

## Design of a Compact Permanent Magnet Cyclotron Mass Spectrometer for the Detection and Measurement of Trace Isotopes.

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### ABSTRACT

A technique for the detection of trace amounts of rare isotopes, Cyclotron Mass Spectrometry (CMS), is described. This technique uses the relationships between particle mass, charge, magnetic field strength and cyclotron orbital frequency to provide high mass resolution. The instrument also has high sensitivity and is capable of measuring isotopes with abundances of  $< 10^{-12}$ . Improvements now being implemented will lead to further increases in the sensitivity and enhance operating parameters such as cost, portability, and sample throughput.

### 1. INTRODUCTION

Measuring the abundance of trace isotopic constituents has applications in many fields. In biomedicine, the use of carbon-14 ( $^{14}\text{C}$ ) has an important role in studying the metabolism of drugs and DNA sequencing. Archaeological applications include the radiodating of carbon containing samples to obtain accurate ages of fossils and anthropologic artifacts. In planetary geology, the abundance of the isotopes of iridium in past geological periods has given rise to the "catastrophe" theory of mass extinctions. Environmental applications of the trace detection of isotopes also abound, such as in the measurement of the  $\text{CO}_2$  content of ice cores and deep ocean water or measuring the distribution of combustion byproducts in the atmosphere. In general, these applications require a very high sensitivity and selectivity. The combination of the samples being very dilute in the isotope desired ( $< 10^{-9}$ ) and containing other atoms and molecules with almost the same atomic or molecular weight makes essential a detection scheme with high sensitivity *and* specificity (or resolution.)

One method of high-sensitivity detection has been Accelerator Mass Spectrometry (AMS).<sup>1)</sup> In this technique, the sample of interest is ionized, and a charged-particle accelerator is used to detect single atoms of the isotope of interest. The first use of an accelerator as a mass

analyzer was made in 1939 by Alvarez, who measured  $^3\text{He}$  at natural abundance using a cyclotron. The "modern" era of AMS began in 1977, when Muller proposed using a cyclotron for carbon and beryllium measurements<sup>2,3)</sup>, and the groups at the University of Rochester, and McMaster University demonstrated  $^{14}\text{C}$  detection with tandem accelerators<sup>4,5)</sup>. AMS has developed over the last decade and a half into a powerful tool for use in the detection of trace quantities of materials, having been employed in a variety of environmental, archaeological, and geological studies. Virtually all work is performed on large tandem accelerators. By using the high energy ( $\geq 1$  million electron-volts) inherent in these accelerating schemes and ion detection techniques capable of measuring a single particle, analysis of isotopes which are present in samples at a level of 1 part in  $10^{15}$  has been achieved.

Unfortunately, the use of AMS is not widespread. AMS measurements are performed at central, multimillion dollar facilities, with experimenters bringing or shipping their samples to the site to be analyzed. There are several reasons for this. First, the facilities are large and expensive to acquire, operate, and maintain. The utility requirements are substantial, and a multi-person crew is required to operate the instrument. Second, because of the large accelerating voltages used, substantial amounts of radiation are produced, necessitating heavy shielding. Third, the sample preparation is complicated, requiring the production of solid samples suitable for use in sputter ion systems. Combined, these factors have made AMS a relatively expensive technique with low sample throughput.

To retain the advantages of conventional AMS without the disadvantages of the large accelerator facilities, the original idea of using a cyclotron as the accelerator has been revived, with the new wrinkle that the cyclotron be small and that the accelerating voltages be modest. In this incarnation, the technique has been dubbed Cyclotron Mass Spectrometry (CMS.) Here, the large cumbersome accelerator has been replaced by a compact, low-energy cyclotron. Previous work at LBL<sup>6,7)</sup> has demonstrated the principle of these devices and has shown that CMS can have significant detection advantages over the scintillation methods used routinely in biomedical research for  $^{14}\text{C}$ . As a result, several small cyclotron spectrometers are now

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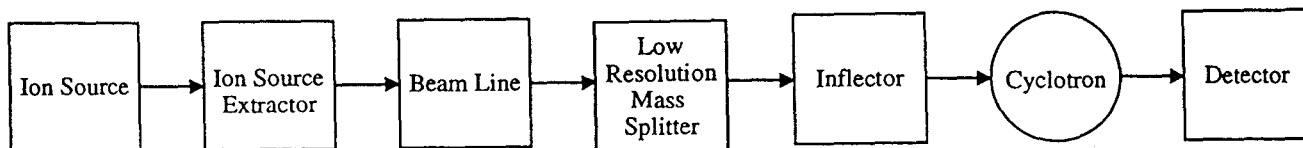


Fig 1 Schematic diagram of the cyclotron mass spectrometer

under development around the world. However, the utility requirements, sampling methods, and sensitivity of CMS are still such that widespread application of the technique to common problems is not currently feasible.

