magnetic analyzers together as a successive focusing device with a separate ion detector, (2) the second magnetic analyzer and the electrostatic lens together as a double-focusing instrument, or (3) all three lenses as a high abundance sensitivity, high resolution instrument. Furthermore, the present arrangement allows only resolved ion beams to enter the electrostatic lens; this reduces the buildup of voltage on the deflecting plates by electrostatic charges.

In selecting the angle for the electrostatic lens, consideration was given to reducing the line width resulting from angular aberration to a minimum while keeping the object distance of the lens at a convenient distance from the second magnetic lens. The value of the spherical aberration for a magnetic lens is given by $\alpha^2 R$. The net angular aberration of a symmetrical magnetic and electrostatic lens as a function of the electrostatic sector angle is indicated in Table 2.

TABLE 2. Net Angular Aberration of Symmetrical
Magnetic and Electrostatic Lens

<u>Lens Angle, deg</u>	<u>Net Angular Aberration, $\alpha^2 R^*$</u>
60	1.8
90	0.6
127	0.3

* α = half-angle of divergence of ion beam

R = radius of curvature for both the magnetic and electrostatic lenses.

FIGURE 3. Rear View of Assembled Three-Stage Spectrometer

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FIGURE 4. Subassembly of 20-inch Magnet

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Since for small divergent angles the contribution toward the ion beam width due to the spherical aberration of the magnetic analyzer $\alpha^2 R$ is only a small percentage of the beam width, the correction achieved by a 90-degree electrostatic lens is sufficient to keep this term negligible. The relatively small additional aberration correction in increasing the sector angle toward 127 degrees would not be worthwhile. It certainly would not be practical because the object and image distances would decrease toward zero; this makes using an electron multiplier impossible at the focal point S_3 .

With usage as a two-stage mass spectrometer, the dispersion at S_3 is equivalent to that of a single 40-in. radius-of-curvature magnetic analyzer. However, the angular separation of two different mass or energy ion beams at S_3 is four times that which would occur in a single 40-in. radius of curvature. Therefore, in conjunction with the electrostatic analyzer, the complete system from S_1 to S_4 does not fulfill the conditions for double focusing. However, for experiments in which thin films are placed at the intermediate focal point S_2 , essentially a new source is formed at this point with a varying degree of energy spread. The subsequent magnetic and electrostatic lenses then act as a double-focusing spectrometer for such beams.

PHYSICAL DESCRIPTION

The magnets, the electrostatic analyzer, spectrometer tube and component parts, electron multiplier-detectors, and the vacuum system are photographed in Figures 3 to 6, 7 to 10, 12, 13, and 15.

The magnets were designed for field strength of 10,000 gauss and an attempt was made to keep the design consistent with simple fabrication and assembly. The decision to restrict the magnet-focusing motion to a single degree of freedom (as indicated in Figure 1) was based on experience that other small adjustments yield no better performance. With a long-focal-length system at least, this seems to be the case. Thus, a base plate was designed with simple 4-in.-diam roller bearings, four for each base plate. The roller bearing axles were mounted in commercial ball bearings. A key-way constrained the translational motion and a small-wheel-actuated screw-drive provided a convenient focusing adjustment (see Figure 3). In assembly, the focal points of the spectrometer were determined by accurate positioning and leveling of the base-plate assembly. The base plates were mounted on suitable concrete piers and leveled by shimming.

The general magnet design can be seen clearly in Figure 4. The yoke pieces are approximately 8 in. thick; the pole piece is 2 in. thick. The pole pieces were hydrogen-annealed; the yoke pieces were annealed only

in air. The pole pieces are retained to the upper yoke section by welding. The uniformity of gap dimension was achieved by four brass spacer blocks, each machined to 0.750 in.

Each exciting coil comprised 21,500 turns of 0.038-in.-diam copper wire coated with heavy Formex insulating coating. The resistance of each coil was approximately 1,170 ohms. A field strength of 10,000 gauss was achieved with 0.365 ampere. The maximum measured field strength was 11,600 gauss at 0.500 ampere. This was a limitation imposed by the power supply rather than saturation of the magnetic circuit.

