CYCLOTRONS AS MASS SPECTROMETERS*

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Summary

The principles and design choices for cyclotrons as mass spectrometers are described. They are illustrated by examples of cyclotrons developed by various groups for this purpose. The use of present high energy cyclotrons for mass spectrometry is also described.

Introduction

Particle beams from cyclotrons have been used primarily for studies of their interaction with target nuclei and atoms, and for applications such as production of isotopes formed in these interactions. A different kind of application is mass spectrometry. The cyclotron resonance principle effectively selects one charge/mass (q/m) ion species for acceleration and rejects ions with nearby q/m values. Mass spectrometry has been done both on existing cyclotrons and on specialized machines constructed for this purpose.

The beginning of cyclotron mass spectrometry is probably the well known discovery of ^{3}He in the Berkeley 60-Inch Cyclotron by Alvarez and Cornog[1] in 1938. They took advantage of several fortunate coincidences, including the shimming of the magnetic field for ^{3}He by eddy currents during magnet turn-off, as described by Alvarez[2]. The ^{3}He was identified by cyclotron resonance, dE/dx in an ion chamber and total range - the same techniques which are used today. The abundance of ^{3}He relative to ^{4}He was measured for helium from gas wells and from the atmosphere. The field of accelerator mass spectrometry was reviewed recently by Litherland,[3] and the application of accelerator mass spectrometry to radioisotope dating was reviewed by Muller.[4] Conferences have been held on radiocarbon dating with accelerators[5] and accelerator mass spectrometry.[6]

Principles

The resonance condition for acceleration in a cyclotron field B, tells us that RF angular frequency ω_{RF} is given by:

$$\omega_{RF} = H\omega_p = HBq/m \qquad (1)$$

where ω_p is the particle angular frequency and H is the harmonic number, ω_{RF}/ω_p. Only particles with the correct value of q/m will accelerate without phase slip. Others will slip, and stop accelerating after enough revolutions. In a synchronous magnetic field the change in phase, ϕ, during aceleration, is given in terms of the frequency, f, and frequency error, Δf, by:

$$\Delta \sin\phi = 2\pi\ H\ N_0\ \Delta f/f \qquad (2)$$

where N_0 is the number of turns an in-phase particle would have. Since $\Delta f/f$ is a ratio, it can refer to either particle or RF frequency. The frequency error corresponds to a q/m difference according to Eq. (1): $\Delta f/f = \Delta(q/m)/(q/m)$, or for constant charge $\Delta f/f = -\Delta m/m$. Eqs. (1) and (2) also give the required accuracy on magnetic field, since $\Delta f/f = \Delta B/B$ for constant q/m. An in-pnase particle has $\phi = 0$ or $\sin\phi = 0$. When it slips to $\phi = 90^o$ or $\sin\phi = 1$, it stops accelerating. So we can estimate the frequency change needed to stop acceleration by setting $\Delta\sin\phi = 2$ in tuning from $+90^o$ to -90^o. The resolution, or full width of the tuning range is then, from Eq. (2):

$$R = f/\Delta f = \pi\ H\ N_0 \qquad (3)$$

So the resolution of a cyclotron with 200 turns is about 600 on first harmonic acceleration, and 1800 on third harmonic. In practice R would be decreased due to a finite phase width, but increased due to phase clipping at extraction.

To obtain high resolution we need to increase the turn number, N_0, and/or the harmonic number, H, according to Eq. (3). The turn number can be increased by increasing the energy, or by reducing the dee voltage until the source clearance limit or magnetic field synchronism limit is reached. The harmonic number can be increased in an existing cyclotron by reducing the ion charge state and thus the particle frequency. In most existing cyclotrons it would be expensive to increase the maximum RF frequency. However in a machine dedicated to mass spectroscopy, using low dee voltage, a high frequency RF system and thus high value of H would be easy to design. It is significant that tne energy and radius can be made small and the resolution can still be high if the HN_0 product is large, as we shall see in some of the dedicated cyclotrons.

