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DETECTION OF RADIOCARBON
IN THE CYCLOTRINO

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ABSTRACT

A small low energy cyclotron (the "cyclotrino"), which was proposed for direct detection of radiocarbon in 1980, has now detected radiocarbon at natural abundance. This device combines the suppression of background through the use of negative ions with the high intrinsic mass resolution of a cyclotron. A high current cesium sputter negative ion source generates a beam of carbon ions which is pre-separated with a Wien filter and is transported to the cyclotron via a series of electrostatic lenses. Beam is injected radially into the cyclotron using electrostatic deflectors and an electrostatic mirror. Axial focusing is entirely electrostatic. A microchannel plate detector is used with a phase-gated output.

Data is presented showing resolution of radiocarbon at natural abundance. In its present form the system is capable of improving the sensitivity of detecting ^{14}C in some biomedical experiments by a factor of 10^4. Modifications are discussed which could bring about an additional factor of 100 in sensitivity, which is important for archaeological and geological applications. Possibilities for measurements of other isotopes are discussed.

1. INTRODUCTION

In response to the increasing demand and high cost of accelerator mass spectrometry (AMS) techniques, the small cyclotron project was begun at Berkeley in 1981 [1]. The basic concept was to combine the excellent properties of a cyclotron used as a mass spectrometer with the capabilities of negative ion sources to reject unwanted backgrounds such as ^{14}N [2,3].

A small cyclotron (the "cyclotrino") was built at Berkeley to test these concepts [4-7]. This cyclotron incorporated a miniature Cs sputter negative ion source at the center of the cyclotron, injecting negatively-charged carbon ions at about 3 keV. Extraction energy was about 40keV at about a 10.5 cm radius. The cyclotron was operated at the 11th to 15th harmonic to obtain the necessary resolution and relied solely on electrostatic focusing for ion confinement. It was found that such a device does indeed produce the necessary resolution for detection of ^{14}C.

Unfortunately, ^{14}C could not actually be detected in the early version of the cyclotron because of the low output of the internal ion source which was used. It was necessary to rebuild the cyclotron to provide for a high-current external ion source. The cyclotron reported here is very similar in design to the earlier machine, with the addition of an external ion source and injection beamline.

Our initial work has emphasized ^{14}C, partially because of the many users interested in this isotope, and partially because the required mass resolution and beam currents are relatively easy to obtain. Measurements of other radioisotopes, such as ^{26}Al, ^{10}Be and ^{3}He are also possibilities with the cyclotrino but have not been pursued.

While the initial impetus for the small cyclotron was archaeological research, biomedical researchers have become interested in this tool as well. In studies of the human body only a limited amount of ^{14}C can be safely given to a patient. For some metabolic studies it is necessary to separate blood into its various constituent fractions and to determine the amount of ^{14}C in each fraction, which may be so small as to contain less than 1 μg of carbon. Decay counting does not yield the required sensitivity for many such studies. A small cyclotron having a carbon current of 10 μA and a detection efficiency of 0.1% would improve the count rate for a 1 μg sample by 5 1/2 orders of magnitude.

2. CYCLOTRON DESIGN

Design of this cyclotron is essentially the same as that described previously by our group [4-10]. A beam of carbon ions is introduced into the cyclotron. The cyclotron's magnetic field and RF frequency are tuned such that only the desired species (^{14}C) is accelerated; other species eventually fall out of phase with the RF and cease being accelerated, never reaching extraction radius. The cyclotrino operates with negative ions to avoid interference from ^{14}N. With ^{14}N eliminated, the nearest interfering mass is the molecular ion ^{13}CH, which is heavier than ^{14}C by a part in 1800. This sets the minimum resolution requirements. The necessary resolution is obtained by operating at a high harmonic (11th to 15th) of the fundamental cyclotron frequency and by using a very flat magnetic field, which causes orbits to be isochronous and hence allows ions to make many turns (50 to 100) in the cyclotron. Focusing is purely electrostatic; the flat magnetic field precludes weak focusing. While beam bunching has been suggested and would improve transmission, this machine uses an unbunched, continuous beam.