Both the spectrometer tubes and the electrostatic analyzer plates were formed in carbon molds. The spectrometer tubes were 1-1/2-in.-OD Pyrex tubing with a 3/32-in. wall. These tubes were first given a nominal 20-in. radius by hand-working and were subsequently placed in the carbon mold shown in Figure 5. These carbon molds were quite accurately machined to dimension and utilized for forming a large number of tubes. After fabrication, the tubes were coated on the interior surface with stannous chloride; this conducting film is required to prevent electrical charge buildup from the ion beam.

The electrostatic analyzer was formed from rectangular Pyrex plates 2-1/2 in. wide and 5/32 in. thick. Both the convex and concave molds that formed the cylindrical surfaces are shown in Figure 6. These molds were machined to radii one centimeter less and one centimeter greater than the mean 20-in. radius. The inner surfaces were coated with gold which gave a resistance across the long dimension of only a few ohms. A subassembly of the analyzer is shown in Figure 7. Ceramic pieces were used both as dimensional spacers and supports. A 4-in.-ID Pyrex pipe made by Corning Glass Works housed the electrostatic analyzer section. An assembled view is shown in Figure 8.

The component parts that connect the glass sections and that contain variable slit assemblies at S_1 , S_2 , S_3 , and S_4 will be described briefly. A duraluminum cylinder was fabricated so as to accommodate a high-voltage source, differential pumping, a vacuum interlock, and the variable collimating slit assembly. This unit is shown in Figure 9, before mounting of the source and slit assembly. The vacuum interlock consists of a polished stainless steel rod, 1/2-in. diam, which conforms to a convex cylindrical surface with an intervening Teflon gasket. A 1/8-x-1-in. slot in this rod allows beam passage in the normal position; a 90-degree rotation completely isolates the analyzing region from the source region. Pressures of 10^{-6} mm of mercury are maintained against external atmospheric pressure. This valve makes source sample interchange convenient and reduces harmful oxidation of the electron multiplier dynodes. A micrometer screw allows adjustment of the collimating slit from 0.000 to 0.040 in. The source can be adjusted so as to maximize the ion beam incident at the detector by means of a screw actuated through a vacuum-tight bellows.

FIGURE 5. Carbon Mold for Forming Spectrometer Tubes

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FIGURE 6. Convex and Concave Molds that Formed Cylindrical Surfaces of Rectangular Pyrex Plates of Electrostatic Analyzer

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FIGURE 7. Subassembly of Electrostatic Analyzer

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FIGURE 8. Assembled Electrostatic Analyzer Section

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FIGURE 9. Vacuum Interlock and Source Holder

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FIGURE 10. Closeup of Second Slit Assembly

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Figure 11. Film Holder and Slit Assembly

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Two slit assemblies have been used at S_2 . The first assembly consisted of a pair of rotating vanes which could be set at the desired slit width by means of an external magnet. A detail of a second slit assembly, which has been used at S_2 for more recent work, is shown in Figure 10 and in the schematic diagram, Figure 11. The film holders can be independently interposed or withdrawn from the beam.

The large adapter at S_3 was machined from a single block of duraluminum (see Figure 12). It contains a slit system with a 0- to 40-mil micrometer adjustment, a connector for the 1-1/2-in. Pyrex spectrometer tube, an adapting flange for the 4-in. Pyrex pipe that houses the electrostatic analyzer, and a vacuum interlock that separates the electrostatic analyzer region from the preceding section. A 20-stage electron multiplier, which can be moved in such a manner that the first dynode either intercepts the beam at S_3 or can be withdrawn so that the beam continues through the vacuum valve and electrostatic analyzer, is also mounted in this block. Adjustment of the dynode position with respect to the vertical line beam is especially convenient and an optimum position is determined through trial and error, by maximizing the number of output pulses from the multiplier.

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The adapter at S_4 shown in Figure 13 serves to join the 4-in. Pyrex 90-degree vacuum pipe of the electrostatic analyzer to a standard Corning 60-degree glass elbow in which is mounted the second 20-stage electron multiplier. The last slit system is actuated from this brass flange by micrometer screws; the electrical leads for the analyzer plates were placed through appropriate glass-to-metal terminals in this flange. The slits at S_3 and S_4 were made with each slit edge independently adjustable so that the slit could be centered and its width varied.

The vacuum system consists of five Consolidated Vacuum Corporation Model GHG-10 mercury diffusion pumps backed by a single fore-pump. Pressures of 10^{-7} mm of mercury can be obtained easily and the glass components eliminate the necessity for bakeout. Separate ion gauges in the source, magnetic analyzer, and electrostatic analyzer regions with vacuum interlocks separating these three regions facilitate leak-checking.