This paper describes the development program now underway at Lawrence Berkeley Laboratory (LBL) to improve the performance and operating characteristics of CMS. The new CMS system will be more compact, less expensive, and have a higher sample throughput than existing AMS and CMS instruments. To meet these goals, improvements are now being undertaken in the ion source, the injector system, and the cyclotron magnet system. In this paper, these improvements are briefly described and the general operating characteristics of the new machine presented.

## 2. CMS IMPROVEMENT PROGRAM

Figure 1 shows a schematic diagram of the LBL CMS system. To improve the performance over existing devices, changes are being made in the ion source and beam transport and injection line. This will lead to enhanced sensitivity and increased sample throughput. In order to reduce the size, weight, and complexity of the system, the magnetic field of the cyclotron will be produced by permanent magnets rather than electromagnets. These improvements are described below.

The ion source typically used in AMS (and present CMS) is a cesium sputter ion source. A solid sample (graphite for  $^{14}\text{C}$  analysis) is prepared and bombarded by an atomic cesium beam. The resulting ions are then collected and sent to the accelerator. This type of source can produce modest ion currents with modest beam quality. However, sample preparation, i.e. the production of a solid sample, can be laborious and time consuming, leading to low sample throughput. In addition, the many steps in preparing the sample make contamination of the sample more likely.

At LBL, substantial experience has been obtained in developing negative ion sources for fusion and ion implantation applications using magnetic multicusp sources.<sup>10)</sup> In these devices, medium-density, low temperature electric discharge plasmas are used to directly produce negative ions from gas phase precursors. These

sources have high negative ion emission yields and superior beam emittance. Recent experiments have shown that  $\text{C}^-$  can be formed in these sources as well. Figure 2 shows the negative ion yield from a multicusp source using  $\text{CO}_2$  as the precursor gas. As can be seen, a substantial fraction of the ions produced is  $\text{C}^-$ . Further research is underway to optimize this type of source for  $\text{C}^-$  production. If successful, this source will provide a simple to operate, high throughput source of negative ions without the need for complicated processing procedures.

After their production, the ions are transported to the cyclotron analyzer. In a previous instrument, the ions were radially injected. Although this radial injection system was relatively straightforward to implement, it introduced losses to the ion beam. As the sensitivity of the detection depends on the ion current available, it is important to minimize these losses.

One method to possibly improve the transmission of the beam is to use axial rather than radial injection of the ions. Axial injection, in general, is very efficient, and may be simpler than undergoing the several bounces needed for radial injection. To this end, trajectory calculations have been performed to design a spiral inflector, an electrostatic channel which twists or "tilts" as it guides the ions down the axis of the machine and into the midplane. Several designs have been produced. Figure 3 exhibits one candidate, showing one of the two parallel electrodes. All these designs guide the ions to the cyclotron midplane with ion orbits centered on the machine axis, but the deflectors differ in the detailed