A 12-inch NMR-type laboratory magnet is used, operated at about 10kG. Beam is injected at an energy of 5 keV with a 2.8 cm radius and extracted at about 40keV with a 10 cm radius. Ions must gain about 260 eV in the first two gap crossings for about half of them to clear an electrostatic deflection channel in the center of the cyclotron [10].

This cyclotron is typically operated with a harmonic of 11 to 15 and a peak dee voltage of 300 to 400 volts. The cyclotron frequency is about 1 MHz with a magnetic field of about 1 Tesla, so the dees must be driven at 11 to 15 MHz at 600 to 800 volts peak-to-peak.

As mentioned above, focusing is purely electrostatic. This provides sufficient verticle focusing to contain most of the beam from the ion source. The magnetic field provides radial focusing with $v_r = 1$. There are no phase oscillations, but there is significant phase bunching. These focusing mechanisms are discussed in detail elsewhere [10].

The cyclotron acceptance is approximately 40π mm-mrad. Extrapolating from estimates of the source emittance at 20 keV, it was estimated that about 80% of the source output should fall within 40π mm-mrad at 5 keV. Thus, with appropriate focusing, nearly the entire beam profile should be accepted. However, beam is accepted over only about a 45° RF phase window due to the focusing and acceleration properties of the cyclotron. Thus, about 12% of the beam is expected to be accepted into the cyclotron.

3. ION PRODUCTION AND INJECTION

3.1 Overview

The cyclotron system is shown schematically in Fig. 1. A beamline transports beam from the ion source to the cyclotron. An einzel lens captures and slightly focuses the highly divergent output of the ion source. An electrostatic quadrupole lens further focuses and steers the beam into the object slit of a Wien filter. The Wien filter (a velocity selector) performs the dual purpose of reducing the ion load entering the cyclotron (including elimination of the high energy tails from ^{12}C and ^{13}C) and allowing ion source output to be monitored. It focuses the mass 14 beam from the object slit to an image slit, deflecting the mass 12 and 13 ions to a Faraday cup to provide normalization for ion source output fluctuations. The mass 14 beam (containing ^{14}C and molecular ions such as ^{13}CH) is focused and steered into the cyclotron with a combination of four electrostatic quadrupole lenses. The beam is injected into the cyclotron radially using electrostatic deflection channels and an electrostatic mirror.

3.2 Injection details

The injection beamline is shown in Fig. 2, with the approximate beam envelope shown in Fig. 3.

A Cs–sputter negative ion source is used (General Ionex model 860), capable of more than 20 μA of C^- output using graphite or CO_2. The source is designed to operate with about 20 keV extraction energy. However, this machine is designed to have only a 5 keV input beam energy; thus, beam properties were not well known. Extrapolating from performance at 20kV, estimates were that at 5 keV the beam should have an emittance of about 40π mm-mrad with an apparent origin over a region of about 1 mm diameter at about the actual sample location. These estimates are probably not extremely accurate, since there is significant loss of beam before the exit of the Wien filter. An axially asymmetric lens similar to that of Riddle [11] and Drummond [12] was designed in an attempt to reduce the spherical aberration present in axially symmetric designs. This lens seems to work fairly well, but a significant amount of aberration seems to be introduced by the combination of the ion source and einzel lens.

Good control is needed of beam characteristics (beam position, beam width, and focal plane locations) on injection into the cyclotron. A combination of four electrostatic quadrupole lenses provides the needed flexibility. By independent adjustment of the powers of the four lenses, control is gained over the four degrees of freedom associated with beam focal properties. (These four degrees of freedom may be described as the position and size of the beam focus for each of the two dimensions orthogonal to the beam direction.) Beam steering may be accomplished by driving the quadrupoles asymmetrically; this allows adequate control of the four additional degrees of freedom associated with beam position. Custom-built power supplies allow independent adjustment of the focal power and vertical and horizontal deflections of the quadrupole lenses [10].