Perhaps the most desirable feature of the instrument is its adaptability to modification or change for various types of experimental investigations. The reversal of magnetic and electric polarity of magnet M-2 and the subsequent analyzer are especially valuable in investigating negative ions formed in the S_2 region. Work with the instrument during the last year and a half has clearly demonstrated that this spectrometer is more than adequate for a large uncompleted program of research problems. It is also hoped that it will serve as a useful prototype for future mass-spectrometers of modest dimensions.

ELECTRONICS

A block diagram of the electronic circuitry that is associated with the 3-stage spectrometer is shown in Figure 14. The electronic circuitry is divided into four sections, as indicated by the dotted lines, which include the source, the magnetic analyzer, the electrostatic analyzer, and the detector sections. The negative and positive voltage supplies for the electrostatic analyzer lens are included in the source section since the negative supply serves as a reference for the source accelerating voltage.

Either of two high-voltage supply units is used for accelerating positive ions at the source and each unit also includes the negative and positive supply for the electrostatic analyzer. The first unit has a range of 2500 to 10,000 volts for source voltage and 100 to 400 volts for each plate of the electrostatic lens. The output voltage of the negative supply is controlled by varying the resistance of the feedback voltage divider. Since this output voltage serves as a reference for the positive voltage of the electrostatic lens and the source voltage, all three output voltages are varied by means of the same controls. The ratio of source

FIGURE 12. Adapter at S-3 Containing Variable Slits
and Vacuum Interlock

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FIGURE 13. Adapter at S-4 Showing Variable Slits

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Figure 14. Block Diagram of Electronic Circuitry for Three-Stage Spectrometer

voltage to negative electrostatic voltage is nominally 25.4 to 1 for this instrument, but can be varied from 23-to-1 to 27-to-1. Beam centering at S_4 is accomplished by adjusting the positive electrostatic voltage relative to the negative voltage over a small range. Long-term stability, regulation, and ripple for all three output voltages are of the order of 0.002%. This unit was designed and constructed by the New Jersey Electronics Company, in accordance with KAPL specifications.

Since the source filament must operate at positive ion accelerating potentials, the filament power supply is insulated for 15 kv. This unit supplies up to 20 amp at 5v dc and is current-regulated by means of a feedback servoamplifier. Ripple is minimized by using 3-phase ac power and a special transformer with 12 taps on the secondary winding supplying 12 selenium rectifiers in parallel.

A second special high-voltage unit, also supplied by New Jersey Electronics Corporation has a source-voltage range of 5000 to 50,000 volts and an electrostatic-lens-voltage range of 200 to 2000 volts. The stability, regulation, and ripple are $\sim 0.01\%$ for this unit since at these higher operating voltages the mass-spectrometer requirements are not so stringent. A source filament current supply that is regulated for line voltage variation is included as an integral part of the unit. It consists merely of a Variac control and a high-voltage, insulated, step-down transformer supplying up to 20 amp at 5 volts.

Provision is made on both of these high-voltage supplies for connection to a unit known as a four-channel modulator which was designed and constructed at KAPL. It serves two functions: (1) A motor-driven potentiometer linearly varies the negative reference voltage and hence the source accelerating voltage in order to sweep the mass spectrum. (2) The high voltage is modulated in a step-function for preset time periods and preset mass positions. The second function is illustrated in the detector section of the block diagram in Figure 14. The 1-Mc scaler shown is a Model 5510, Berkeley Universal Counter and Timer which also furnishes the 1-sec time base for the modulator unit. Two "glow tube" type counters in the latter provide counting periods, for each of the four channels, of 1 to 10 sec in 1-sec steps and 10 to 100 sec in 10-sec steps. These ranges can be varied by selecting the output of the frequency divider in the counter and timer. The appropriate period may thus be chosen for each channel, depending on the relative isotopic abundance. The ion-accelerating voltage for the desired isotopic mass position is selected by a 10-turn potentiometer for each channel. These potentiometers each have a range of 100 volts when used with the 10-kv supply and 500 volts with the 50-kv supply. The four channels are sequentially "scanned" by

means of a solenoid type "stepping-switch" which is actuated at the end of each counting period. There is a delay of two seconds, during which time the switching takes place and the accelerating voltage stabilizes. Also, during this time, the number of pulses accepted by the 1-Mc scaler is printed on tape by a digital recorder.

