Title
HEAVY ELEMENT MASS SPECTROSCOPY WITH THE BERKELEY 88-INCH CYCLOTRON

Permalink
https://escholarship.org/uc/item/2t2567tn

Author
Stephenson, E.J.

Publication Date
1977-08-01
HEAVY ELEMENT MASS SPECTROSCOPY WITH THE
BERKELEY 88-INCH CYCLOTRON

E. J. Stephenson, D. J. Clark, R. A. Gough, W. R. Holley, and A. Jain

August 1977

Prepared for the U. S. Energy Research and Development Administration under Contract W-7405-ENG-48

For Reference

Not to be taken from this room
DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.
HEAVY ELEMENT MASS SPECTROSCOPY WITH THE BERKELEY 88-INCH CYCLOTRON*

E. J. Stephenson, D. J. Clark, R. A. Gough, W. R. Holley, and A. Jain**

Nuclear Science Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720, U.S.A

Abstract

A method is discussed for using the Berkeley 88-Inch Cyclotron as a mass sectrometer for all atomic masses. Low background radiation permits the detection of trace elements with a sensitivity of 1 part in $10^5$ to $10^{17}$, a mass accuracy of $\Delta m/m = 5.8 \times 10^{-5}$, and an abundance accuracy of a few percent. These results are based in part on mass scans using an ion source containing rare earth ores.

* Work performed under the auspices of the U.S. Energy Research and Development Administration.
** On leave from Bhabha Atomic Research Centre, Calcutta, India, with support from the International Atomic Energy Agency and National Research Council.
1. Introduction

Although cyclotrons have been used extensively to produce beams for experiments in nuclear and atomic physics, only on very few occasions have they been employed as mass separators for trace element detection. In this role, the cyclotron was used to show that $^3$He was stable\(^1\). More recently, the Berkeley 88-Inch Cyclotron has been used to search for free, integrally-charged quarks in hydrogen\(^2\), and is being developed for radioisotope dating\(^3\).

The quark search\(^2\) demonstrated the feasibility of light ion mass analysis with a sensitivity better than 1 part in $10^{14}$. We wished to investigate whether the cyclotron could be used to separate and accelerate any atomic mass and identify it unambiguously. We present here the results of several experiments with the cyclotron to demonstrate that mass analysis is possible for all atomic masses, and to illustrate the present levels of sensitivity, accuracy, and mass resolution for the Berkeley 88-Inch Cyclotron. It was expected that, with some improvements, the cyclotron could be used to search for naturally occurring superheavy elements\(^4,5\).

In a mass analysis, a sample to be examined is placed in the ion source discharge, where its constituent atoms are ionized and extracted as a beam into the dee for acceleration. Then, the cyclotron is tuned to accelerate those atoms with a specific charge-to-mass ratio and the amount of accelerated beam is measured. A large intensity range may be covered by measuring beam currents with either a Faraday cup or with a charged particle counter placed directly in the beam line.
Background in the mass analysis was expected to be low. The requirement that the accelerating particles cross the dee gap isochronously for many turns removes from the beam ions of slightly different charge-to-mass ratio (e.g., many molecular ions from atomic ions) and ions created by collisions or charge exchange in the residual gas inside the cyclotron. When intensities permit counters to be used in the beam line to measure ion energy, high charge state atomic ions may be easily distinguished from low charge state molecular ions. The production of molecular ions may be suppressed by the use of high arc power in the ion source. In some cases, the high energy of the emerging beam makes it possible to identify individual ions on the basis of their range or stopping power.

The background in radioisotope dating is more difficult to remove because the radioisotope beam always contains a component from the stable isotope at that mass value. Usually the mass resolution of the cyclotron is inadequate to separate them. The removal of this component by an absorbing medium is being developed\(^3\)). In cases such as \(^{14}C\)-dating where the \(^{14}N^-\) ion is unstable, the use of a tandem accelerator with its negative ion source may prove useful\(^6\)).

2. Experimental method

To achieve the greatest sensitivity in a mass scan, the source and operating condition of the cyclotron must be chosen to maximize the beam current. The configuration chosen depends on the mass sought in the analysis. Searching for light ions is best accomplished with a filament source\(^7\)). A Penning Ion Gauge (PIG) source\(^8\)) is preferred for heavy ions, because it produces the high charge states, or
high charge-to-mass ratios $q/m$, necessary for efficient acceleration by the cyclotron. For even the heaviest masses, the peak production never rises above $q = 6$ (ref. 9), so the acceleration of ions such as $^{238}\text{U}$ with $q/m$ as low as $1/40$ (electron charges/amu) is desirable. Small values of $q/m$ require low particle revolution frequencies during acceleration. On the Berkeley 88-Inch Cyclotron, the limitation in the available dee frequencies is overcome by operating on an odd higher harmonic, i.e., by making the dee voltage reverse an odd number of times while the particles traverse the half turn through the dee. (In this discussion, $h$ will be the ratio of the cyclotron dee frequency to the particle revolution frequency).