A Wien filter (also known as a crossed-field separator or a velocity selector) is used in this system to pre-separate the incident ion beam, reducing the amount of ^{12}C which is injected into the cyclotron itself. A Wien filter is composed of an electric and a magnetic field which are orthogonal to one another. The ion beam travels along an axis orthogonal to both. The electric and magnetic fields are adjusted to pass the desired species undeflected; any species with incorrect velocities are deflected away from the main path.

The Wien filter in this system has been built with samarium cobalt permanent magnet material and a soft steel yoke, placed entirely in vacuum [10]. The magnetic field is 4.2 kG over an effective length of 12.0 cm, with a width of about 4.7 cm and a gap of about 2 cm.

The electric field is produced between parallel plates inserted in the magnet gap, and with guard electrodes near the magnet poles [10]. The electric potentials are applied asymmetrically in this design, with one electric field plate grounded and the other operated at a high negative voltage (about 2500 V). This slows the incident 5 keV beam significantly, shortening the necessary length of the Wien filter since ions spend more time in the crossed field region.

The ion beam is injected into the cyclotron using a novel radial injection method [13]. This was used because of concern that the more traditional axial injection would cause distortion of the magnetic field near the center and destruction of orbit isochronicity, due to the need to bore a hole through one of the magnet pole pieces. The radial injection system is shown in Fig. 4, with the approximate beam envelope in Fig. 5. Beam crosses

the magnet edge at an angle, providing the proper focusing characteristics for a nearly parallel incident beam to be axially focused to a beam waist at the first dee gap crossing.

After entering the magnet region it is deflected in an electrostatic channel. The beam then curves through a 180° arc, whereupon it strikes an electrostatic mirror at normal incidence. After reflection the beam travels along a circle which is nearly, but not exactly, centered on the cyclotron axis. A final electrostatic deflection channel near the center of the cyclotron shifts the beam orbit slightly to center it and to provide clearance between the electrostatic mirror and the beam's first orbit.

4. ION EXTRACTION AND DETECTION

An electrostatic channel is used to deflect ions out of the cyclotron. The entrance to the channel is located 90° from the dee gap (Fig. 4). This prohibits any low energy ions from entering the channel by drifting along the dee gap. The first order optical effects of the channel and the crossing of the B-field edge combine to produce a beam focus in the axial dimension at the detector's dynode.

A custom microchannel plate detector was designed and built to detect the output ions [14,15]. This detector uses an aluminum oxide conversion dynode which is impacted by the extracted ions at glancing incidence. A number of secondary electrons are liberated from the aluminum and pulled into the microchannel plate. This should allow discrimination between the 30-40 keV output ions and any low energy ions or photons, which will release fewer secondary electrons.

A large number of photons, presumably UV, seems to be produced in the injection channel. This is probably due to secondary electrons which are produced in the channel by incident ions, gain energy from the electric field, then strike surfaces at high velocity. Copper plated optical shields were installed to block and absorb these photons. Unfortunately, a significant amount of UV still strikes the detector. The tail of the UV-induced single electron distribution extends well into the distribution for high energy ions. Because of this it was necessary to reduce the angle at which ions strike the dynode from 12° to about 6°, shifting the peak of the high energy ion distribution from about 10 secondary electrons to about 17 secondary electrons. This unfortunately reduced the projected area of the dynode by a factor of two, making alignment much more critical and causing some ion loss at the detector.

In a further attempt to reduce the interference of UV photons, pulses from the detector were gated in time. Pulses were only accepted if they fell into a phase window of about 90° width. This reduced the number of UV photons by about a factor of four, while reducing the number of high energy ions by about 25%.