The beam intensity in a cyclotron is limited by the output of the ion source, the transmission during acceleration, and by deflection efficiency in the case of an external beam. Good transmission requires careful magnetic field design to prevent phase slip losses, adequate transverse focusing, and freedom from serious resonances and field errors. It is much more critical for a design with a large number of turns. An upper limit on beam intensity is reached when the axial defocusing space charge force equals the axial electric and magnetic focusing forces. An approximate relation for this current limit was given by Blosser and Gordon[7]:

$$I = \epsilon_0 A \omega_p \nu_z^2 \times (\Delta\phi/2\pi)(\Delta E/q) \qquad (4)$$

where MKS units are used, and previously undefined symbols are: ϵ_0 = permittivity, A = full beam aperture, $\Delta\phi$ is beam phase width, and ΔE is energy gain per turn. For present high energy cyclotrons, this limit is typically a few milliamps of protons. For small mass spectrometer cyclotrons $\Delta E/q$ is small for high resolution, so the current limit can be in the μA or nA region. Fortunately only small currents are needed to make a mass measurement, particularly if particle detectors are used. But this limit can be important when there is an intense contaminant beam accelerating to partial energy along with the desired low intensity beam.

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Design Choices

In the design of a cyclotron dedicated to mass spectrometry, there are a number of design choices for the ion source, magnetic field, RF system, and extraction and detection systems. The energy can be low, 100 kV to a few MeV, so the size of the machine can be small.

Internal ion sources have been used which are either hooded or open and with or without dc pre-acceleration. The open arc has the advantage of eliminating the lower limit on dee voltage imposed by source structure. But it sets low limits on beam intensity because the source gas fills the acceleration chamber. Also only gaseous feed can be used in an open arc. The hooded arc provides both gas and solid source feed. An external source has the advantage of more space to mount and change solid samples, and the background gas and activity from the cyclotron can be eliminated. In the LBL 88-Inch Cyclotron for example a significant ^{14}C background from the accelerator vacuum tank limited the sensitivity of dating measurements.[4] For source feed the tandem electrostatic accelerators have found that negative ions can solve the background problem of ^{14}N while detecting ^{14}C, since ^{14}N forms no negative ion. The same technique can be used with cyclotrons. The use of negative ions would also eliminate ^{3}He in ^{3}H and ^{36}Ar in ^{36}Cl. Space for a sputtering type negative ion source could be provided by an external source or an internal source with pre-acceleration.

The simplest magnetic field design would be an azimuthally constant field, with or without a radial gradient. A radial gradient field as used in the classical cyclotron, limits the HN_0 product and thus the resolution is limited to about 200, due to phase slip. If we use a uniform field with no radial or azimuthal gradient, we remove the limits on HN_0 and resolution, but lose magnetic axial focusing and have to depend on electric focusing. This is usually considered inadequate beyond about 10 turns since the largest electric term in ν_z^2 at high energy is the phase focusing term:[8] $H \sin\phi/(4\pi N_0)$.

This term is proportional to $1/N_0$, and so decreases to .0056 at $N_0 = 10$ and .00056 at $N_0 = 100$ for $\sin\phi = 45^0$ and $H = 1$. However in a small machine with narrow gaps, a high harmonic number will increase this focusing as well as increase the resolution. If $H = 50$, the ν_z^2 contribution goes up to .028 at $N_0 = 100$, giving $\nu_z = .17$, a typical design value for many cyclotrons. So it appears that a uniform magnetic field can be used, if the phase is shifted to the lagging side for axial focusing. Several cyclotrons utilize this design feature. Electrostatic focusing can also be supplied by putting a positive bias on the sides of the acceleration chamber which the magnetic field crosses, to confine positive ions.

Of course a sector focused magnetic field could also be used for axial focusing. An advantage of this design is the availabiltiy of a valley for injection from an external source. Also more turns could be used since the magnetic focusing continues to be strong, independent of N_0.

The RF system requirement is quite modest, since the cyclotron is small and the dee voltage is low. With low power, wave shapes other than a sine wave can be used. A tapered dee can be used, with large RF width in the center for first turn clearance and small width for lower energy gain and higher turn number at larger radius.

An electrostatic deflector to extract full energy beam is useful to eliminate spurious beams from the detector, and allow the counting of single particles with a channel plate or solid state detector.