The remaining circuitry in the detector section is associated with the detection and recording of positive ions, or in some instances, negative ions. Each of the two electron multipliers previously mentioned has its own preamplifier, either of which may be connected to the main amplifier and discriminator unit. Equalized pulses from the discriminator can be fed through a fast, i.e. 10-Mc scale of 100, or bypassed through a relay directly to a scale of 10,000. The relay is actuated by a switch on each channel of the modulator unit. From this point, the pulses are also fed to the 1-Mc scaler mentioned in connection with the modulator unit. The output of the scale of 10,000 is integrated in a count rate meter and the resultant current is plotted on a chart recorder. Either the scale of 10,000 or the 1-Mc scaler can be used for pulse counting and their outputs can be compared for accuracy.

The magnetic analyzer section of Figure 14 includes the magnet power supply and the nuclear magnetic resonance field control equipment. The power supply furnishes a current of up to 0.5 amp to the excitation coils of the two magnets in a series-parallel arrangement. The two coils of one magnet are connected in series with the two coils of the other so that the current is the same for both, and the fields are therefore very nearly identical.

It should be noted that, in practice, no two magnets with pole-face areas of sizeable dimensions can be constructed so that their integrated field strengths are equal. Hysteresis effects would produce differences even if the magnets could be adjusted to the identical value. In any ion beam work or charged-particle analysis the magnetic field adjustment must be made experimentally. Matching of the paired fields of this spectrometer is accomplished by a shunt which varies slightly the relative current in the coils of the two magnets. The adjustment of the shunt control is determined by maximizing the ion beam incident on the detector slit.

The nuclear resonance equipment, a type developed by Pound,⁸ includes standard units that consist of a Model 103 power supply, Model 102 oscillator, Model 104 lock-in amplifier, and Model 105 modulator, supplied by the Nuclear Magnetics Corporation. The equipment furnishes a resonance absorption signal whose linear relationship between magnetic field and oscillator frequency makes it useful for field control, measurement, or monitoring. The field-sensing probes contain a lithium-chloride solution

which provides both proton and Li^7 resonances. Four interchangeable probes cover a total range of 282 to 7300 gauss for proton resonance and 2000 to 18,750 gauss for Li^7 . In practice, only the proton resonance is used for field regulation since its range is adequate for most work and the Li^7 signal is too weak for reliable control.

As shown in the diagram, the signal from the nuclear resonance oscillator is fed to the lock-in amplifier (phase detector) and the resulting error signal is used to control the magnet power supply. The probe senses the field of one of the magnets, but since the coils of both magnets are excited in series by the same supply, both fields are regulated by the same probe. Since the oscillator now serves as a reference, the field stability is dependent on the stability of the oscillator, which was found to be better than one part in 20,000 for an average 24-hr period. Precise measurement of the oscillator frequency, and therefore the field magnitude, is obtained with a Berkeley Model 5571 frequency meter which has an accuracy of ± 1 cycle. The crystal oscillator in the latter unit, which serves as a time base for frequency counting, is stable to better than one part in 10^6 . For more precise field measurement, the crystal oscillator can be calibrated against National Bureau of Standards station WWV.

A unique method of monitoring the fields of the paired magnets to detect any differential drift is also shown in Figure 14. Two nuclear resonance probes are connected in parallel to a second oscillator unit which will produce a resonance signal for the field of either magnet. Since the fields are nearly identical, as mentioned above, the two resonance "peaks" are superimposed on an oscilloscope screen by passing a direct current of 0.5 amp or less through the Helmholtz modulation coil of one of the probes. When thus superimposed, the amplitude of the displayed signals is doubled as shown in Figure 15. A change of 0.05 gauss in relative field strength of the paired magnets is evident when there is a decrease in the amplitude of the superimposed resonance signal.

Since the procedure of connecting two probes in parallel alters the frequency range, three pairs of probes were supplied by the manufacturer on special order. The total range of these probes for proton resonance is from 6.9 Mc to 31.0 Mc or 1600 to 7400 gauss.