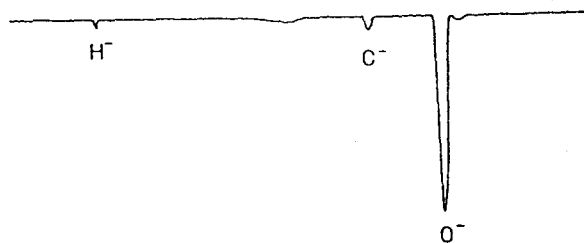


Fig 2  $\text{C}^-$  ion output from a magnetic multicusp source

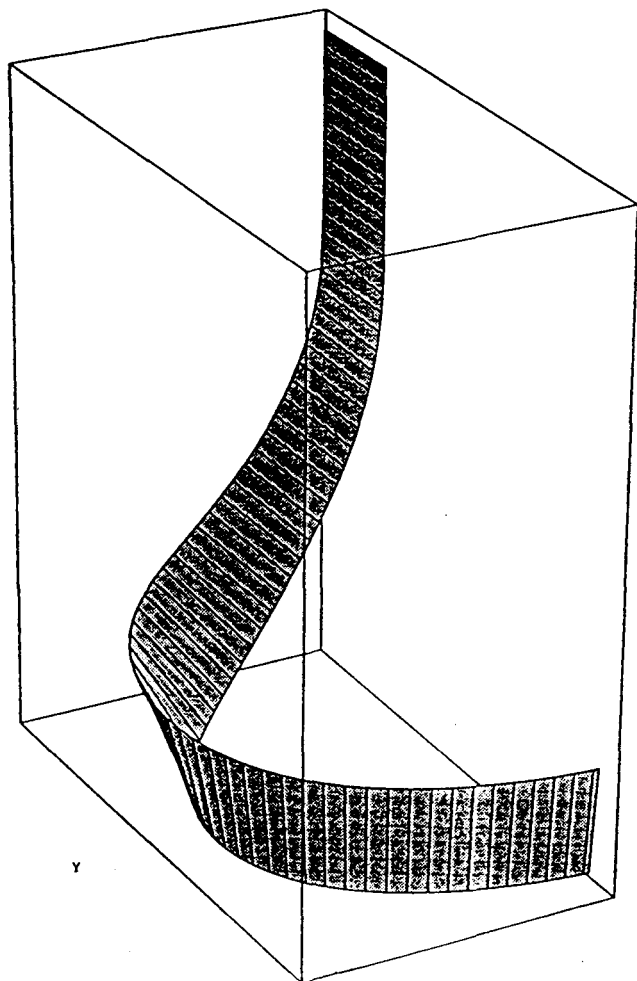


Fig 3 Inflector design for axial injection of ions into the cyclotron.

specification of the tilt angles and electric field. Acceptance calculations are being performed to optimize the design.

The third improvement now being implemented in the CMS will substitute permanent magnets for the existing electromagnet now used in producing the magnetic field. This change has several important advantages. First, the overall size and weight of the magnet structure is reduced, as the magnet coils and power supplies are eliminated. Second, the electrical power and cooling water consumption of the instrument is minimized. With permanent magnets, the CMS will be quite portable and could be placed aboard aircraft, small boats, or out in field locations. It will also reduce operational costs. Using samarium cobalt as the magnetic material, a design for the magnet structure has been produced which has both the required field ( 1 T) and uniformity ( 1 part in  $10^4$ ). Figure 4 shows a view of the

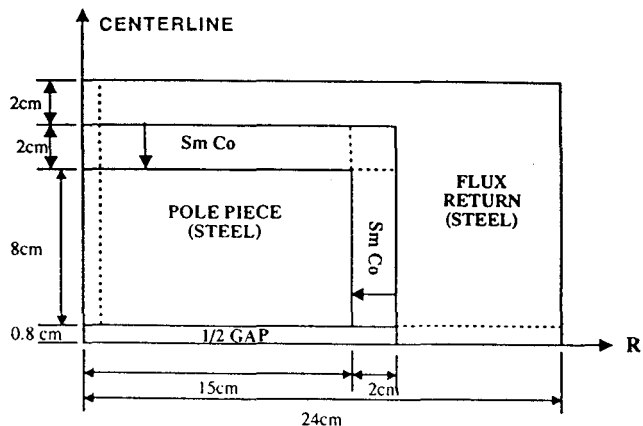


Fig 4 Schematic diagram of permanent magnet system for the cyclotron.

design. This figure shows one quadrant of the machine. The cyclotron has axial symmetry about the centerline. The midplane (marked R in the figure) is a mirror plane, with the exception that the magnet field vectors in the SmCo points away from the pole face in the lower half of the machine. In this concept, the pole faces of the magnet are part of the vacuum vessel. A more detailed description of this design appears elsewhere in these proceedings.<sup>11)</sup>

### 3. SYSTEM CHARACTERISTICS AND PROJECT STATUS

The advanced CMS is being designed and optimized for use with  $^{14}\text{C}$ . Some of the operating characteristics are shown in Table 1. Construction of the permanent magnet structure and the axial injection system should be finished by the end of 1992. Ion source development is occurring concurrently, with integration of the source and cyclotron to take place in 1993.

In addition to these developments, additional improvements are anticipated. In particular, improvements to the beam transport and low resolution mass selector will be made to decrease their size. Earlier devices used a Wien filter for this purpose, but a bending magnet analyzer may be used instead. In addition, advanced accelerator structures may also be used, such as the dee structure proposed by Chen et al.<sup>8)</sup> This advance would allow for an even higher mass resolution in the same cyclotron structure.

### 4. CONCLUSIONS

Cyclotron mass spectrometry (CMS) is a potentially powerful analytical technique with applications ranging from studies of global warming constituents to the biological metabolism of pollutants and pathogens. A

Parameter	Value
Ion source	Magnetic multicusp
Species	<sup>14</sup> C
Injector type	Spiral inflector
Injection energy	5 kV
First orbit radius	4 cm
Extraction radius	12 cm
Extraction energy	< 50 kV
Pole face radius	15 cm
Pole gap	1.6 cm
Magnetic Field	1 T
Field Source	SmCo Magnets

Table 1 Cyclotron design Parameters

development program is now underway which will increase the sensitivity and improve operational characteristics, such as portability and sample preparation, while at the same time reducing the cost of the instrument and its operation. These improvements will make CMS more widely available for routine analysis of trace materials.

#### 5. ACKNOWLEDGMENTS

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