Various Cyclotron Designs

The principles and design choices of the previous sections have been used by various groups since 1938 to design and build cyclotrons for mass spectrometry. The following sections give examples of these designs.

No RF, Pulsed Beam, Time-of-Flight

A "Chronotron" was built by Goudsmit[9] in 1948, which used the fact that ion revolution time in a magnetic field is proportional to m/q, the inverse of the frequency relation of Eq. (1). The source is pulsed, and ions follow helical trajectories in a uniform magnetic field over one or more revolutions. They are focused radially to a point above or below the source. Ions of different q/m are separated by their time-of-flight. In an experimental test[10] a resolution of 1000 was obtained at mass 100, with 1 or 2 particle revolutions.

A variation of this design is the time-of-flight mass spectrometer proposed by Hipple and Thomas[11] in 1949, Fig. 1. Here a uniform electric field

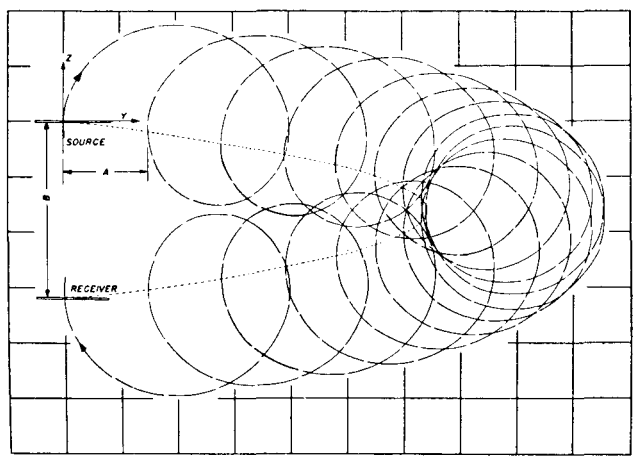

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Fig. 1. Ion path in proposed time-of-flight spectrometer with time varying electric field.[11]

slowly reverses from the +Z to the -Z direction as the ions travel from the source to the receiver. This permits many revolutions of the ions, to give high resolution of q/m by time-of-flight measurement of pulsed beams. The authors also suggest that the electric field could be set to zero for many revolutions at some time in the cycle to increase the number of turns, and the resolution. There is no axial focusing in these time-of-flight spectrometers.

Uniform RF Field, Many Turns

Cyclotron acceleration occurs in an RF electric field which is uniform in space, but only on first harmonic mode. So for high resolution we need many turns. This idea was proposed by Richardson[12] at Berkeley to separate uranium isotopes during World War II. The principle is shown in Fig. 2, and a frequency sweep in Fig. 3. Clear cyclotron resonances were obtained, but only at low beam intensities. At higher intensities, fluctuating plasma electric fields of 10 V/cm interfered with

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the accelerating RF fields of a few V/cm. So a separator would require a very large device with high RF fields. Such a device was not built.

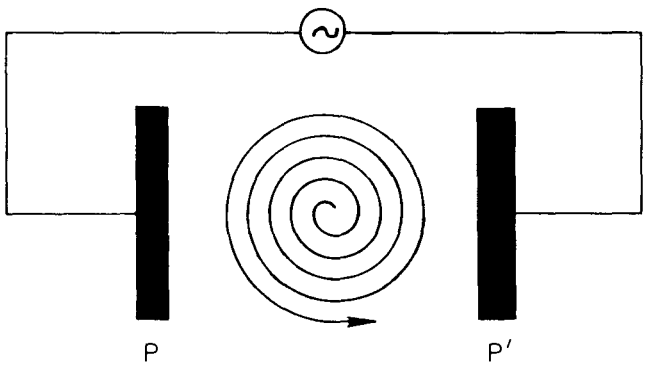

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Fig. 2. Schematic illustration of resonance method for uranium isotope separation proposed by Richardson at Berkeley.[12]