5. SYSTEM PERFORMANCE

5.1 Transmission

System losses are illustrated in Table 1. The transmission of the most complex part of the system (from the output of the Wien filter to the extraction channel of the cyclotron) is quite good, at about 2.5%. A factor of 8 loss in the cyclotron would be expected due to phase acceptance alone. (Only ions with a phase between about 15° and 60° have enough energy gain and focusing to reach extraction radius.)

Table 1
Transmission losses in cyclotron system.

From:	To:	Loss Factor
ion source	Wien first slit	4*
Wien first slit	Wien second slit	5
Wien second slit	injection channel	1
injection channel	mirror	2
mirror	inner deflector	2
inner deflector	extraction radius	10
extraction radius	detector	5
phase discrimination		1.3
amplitude discrimination		4
Wien output	extraction radius	40
Wien output	detection	10^3
ion source	detection	2×10^4

* estimated

The overall efficiency of the system is somewhat lower than expected, with unexpected losses seen at each end of the system. Loss at the beginning of the beamline is probably due to aberrations (and possibly some mispositioning) of the einzel lens. It

should be possible to improve this performance by re-designing the einzel lens on the basis of detailed emittance measurements or computer calculations, yielding a transmission increase of a factor of 5 to 10.

There is also unexpected loss in ion detection due to the discrimination of ions from UV photons. The loss from extraction radius to detector is larger than expected; a factor of two would be expected [4] while a factor of five is seen. This is probably due to part of the beam missing the dynode. (The dynode should be able to collect the entire beam; this loss is probably a result of minor misalignments and uncertainties in beam focus position.) A factor of four loss is due to setting a high discrimination threshold.

It may be possible to reduce these detection losses by a factor of four or so by extending the size of the dynode and changing the position of the microchannel plate (to present a larger projected area to the beam) and placing the dynode at more of a glancing incidence (to produce more secondary electrons from each incident ion). It is also possible to add a beam buncher. This has been suggested for this system [8] and has been investigated in detail by Karadi [10]. A beam buncher should yield another factor of four increase in efficiency. Making all of these improvements (einzel lens, beam buncher, and detector modifications) should boost system efficiency by two orders of magnitude.

5.2 ^{14}C Measurements

A sample of modern carbon prepared by Erv Taylor from the OX-1 standard was measured in the system (Fig. 6). The ^{14}C peak is easily identified. The shape of the peak was taken from an enriched sample of ^{14}C (Fig. 7). At the ^{14}C peak, the count rate was about 0.2 counts per minute with an ion current of roughly 10 μA.

Data from the sample of enriched carbon is shown in Fig. 7. This sample had been enriched roughly 15 times above modern levels in a reactor, courtesy of Frank Asaro and Helen Michel. The enrichment of this sample allowed a more efficient detection setting than that in Table 1; the amplitude discrimination setting was lowered, giving a loss factor of about 1.5 rather than 4. The measured count rate for this sample was actually higher than expected by about a factor of 6. It is not completely understood why this is so; it is possible that the enrichment was higher than expected, and it may be that other loss factors were reduced from the values in Table 1 due to the lower ion currents used for the measurements of the enriched sample. At the peak, the count rate was about 15 per minute with an ion source output of roughly 4 μA.

6. APPLICATIONS AND IMPROVEMENTS

The cyclotrino offers great potential for biomedical research. Samples with enriched levels of ^{14}C can be easily measured with a 10^4 increase in sensitivity over that of conventional scintillation counting. This conclusion is based on measurements with the OX-1 standard; a factor of 5 enrichment over natural abundance would give a count rate of about one count per minute. Since a factor of 7×10^4 enrichment is necessary to give 1 decay per minute for a 1 μg carbon sample, this represents an improvement of four orders of magnitude over traditional decay counting.