Another procedure for field control is used in the investigation of energy degradation of positive ions in passing through thin films.⁹ Here, one magnetic analyzer field is held constant by nuclear resonance control while the field of the second magnetic analyzer is swept over a wide range. This type of control, which senses the magnetic field, is

(A)

(B)

FIGURE 15. Resonance Peaks with Different and Equalized Magnetic Fields

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essential in this procedure since there is appreciable interaction between the fields of the two magnets. Another power supply excites the coils of the second magnetic analyzer and this supply is regulated by a second set of nuclear resonance equipment. To sweep the field of the second analyzer, a motor drive and a speed reducing gear assembly is employed to rotate the oscillator tuning capacitor. A unit of this type, designated Model 108 Autoscan, was developed by the Nuclear Magnetics Corporation. Both the speed and range of rotation are adjustable; the latter is limited by a reversing switch. Push buttons were added as a convenience so that the direction of sweep may be reversed at any time. Accurate determination of the energy degradation is accomplished by precise measurement of the nuclear resonance oscillator frequency at the field required for detection of the degraded ions that have traversed the film.

EXPERIMENTATION RESULTS

Abundance Sensitivity

Preliminary measurements of the abundance sensitivity of the 3-stage mass spectrometer indicated that the abundance sensitivity was greater than 10^7 in the intermediate mass region. To measure a ratio larger than 10^7 , an indirect method had to be employed, since such a large ratio could not be measured with the electron multiplier. However, by collecting the total charge of a Na^{23+} beam by means of a vibrating reed electrometer and then using the electron multiplier to count the ions at the mass 23.5 position, a ratio of 10^{10} was obtained. Since impurity peaks greater than this level were found at every integer mass position, the abundance sensitivity at mass 24 can only be said to be $>10^{10}$.

The higher abundance sensitivity of the 3-stage spectrometer was obtained quite independently of the particular mass region under examination. The above-mentioned figure of 10^{10} for the ratio of the signal of Na^{23+} to the 23.5 mass position is conservative. This measurement presented difficulty in obtaining a sufficiently intense ion beam. In a single measurement, a figure of 10^{11} to 1 was obtained although, because of variable ion emission, this could not be reproduced. In a single-stage mass analyzer, the Na^{23+} beam would have been observed over a region of several mass units ranging from probably 1 ppm at one mass unit distance and decreasing only approximately one additional order of magnitude. It appears that, in order to achieve a 10^{11} to 1 signal to noise ratio, the effects of certain phenomena that are normally considered negligible must be minimized. Interactions that cause ions to follow paths that differ from calculated ion optics, even though they are generally not apparent, must include (1) wide- as well as small-angle elastic gas scattering, (2) inelastic collisions with gas molecules with subsequent degraded ion

energies and the production of secondary ions of very low energy, (3) inelastic encounters with slit edges, tube walls, etc., (4) charge exchange with residual gas molecules, and (5) reflection and diffraction phenomena.

On the basis of a number of experiments in which charge exchange, energy degradation, and scattering were measured qualitatively, we are of the opinion that the marked improved abundance sensitivity of the instrument, operating as a three-stage spectrometer, can be attributed to the double-focusing action upon ions traversing the field-free region in the neighborhood of S_2 . In this region, ions that have undergone inelastic encounters are nearly completely filtered out by the second magnet and the electrostatic analyzer combined. It is true that this effect is smaller than the contribution from small-angle elastic scattering but it appears to be the next most important factor in explaining the spatial dispersion of ions of a given mass over a wide range. The spectrum shown in Figure 16 of the triplet at mass position 16 was obtained with the 2-stage magnetic analyzer. A comparison of this spectrum with the spectrum shown in Figure 17 graphically illustrates the marked increase in abundance sensitivity obtained with the 3-stage analyzer.

Isotopic Sensitivity

The proposed methods, previously described, of increasing the isotopic sensitivity by reducing the number of undetected ions have been investigated. The 4-channel modulator and the simultaneous collection techniques gave encouraging results. The resolution of the 2-stage spectrometer in the U^+ region, as shown in Figure 18, is sufficient for the use of detector slits as much as ten times the beam width in size. Under these conditions, the use of the 4-channel modulator is completely automatic; no attention is required other than the original voltage adjustments for each isotope to be counted.

The results of isotopic analysis in which the 4-channel modulator was utilized to alternately record each of four uranium isotopes for a preset counting time are shown in Table 3 below. All the errors quoted are standard deviations for a series of isotopic ratio determinations.