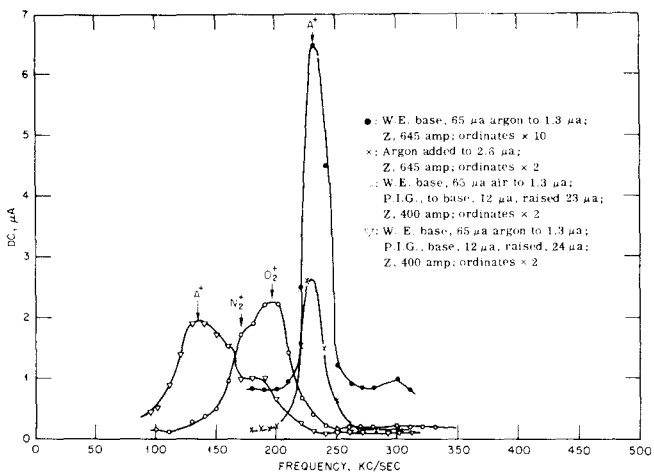

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Fig. 3. Resonance curve data from model for uranium separation cyclotron.[12]

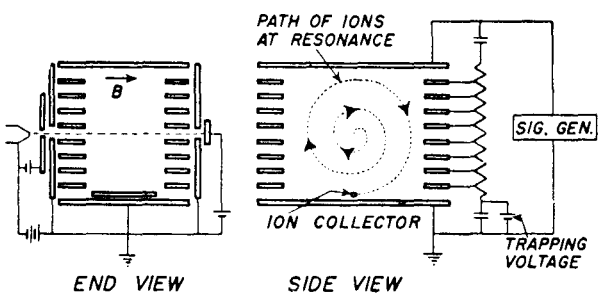

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Fig. 4. Omegatron mass spectronmeter developed at the National Bureau of Standards.[13] Orbit diameter is 2 cm.

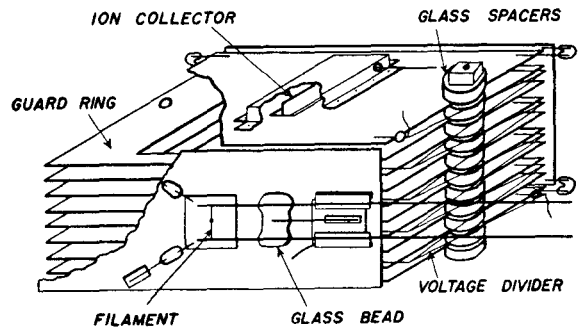

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Fig. 5. Cutaway view of omegatron.[13]

A uniform RF accelerating field for cyclotron mass spectrometry was used again in the "omegatron" at the National Bureau of Standards, described by Sommer, et al. in 1951.[13] This device is shown in Figs. 4 and 5. It consists of an accelerating chamber 3 cm x 2 cm x 5 cm placed in a uniform magnetic field of 5 kG. An electron beam along the magnetic axis forms the ions from the vacuum tank gas. The RF field of .1 V/cm, and 7 MHz is applied across plates 2 cm apart, with voltage divider rings. Proton beam accelerates to a radius of 1 cm and an energy of 1 keV, where it reaches the ion collector. Operating pressure is 10^{-7} torr. Beam currents are small, about 3×10^{-14} A. There are 7000 turns, giving a resolution of 35,000. Axial focusing is supplied by electostatically biasing the divider plates to +.1V, to give a restoring force in the magnetic field direction. Heavier ions can also be accelerated, such as mass 28 with a resolution of 5000. The application of this cyclotron was measurement of the proton magnetic moment, in terms of its cyclotron frequency. The omegatron proved that a high resolution mass spectrometer can be built in a very small volume by using the cyclotron resonance principle.

The omegatron has been further developed by McIver and colleagues at Stanford and Univ. of Calif., Irvine,[14] into a versatile mass spectrometer, now called an Ion Cycloton Resonance (ICR) spectrometer, Fig. 6. The containment time was increased to several seconds, compared to 1 ms in the omegatron, by positive biasing of only the side plates perpendicular to the magnetic field, with a negative bias on the other sides. Also the ions were collected by the entire upper and lower plates, rather than by just a wire, and RF was applied to parallel wires. The ICR spectrometer can be used to study chemical reactions, by pulsing the electron beam current to create ions, waiting for a desired time for reactions to occur, and then pulsing on the RF to analyze the reaction products. Later improvements include frequency sweeps for mass analysis, and detection of ions with pick-up plates, leading to a resolution of 500,000.