Though the device has very good sensitivity for biomedical studies, at present it is not competitive with tandem accelerators for archaeological or geological measurements. However, with improvements in the einzel lens geometry to better match beam from the ion source to the Wien filter, a factor of 5 to 10 increase in efficiency should be attainable. With improvements in the detector system a factor of about 4 should be attainable. The addition of a beam buncher should add another factor of 4. Thus, count rates of 5 per minute without a buncher, or 20 per minute with a buncher should be attainable from modern carbon. This would make the system a practical tool for archaeological and geological applications.

In order to make high precision measurements, a rapid sample changing method is necessary to cancel errors due to small changes in the transmission and detection efficiency of the system. This could be provided with a rotary sample changing apparatus as is used with many tandem accelerator mass spectrometers. Rapid sample changing could also be done by making measurements with CO_2 gas rather than graphite; CO_2 could be injected into the ion source through a porous plug and sample switching could be done by simply switching gas sources.

It would also be possible to measure a number of other isotopes with the cyclotrino [10]. Tritium should be measurable. The nearest interfering mass is different by a part in 500, so the resolution requirements are much more relaxed than for carbon and are easily attained. It will be much lower in abundance than is ^{14}C, which may lead to some difficulties with backgrounds or count rates. If the resolution of the cyclotron can be increased by a factor of about 10 by operating at a very high harmonic (>50), ^{10}Be may be measurable, though operation at such a high harmonic will greatly reduce system

transmission. It should be possible to measure ^{40}K and ^{44}Ti by running at about the 31st harmonic, if there is any interest in studying these isotopes by AMS. But the most promising isotope other than ^{14}C seems to be ^{26}Al. The abundance of the nearest interfering species (^{25}MgH) should be relatively low. The cyclotron resolution would have to be doubled to measure ^{26}Al; this can be done by operating at about the 25th harmonic. Production of a beam of Al⁻ ions is problematic [16]; it seems that detection of ^{26}Al should be easily achieved if adequate ion beams can be produced.

7. CONCLUSIONS

A small low energy cyclotron similar to that of Welch [4] has been combined with a high current external ion source, a beamline, and radial beam injection to detect ^{14}C at natural levels. The system's overall efficiency is about 2×10^{-4}. In its present form, it would be quite useful for measurements of samples enriched in ^{14}C; it could improve the sensitivity of detecting ^{14}C in 1μg biomedical samples by four orders of magnitude over conventional scintillation counting techniques.

Improvements in system efficiency by a factor of 100 should be possible by redesigning the optics of the einzel lens, adding a beam buncher, and optimizing the detector geometry. With these modifications the system would be well suited to archaeological and geological applications.

The system could also be used for measurements of radioisotopes other than ^{14}C. With minor modifications, ^{26}Al and ^{3}He should be detectable. With further modifications, ^{10}Be and other species may be measurable as well.

A small cyclotron such as this has great potential as a mass spectrometer. It offers a huge advantage over decay counting methods for small samples. With efficiency improvements, it would be able to do archaeological and geological measurements which are now done by tandem accelerators, yet would require a small fraction of the space, construction costs, and operating costs.

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1. FIGURES

Figure 1 Cyclotron system schematic, showing injection beamline. (XBL 874-1819)

Figure 2 Cyclotron system, showing injection beamline. (CBB 888-8200)

Figure 3 Approximate envelope of ion beam along beamline. The radial envelope (in the plane of the paper) is shown above, while the axial envelope (the plane out of the paper) is shown below. EQ stands for electrostatic quadrupole lens. (XBL 8911-4210)

Figure 4 Photograph showing the components of the cyclotron. (CBB 888-8210)

Figure 5 Approximate envelope of ion beam in cyclotron. The radial profile is shown above and the axial below. (XBL 8911-4174)

Figure 6 Carbon of modern age. Operation is at the 15th harmonic with 400 V peak RF. RF frequency offset from the ^{13}CH peak is along the horizontal axis. The vertical axis is the normalized count rate, expressed as the counts per coulomb of ^{12}C measured at the output of the Wien filter. (XBL 8911-4185)