TABLE 3. Isotopic Analysis

Sample	U^{235}/U^{238}	U^{238}/U^{234}	U^{238}/U^{236}
	Ratio	Ratio	Ratio
1	18.48 ± 0.11	5.31 ± 0.04	11.40 ± 0.08
2	18.39 ± 0.10	5.30 ± 0.03	11.40 ± 0.11
3	18.32 ± 0.08	5.24 ± 0.05	11.29 ± 0.10

VOLTAGE DISPERSION 42 VOLTS/MASS
 LINEAR DISPERSION 0.034"/MASS
 RATIO OF DISPERSION TO HALF WIDTH ~ 30 } $m=235$

Figure 18. Mass Spectrum of Irradiated Uranium Obtained with Two-Stage Spectrometer

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Figure 19. Resolution of Complex Ion in the Uranium Region Obtained with Two-Stage Spectrometer

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The reproducibility of the measured ratios among analyses is evidently within the error of any given analysis. The average time was approximately two hours for a complete analysis. Sample No. 1 contained approximately 0.5 μgm of total uranium while sample No. 2 contained 1.5 μgm of total uranium.

In addition, a considerable number of analyses were made on a sample of natural uranium. The U^{238}/U^{235} ratio was obtained in each sample with a counting time interval of 1 sec for the U^{238} ion beam and 3 sec for the U^{235} beam. The total number of counts obtained for U^{235} in one complete determination was approximately 10^4 . In each determination, the counts for 100 counting intervals for each isotope were accumulated; the total analysis time was about 15 minutes. No time or temperature trends in the average isotopic ratio of U^{238}/U^{235} were observed in any of the samples. Therefore, the data obtained were grouped in a frequency distribution, and fitted to a normal distribution as indicated in Table 4.

A chi-square test for goodness of fit with the normal distribution gave a p of 0.9. The mean U^{238}/U^{235} ratio and the precision obtained for a complete analysis of each of the natural uranium samples are shown in Table 5.

TABLE 4. Frequency Distribution

<u>U^{238}/U^{235}</u> <u>Ratio</u>	<u>Expected</u> <u>Frequency</u>	<u>Observed</u> <u>Frequency</u>
135-136	2	0
136-137	7	7
137-138	20	25
138-139	33	32
139-140	36	34
140-141	25	24
141-142	11	13
142-143	3	3
143-144	1	1

$$\bar{X} = 139.15$$

$$\sigma = 1.47$$

TABLE 5. Isotopic Analysis of Natural Uranium Samples

Sample	U^{238}/U^{235}	Standard Deviation
	Ratio	
1	139.1	± 1.2
2	139.4	± 0.8
3	139.0	± 1.1
4	139.2	± 1.2

The reproducibility of the ratios obtained from the four samples of natural uranium is excellent. The absolute value of the U^{238} to U^{235} ratio for natural uranium is not necessarily within the limits given here. In this work, only the reproducibility of the ratios was considered, and no attempt was made to correct the mean ratio for any possible systematic error.

Simultaneous Collection

In most mass-spectrometric devices, the counting of individual positive ions of high mass is difficult because of limited dispersion which does not allow the use of large dual collectors in the region of the detector focal plane. In the present instrument this difficulty is circumvented as shown in Figure 2.

The fields of both electromagnets are held fixed by proton resonance control and are "matched" for the best focus. The accelerating potential at S_1 is then adjusted so that isotope B is made to follow the median path. The lighter isotope A is displaced by the amount σ at S_2 and is actually focused along this diagonal, joining S_1 , S_2 and S_3 and detected by the first electron multiplier. With an "S"-shaped arrangement for the magnets, the dispersion at S_3 is 2σ , i.e., twice that of a single symmetric analyzer. At S_3 , isotope B passes into the electrostatic analyzer and is detected by the second electron multiplier.

As indicated in Figure 2, it was necessary to operate the first electron multiplier with its first dynode at ground potential and to use a blocking condenser to isolate the preamplifier from the high voltage on the collector. If operated in conventional fashion, the electron multiplier completely defocuses beam B.

For this first test, a sample of natural uranium was run; isotopes A and B were respectively, U^{235} and U^{238} . The separation (2σ) at S_3 was approximately 0.5 in. This allowed the U^{235} beam to clear the first electron multiplier easily. Both multipliers were fitted with bellows so that the response could be optimized by horizontal positioning. The first multiplier was actually baffled by a shield with a 1/8-in. slit and the usual defining slits at S_2 and S_3 were removed.