An electron cyclotron was used to measure the magnetic moment of the proton in terms of the electron q/m, by Gardner and Purcell of Harvard.[15] The electrons were in a beam crossing a waveguide, which provided the RF field at 9 GHz. The resonance was detected by the change in collected beam current due to beam size change from acceleration. Resolution was 10,000.

High Harmonic, Few Turns

A "Mass Synchrometer" based on three turns of cyclotron deceleration was proposed by Smith at Brookhaven in 1951 and described by Smith and

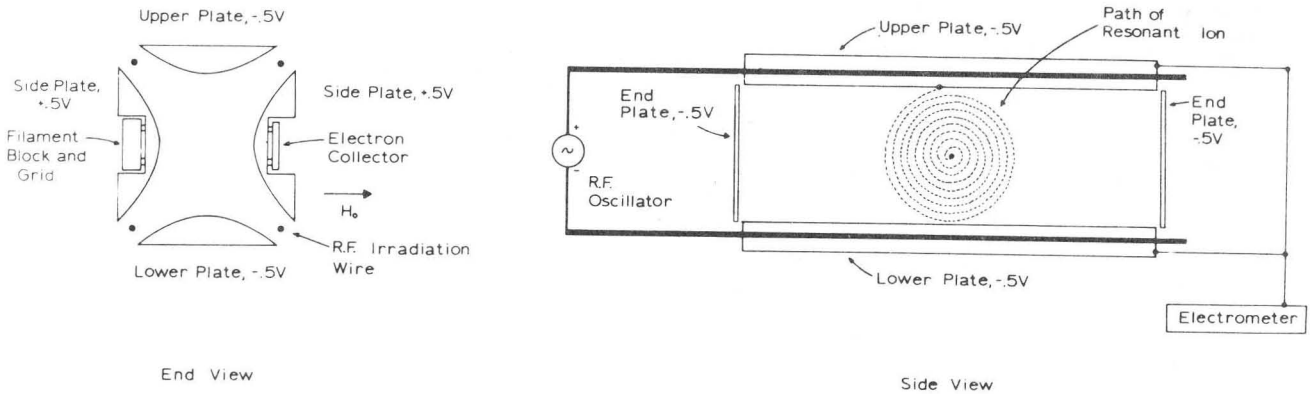

Fig. 6. Ion cyclotron resonance (ICR) mass spectrometer developed from omegatron by McIver at Stanford and Univ. of Calif., Irvine.[14]

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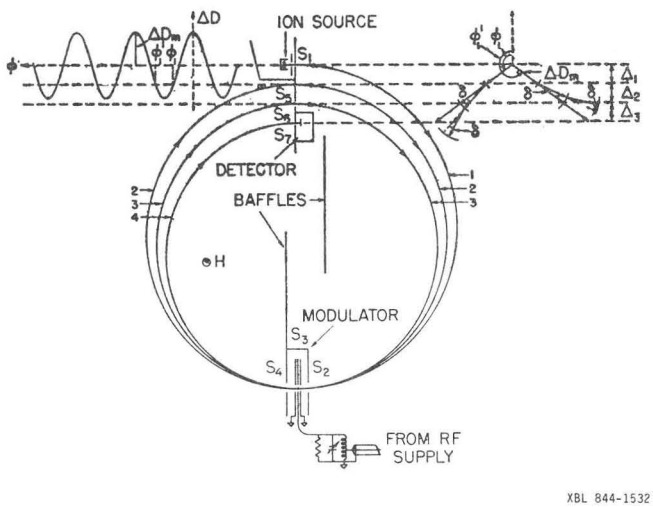

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Fig. 7. Brookhaven mass synchrometer decelerating cyclotron.[16] Diameter of first orbit is 28 cm.