Figure 7 Enriched sample. Operation is at the 15th harmonic with 400 V peak RF. RF frequency offset from the ^{13}CH peak is along the horizontal axis. The vertical axis is the normalized count rate, expressed as the counts per coulomb of ^{12}C measured at the output of the Wien filter. (XBL 8911-4186)

EQ = ELECTROSTATIC QUADRUPOLE LENS
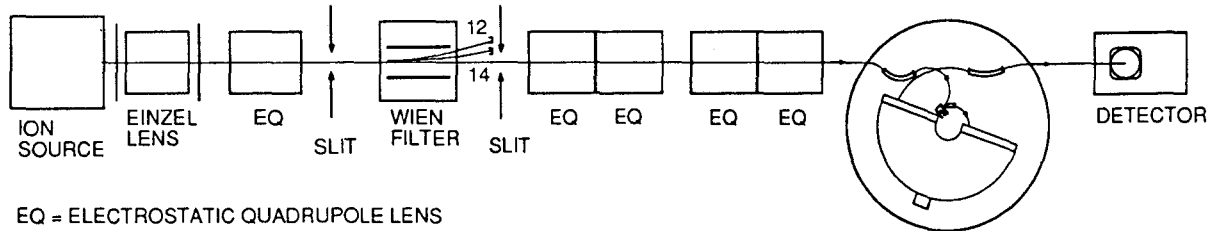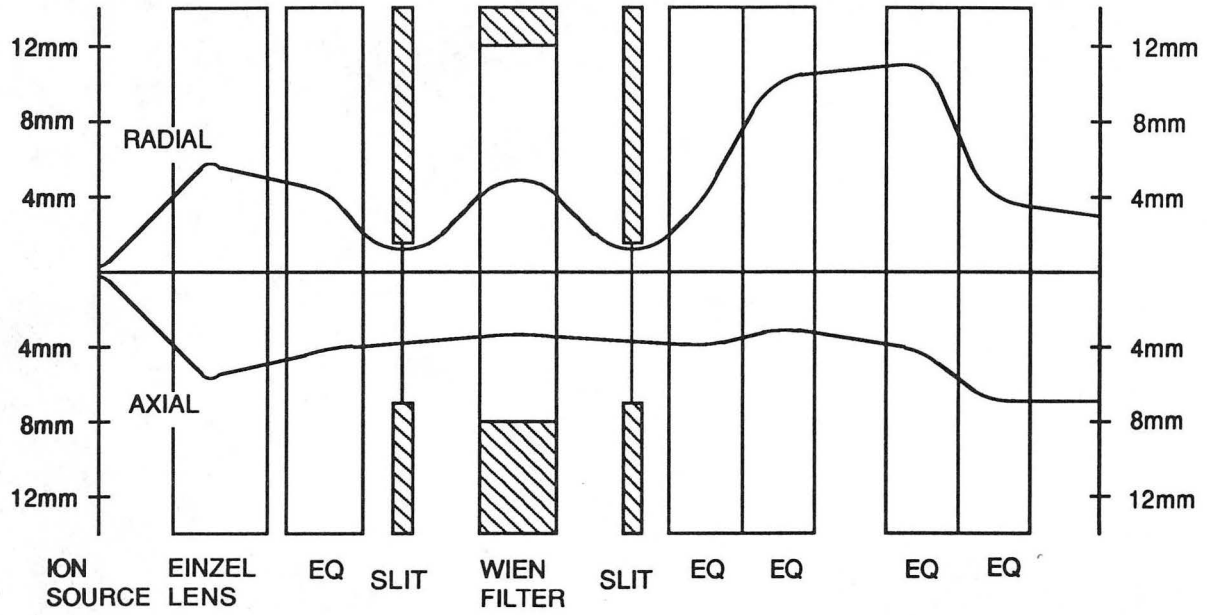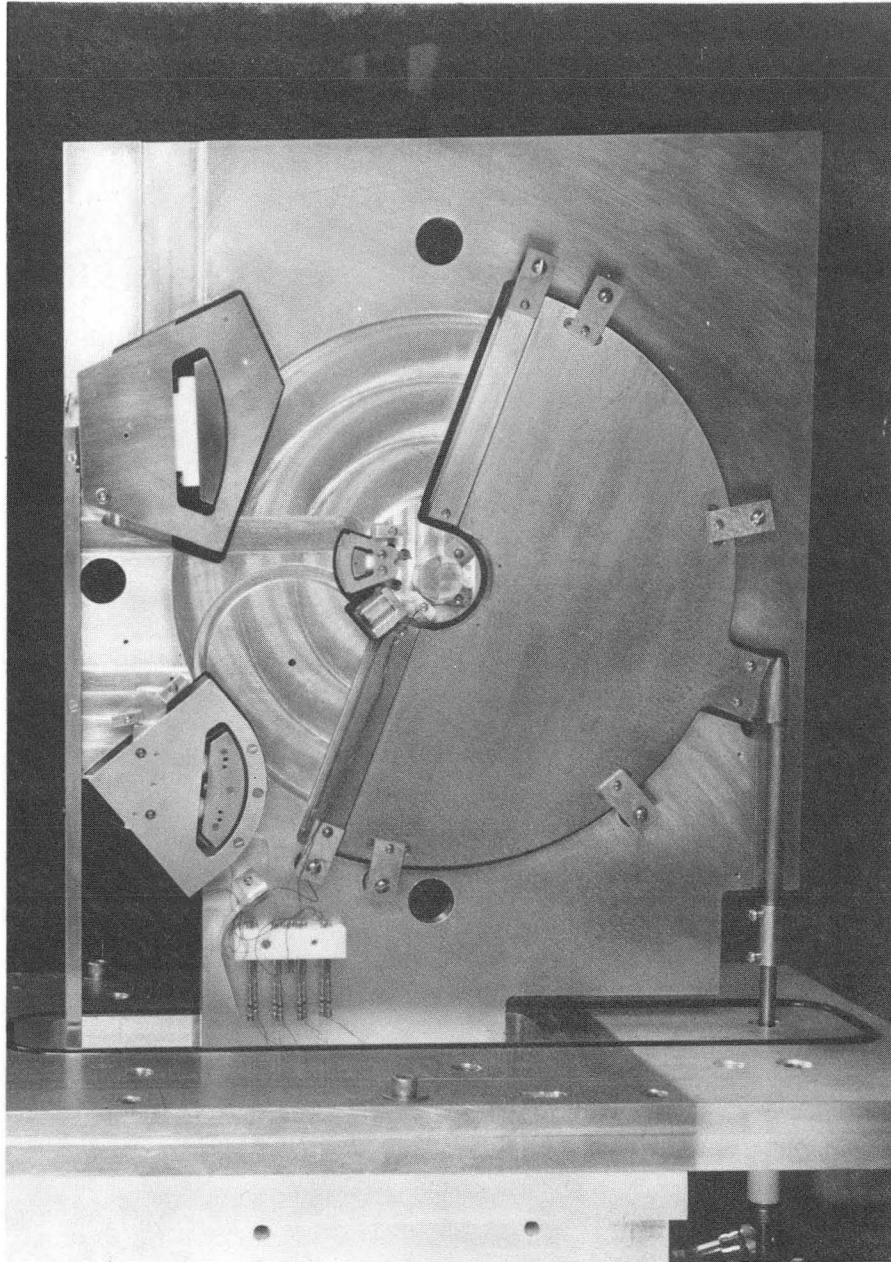
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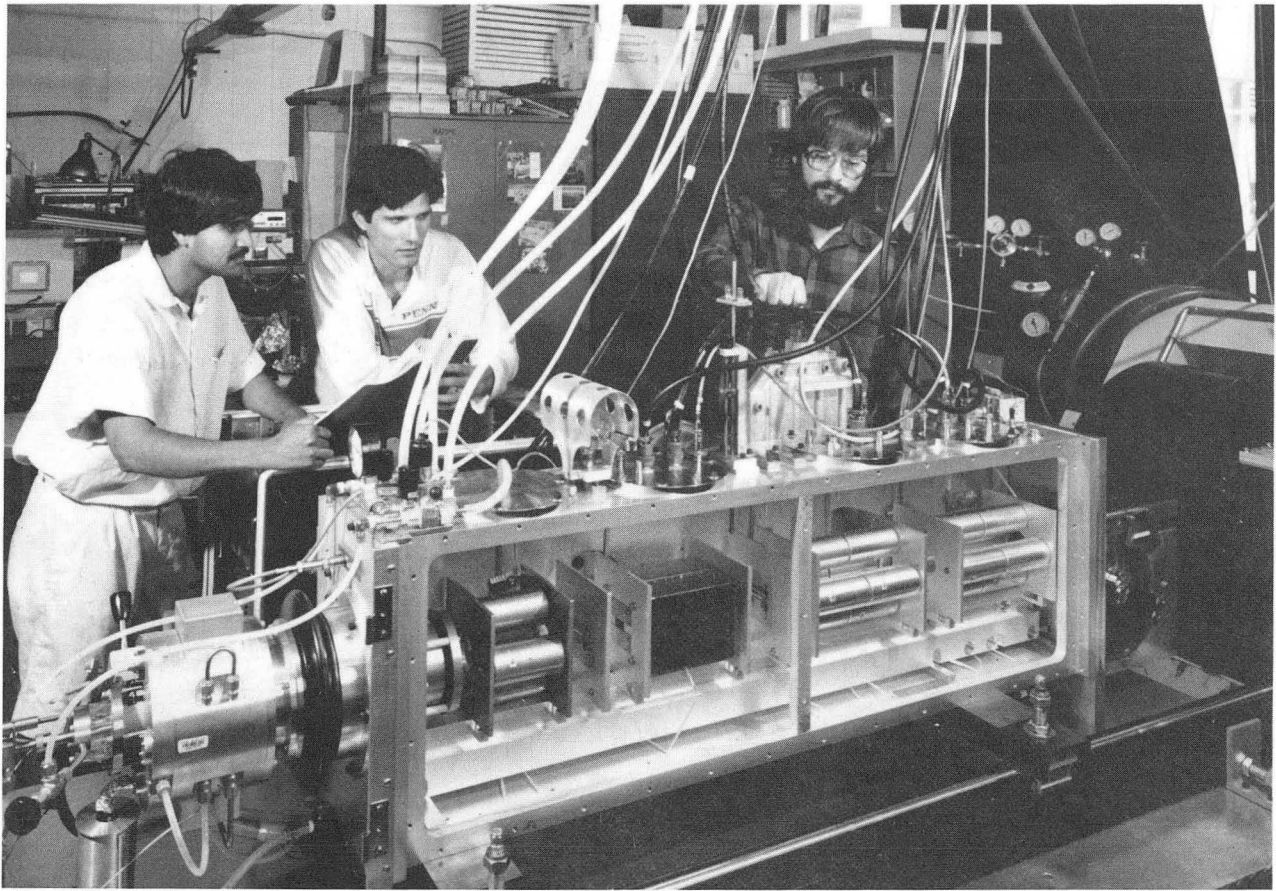
Figure 1

CBB 888-8200

Figure 2

BEAM ENVELOPE IN BEAMLINE

XBL 8911-4210

Figure 3

CBB 888-8210

Figure 4

BEAM ENVELOPES IN CYCLOTRON

Figure 5

XBL 8911-4174

OX-1 Standard
15th Harmonic, 400 V Peak

XBL 8911-4185

Figure 6

Enriched Sample
15th Harmonic, 400 V Peak

XBL 8911-4186

Figure 7