Some interesting results have been obtained even though the work is still in the exploratory stage. First, the two electron multipliers appear to have very nearly the same response. That is, the probability of an output pulse arising from an incident positive ion is very nearly equal for the two multipliers that were tried, even though the kinetic energy of these ions was 10 kv for the first multiplier and 15 kv for the second because the first dynode was at ground rather than at -5 kv. The source accelerating high voltage was ~10 kv.

Secondly, the transmission loss in the electrostatic analyzer under optimum conditions is very small. Apparently all or nearly all the ions that arrive at S_3 finally reach S_4 . This seems to be true, however, only when the total (unresolved) beam current is small. The possibility of charge building up on some insulators inside the system must be investigated.

As expected, variations in ion emission current (of the order of 500%) as a result of raising or lowering filament current to simulate the random emission fluctuations of positive ions caused no measurable change in the ratio.

An interesting result was obtained when the gating time was restricted to 0.01-sec intervals. The sample used in this analysis was 10^{-6} gm of natural uranium. The results of the analysis are shown in Table-6.

TABLE 6. Isotopic Analysis of Uranium

<u>Set</u>	<u>U^{238}/U^{235} Ratio</u>	<u>Total Counting Time, sec</u>
1 (10 readings)	139.5	0.1
2	141.5	0.1
3	139.0	0.07

The ratios have been corrected for counting losses. It is certainly premature to state the general feasibility of doing an isotopic ratio measurement in 0.1 sec, but it may be possible to reduce the sample size materially if such a measurement is necessary. At any rate a need for much additional work in this area seems to be indicated.

Differentiation Between Molecular and Atomic Ions

The differentiation between molecular and atomic ions has been accomplished in two basically different ways with this mass spectrometer. The result of the first method, that of resolving a molecular ion beam from an atomic ion beam is shown in Figure 19. The molecular beam is K_8^{39} , which is contained as an impurity in the uranium beam. This spectrum

was obtained by means of the two-stage magnetic analyzer with a source slit width of approximately 0.008 in. The mass difference between K_8^{39} and U^{234} is 0.258 atomic mass units. It is evident that molecular impurity peaks from atomic ions differing by at least 100 millimass units can be resolved at reasonably high beam intensities.

However, for molecular ions differing in mass from an atomic ion by a considerably smaller amount, such as H_2^+ and D^+ , where the mass difference is only 1.5 millimass units, the resolution would not be sufficient for analytical purposes. However, a second differentiating method, which is independent of the mass difference but operates on the chemical bonds of the molecule, has been developed for this instrument. The method consists of placing thin organic resin (polyvinylchloride-acetate copolymer) films or thin metallic foils (~200 Å thick) in the ion beam at the focal point S_2 shown in Figure 1. The molecular ions dissociate upon traversing the film and the mass fragments are separated from the original atomic ion beam by the magnetic analyzer M-2. The atomic ions merely undergo more or less energy degradation, depending upon their mass and the velocity imparted by the accelerating voltage. The results of this method for the dissociation of a beam of CO_2^+ is shown in Figure 20. The spectrum was obtained from the 2-stage spectrometer, but the three stages were needed to make the mass and energy identification certain. Furthermore, the energy-straggling which accompanies this process broadens the beam considerably as shown in Figure 20. By use of the double-focusing properties of the second magnet and electrostatic lens, the beam may be concentrated into a narrow line. The dissociation of a beam of H_2O^+ by traversing the thin film was also observed. The only products of the dissociation were atomic ions of both positive and negative charge. No H_2^+ , O_2^+ , or OH^- ions were observed.

The unique arrangement of the lens system of this spectrometer also offered the opportunity to study energy loss of heavy charged particles at relatively low velocities. The curves shown in Figure 21 show the results of the measurements to date.

The measurement of the loss of energy for atomic ions was obtained by comparing the magnetic field strength (M-2) required to focus the atomic beam without traversing the film to the magnet field strength required to refocus the beam after the film was inserted. The field strength was determined very precisely by the nuclear magnetic proton resonance oscillator frequency as measured by the crystal referenced frequency counter.