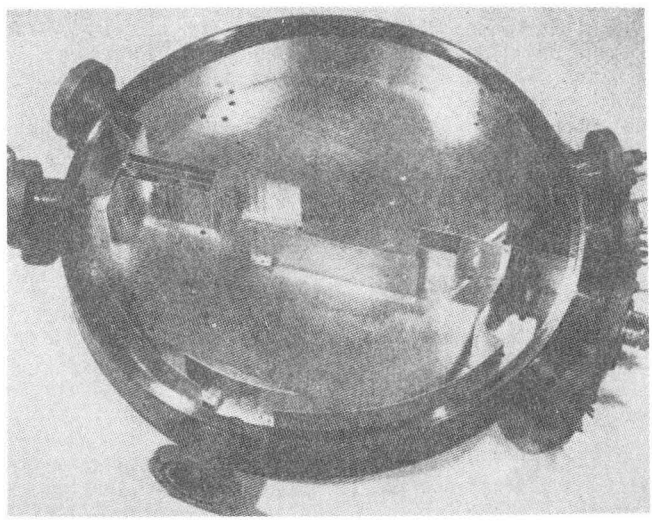

Fig. 8. Brookhaven mass synchrometer.[16]

Damm,[16] Figs. 7,8. A uniform magnetic field was used with a magnet gap of 2 inches. The first orbit size was 11 inches diameter. The decelerating RF voltage of 200 V was applied to a center slit between two grounded slits. The harmonic was about 100, to give R = 25,000. Because of phase selection by slits, the resolution is much larger than predicted by Eq. (3). Ion masses up to 250 could be used. Small currents of 5 x 10^{-11} A were used. No axial focusing was provided, so there was some axial beam loss.

A later version of this spectrometer was built by Smith at Princeton,[17] Fig. 9. This cyclotron has 2 turns, with the selected q/m ion receiving no acceleration. Beam energy was 25 kV. The ion path is a helix with 1% pitch. The orbit is 16 inches in diameter in a 10 kG magnet with a 2 inch gap. RF frequencies of 50-2000 MHz are available. Because of smaller RF gaps of .018 inches, higher harmonic numbers of about 1000 were used, giving resolution of up to 200,000. Beam currents were typically 10^{-11}A.

High Harmonic, Many Turns

This many turn cyclotron begins to look more like our usual picture of a cyclotron. The first example is a decelerating cyclotron described by

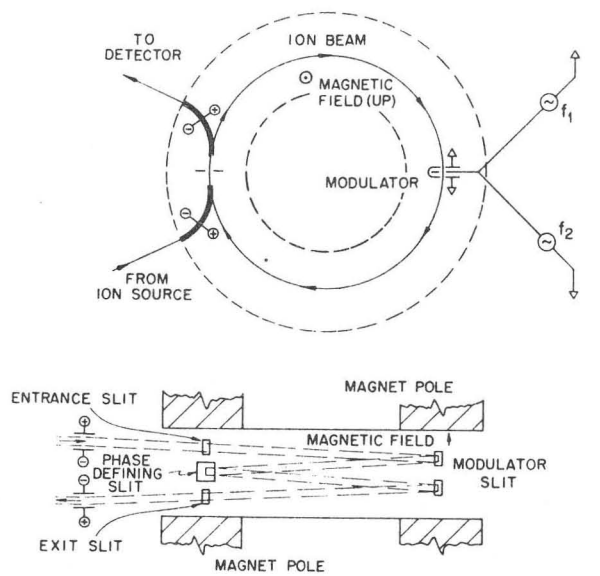

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Fig. 9. New Princeton mass spectrometer.[17] Orbit diameter is 40 cm.

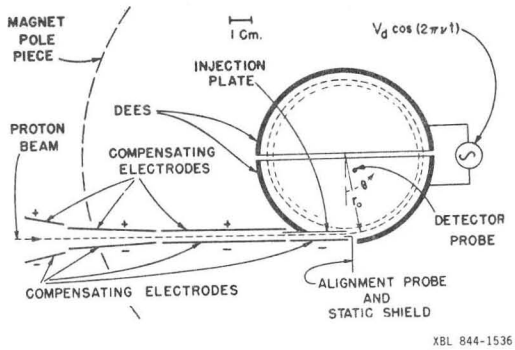

Fig. 10. Decelerating cyclotron designed by Jeffries at Stanford.[18] Outer orbit diameter is 7.6 cm.

Jeffries[18] of Stanford in 1951, Fig. 10. It was designed for protons, to measure their magnetic moment in terms of their cyclotron frequency. The 20 keV beam comes from an external source, is inflected by an electrostatic channel into the outer orbit of 4 cm radius, and then spirals inward to a detector probe at about 15% of injection radius. The magnetic field was uniform to obtain high resolution of 10,000. The axial focusing was provided by electrostatic phase focusing, with the beam running on leading phases for the deceleration mode. RF voltage was 100-300 V, 8-89 MHz, giving harmonics 1 to 11. There were about 500 turns. The maximum turn number was limited by inflector septum clearance, so a very thin septum was used. (.25 mm). Beam currents were 3x 10^{-13} to 10^{-10} A. Jeffries suggests possible improvements of pulsing the dee voltage to clear the septum (although this would reduce the duty factor and beam current), and dividing the dee radially into a high voltage outer section for septum clearance and a low voltage inner section to increase the turn number and resolution.

At Lawrence Berkeley Laboratory a small cyclotron is being devloped by Muller, Welch and colleagues[19] for radioisotope dating research, Fig. 11. This cyclotron is more conventional than those previously described. It has an internal ion source, a beam accelerating to full radius of 10 cm and an electrostatic deflector. Unusual features include a central region cesium source which bombards a graphite target to produce $^{14}C^-$. This avoids ^{14}N contamination, since $^{14}N^-$ is not stable. The $^{14}C^-$ is pre-accelerated with a dc voltage of 3 kV to clear the source components on the first turn. The RF system has a 180° dee with 500-1000 V. For $^{14}C^-$ the full energy will be 36 keV. The magnetic field is uniform for high resolution. Axial focusing is supplied by electrostatic phase focusing, which is strong since the harmonic number is 31. $^{14}N^+$ has been accelerated to full radius and extracted to test the cyclotron. It is expected that with 170 turns and harmonic 31, the resolution should be about 10^4, more than the 1800 needed to resolve $^{14}C^-$ from $^{13}CH^-$.

A recent design for a sector focused cyclotron for mass spectrometry is by Jongen,[20] Fig. 12. This

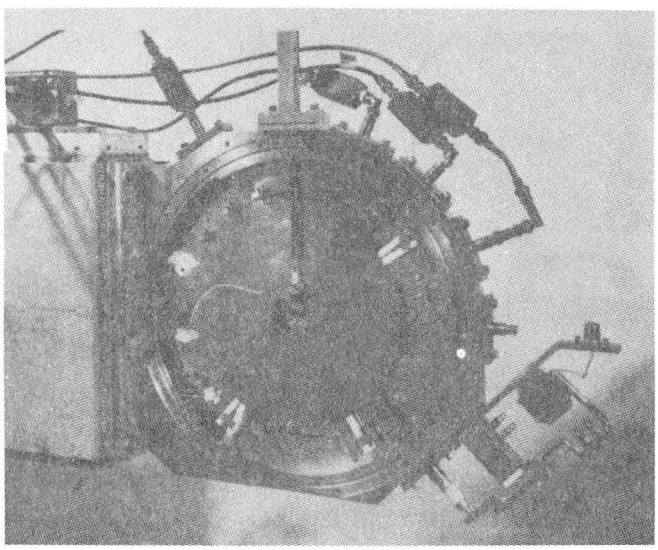

Fig. 11. LBL mass spectrometer cyclotron being developed by Muller and Welch.[19] Orbit diameter is 20 cm.