A continued effort is being made to expand these measurements of relative energy loss and also to investigate charge equilibrium ratios, negative ion production, and multiple charged ions. The need for such data in this field has been pointed out by Allison and Warshaw¹⁰ in their review, "Passage of Heavy Particles Through Matter", which is cited as follows:

Figure 20. Dissociation of CO_2^+ By Thin VYNS Film

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Figure 21. Helium, Nitrogen, and Argon Ions After Traversing Thin VYNS Film

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Unclassified

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"For the heavier ions, which move much more slowly at kinetic energy values equivalent to those of protons, charge exchange is certainly an important mechanism for energy transfer; but in the complete absence of data, speculation must be considered futile."

All the results to date have been obtained with the organic resin films. However, to normalize the curves in Figure 21 to a substance of known relative stopping power, nickel films have been prepared from evaporated nickel foil supplied by Chromium Corporation of America, who guarantee tolerances of ± 50 Å for nickel foil with a nominal thickness of 250 Å. The method of preparing these films has been described by Bashkin and Goldhaber.⁴ Measurements of energy loss for protons up to 50 kev will be compared with results of energy loss by heavy ions through the nickel films.

Molecular Dissociation by Residual Gas Molecules

During the investigation of the mechanism for dissociation of CO_2^+ mentioned above, it was found that without the film placed at the focal point of the first magnetic analyzer, relatively large beam currents of CO^+ , CO_2^{++} , O_2^{++} , O^+ and C^+ were recorded at S_3 even though the only ion beam that could have been transmitted through the first magnet was mass 44 (CO_2^+) or a similar charge-to-mass ratio.

The only explanation for the appearance of the CO^+ , CO_2^{++} , O_2^{++} , O^+ and C^+ at S_3 is that the CO_2^+ must dissociate in the field-free region between the first and second magnetic analyzer. Experiments with A^{40++} and He^{4++} beams have shown that doubly charged ions also undergo charge exchange in the field-free region and are detected at S_3 as A^{40+} and He^{4+} .

The phenomena were previously observed by Mattauch and Lichtblau¹¹ and were discussed by Norton¹² at the Mass Spectrometer Symposium held at the National Bureau of Standards. However, the possibility in a single analyzer of misinterpretation of the $\text{A}^{++} \rightarrow \text{A}^+$ charge exchange with A_2^+ formed in the source or similarly with other elements is not present in the 3-stage instrument. This is because only one ion beam with a unique charge-to-mass ratio is present at S_2 . Hence, there is no ambiguity in determining which event takes place, the charge exchange or the noble-gas molecule formation.

Since a partial dissociation of compounds was observed even with the residual gas pressure less than 10^{-6} mm of mercury, a more complete dissociation could be obtained by means of a differentially pumped chamber at S_2 into which a controlled amount of various gases could be admitted. Actually this method appears more promising for practical dissociation for analytical purposes since the resulting beam is not widened considerably because of energy straggling or large deviations in the direction of the incident ion. Additional experimental data in this area are needed and an attempt will be made to obtain such data in the course of future work.

CONCLUSIONS

The potentialities of the mass spectrometer that was developed have been demonstrated in certain difficult physical measurements as well as in the areas in which analytical machines are usually employed. In addition, considerable instrument experience has been obtained with methods for the control of matched magnetic fields, simultaneous collection of ions and with the requirements for stable operation at 50 kv. Scanning of the spectrum by means of dual magnetic-field variation as well as voltage scanning was successfully accomplished.

The usefulness of this instrument in isotopic analysis is surpassed by its potentialities when applied to measurements of charge exchange, energy loss, cross sections for atomic interactions, small-angle scattering and a large number of measurements in the field of chemical physics, especially molecular dissociation. This instrument is unique in its application to the study of interaction phenomena of a given (e/m) group at a controlled energy with subsequent determination of the resulting momentum and energy of the particles after interacting, especially for ions of high atomic mass and relatively low velocities.

This instrument could be applied to various problems in the field of isotopic analysis, particularly those requiring an abundance sensitivity greater than 10^7 . Most of the problems associated with the production of new nuclides by charged particle bombardment of higher order neutron capture cross sections in low power reactors would occur in this field. Many geological and cosmological problems require the detection and identification of very small amounts of isotopes formed by natural cosmic radiation. If, in such problems, the daughter nuclei formed in the reactions are stable or very long-lived, the sensitivity of this instrument would easily surpass that of radioactive counting techniques.

Application of this instrument would also prove useful in solid-state research, for investigations of impurity concentrations in semiconductors, since the double-focusing properties of the second magnet and electrostatic lens combined allow an arc source to be used for the production of ions.

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