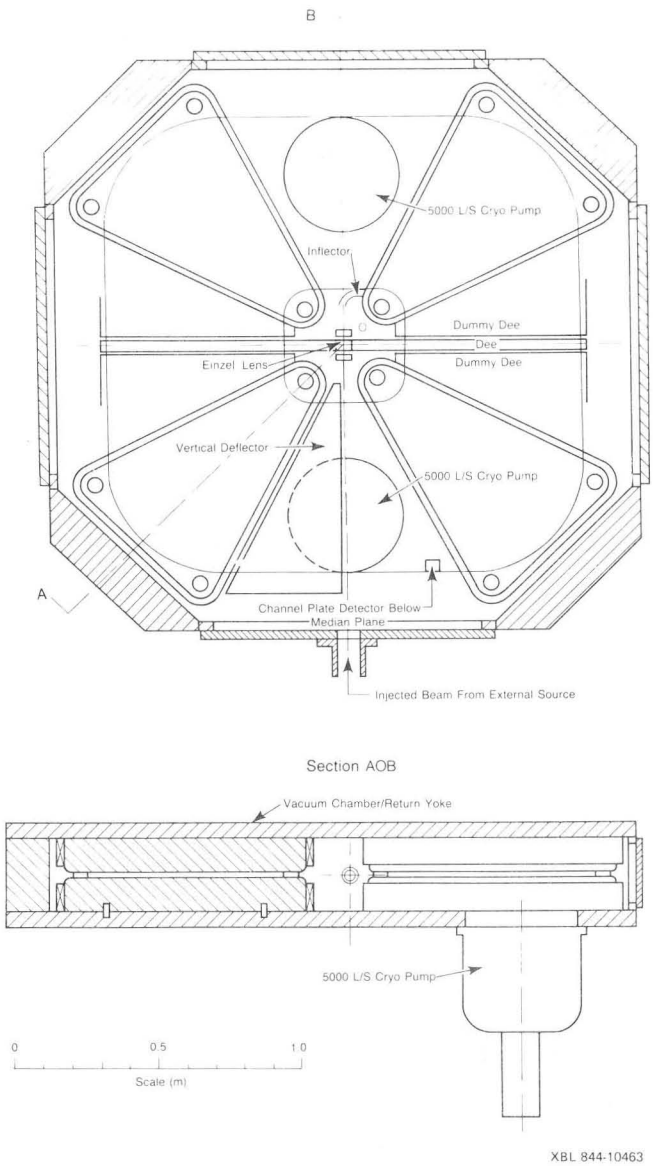

Fig. 12. Proposed mass spectrometer cyclotron of Jongen, Louvain-la-Neuve.[20] Outer orbit diameter is 160 cm.

design uses an external source and injection through a valley. The dee is a narrow strip giving high initial energy gain for inflector clearance, and reduced energy gain at larger radius for increased turn number. The beam accelerates from 20 cm to 80 cm radius. The hill field is 2.5 kG. There are 540 turns with H = 18, giving a predicted R = 30,000. By using pulsed RF amplitude R can be increased to 100,000. Axial focusing is large, with $\nu_z = 1.25$, due to the large magnetic flutter, giving a space charge limited current of 500 nA from Eq. (4).

Present High Energy Cyclotrons

The interest in accelerator mass spectrometry has greatly increased since 1977 when experiments began at several cyclotron and tandem electrostatic accelerators.[3,4] Already in 1973 an Oak Ridge group suggested a search for superheavy elements in the ORIC cyclotron.[21] A search for quarks was done at the LBL 88-Inch Cyclotron in 1977 with 10^{-16} sensitivity.[22] Other cyclotron applications have been described.[23,24] Radioisotope dating experiments were begun in 1977 at the LBL 88-Inch Cyclotron by Muller and his group[25] by dating a ^{3}H sample. For the dating research Muller put together several ideas[4]: using the cyclotron as a mass spectrometer[1,2,22] and counting particles instead of decays. Other isotopes of interest for dating include ^{14}C for formerly living tissue, ^{10}Be for ice and sea deposits and ^{36}Cl for ground water studies. The Grenoble cyclotron was used for ^{10}Be detection with an internal source[26] and with an external source with a quick sample changing design.[27] The ALICE linac-cyclotron system at Orsay has been used for ^{36}Cl detection.[28] Other applications of accelerator mass spectrometry include measurement of reaction cross-sections and long half lives, cosmogenic production and deposition rates of ^{14}C and ^{10}Be, and general trace element analysis. It was the success of these experiments at high energy cyclotrons and tandem electrostatic accelerators that has stimulated the design of the dedicated cyclotrons of the previous section, and of dedicated electrostatic accelerators.

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