The mass analysis of an unknown sample requires the continuous variation of either the cyclotron dee frequency or the magnetic field. With one setting of the radial trim coils, the range over which the frequency or main field may be varied is limited by changes in the relativistic mass increase of the beam or deviations from an isochronous field shape due to saturation effects in the main field magnet (10). For frequency variations at constant magnetic field, this range is given in ref. 10. It is generally better to scan in frequency because the frequency settings are more reproducible than those of magnetic field and because the range of variation becomes large for the higher harmonics. Operation on a higher harmonic generates slow beams, and the relativistic mass increases to be compensated by the radial trim coils is small. In the quark search (2), which was conducted on third harmonic, frequency variations of as much as $50\%$ were possible with a single setting of the trim coils. Operation on fifth or higher
harmonics may be conducted at any frequency with only one setting of the trim coils, since the relativistic mass increase is small enough to be neglected. The expectation that the cyclotron remains in tune during such scans has been confirmed by the observation that intense beams at either end of the scan are efficiently accelerated\(^2\)\).

For a mass scan made by changing the cyclotron frequency at constant magnetic field, the magnetic rigidity of each emerging beam is the same. To the extent that beam steering and focusing are governed by magnetic fields, no changes in beam optics are needed during a scan. However, for the electrostatic deflector at the exit of the cyclotron, the electric force on the beam must remain proportional to the magnetic force, and thus to the ion velocity. Because the velocity at the extraction radius varies in proportion to the cyclotron frequency, the deflector voltage must be continuously adjusted to remain proportional to the frequency. A similar argument may be made for adjusting the peak dee voltage to keep the turn pattern inside the cyclotron the same. However, since the source extraction voltage on the Berkeley cyclotron is the same as the dee voltage, it was often advantageous in the acceleration of very heavy ions to keep the deep voltage at its maximum value to minimize charge exchange losses during acceleration.

Beams can be detected and their intensity measured with the use of either a Faraday cup or charged particle counters. Counters offer the additional advantage of a beam energy measurement that can be related to the charge state of the ions in the beam. Often the energy of intense beams can be measured off frequency resonance. With the lighter ions, the ion range is sufficiently long to permit
counter telescopes to confirm the identification of the beam. Although convenient and easy to use, silicon detectors are subject to radiation damage by intense beams and must be carefully calibrated to give the correct heavy ion energy\textsuperscript{11,12}. The use of counters makes possible the detection of the very low beam currents that give cyclotron mass spectrometry its great sensitivity, and permits discrimination against spurious background radiation. Most spurious radiation originating in the cyclotron is eliminated by placing the counters downstream from a bending magnet.

Through the use of equations that describe the particle orbits and beam energy, the mass and charge of an unknown beam can be determined from the beam energy and the parameters of the cyclotron when the beam is in resonance. A circular orbit inside the cyclotron requires that the centripetal force \( \frac{m v^2}{r} \) be equal to the magnetic force \( q v B \). In a scan at constant magnetic field, all beams are related through the equation:

\[
B = \frac{mv}{qr} = \frac{m}{q} \omega = \frac{2\pi mf}{qh},
\]

where the particle angular velocity \( \omega \) is given by 2 times the ratio of the cyclotron dee frequency \( f \) to the harmonic number \( h \). The harmonic on which an unknown beam is accelerating can be determined from the electrostatic deflector voltage that maximizes the beam current, since that voltage is proportional to the beam velocity, as discussed above. Additional information is provided by an independent measurement of the beam energy in the charged particle counters through the equation (non-relativistic)
\[ E = K \frac{q^2}{m} \]  \hspace{1cm} (2)

The cyclotron constant \( K \) is equal to \( B^2 r^2 / 2 \) where \( r \) is the radius of the beam at extraction.

An accurate measurement of an unknown mass can be made by comparison to a calibration beam of known mass. The cyclotron may be set to the correct \( q/m \) for the calibration beam using the parameter systematics\(^{10}\) for the cyclotron and ion source. Once the beam has been extracted from the cyclotron, its charge state may be confirmed by a measurement of the beam energy and by use of eq. (2). The deflector voltage setting may be used as a reference for future determinations of the harmonic \( h \). After tuning to the unknown beam with only a change in cyclotron frequency, measurements of the harmonic (from deflector voltage), energy, and frequency can be used in eqs. (1) and (2) to solve for charge and mass. The most accurate value of the mass is obtained from eq. (1) when the nearest integer values are used for \( q \) and \( h \), and when the constant value of \( B \) is taken from the parameters of the calibration beam. To obtain the atomic mass, corrections must be made for the electrons missing from the ion and the small relativistic mass increase of the energetic beam. For some studies, precise mass determinations are not needed; e.g., in radioisotope dating\(^3\) or in measurements of the pulse height defect in silicon detectors\(^{12}\).

In many cases, a given mass value can be associated uniquely with some stable isotope of a particular element. In other cases, the element being accelerated may be inferred from a comparison of
several nearby beam currents and the natural isotopic abundances of each element.

3. Experimental results

To test this scheme, a short scan was made using a PIG source doped with heavy elements. A sample containing rare earth silicates was pulverized, mixed with tantalum powder, compressed into a pellet, and placed in a PIG source at the edge of the discharge, opposite the anode aperture. The discharge was supported with argon gas, and a beam of $^{40}\text{Ar}^{2+}$ was used to tune the cyclotron and obtain the value of $B_{\text{used}} \approx n_{\text{eq.}} (1)$. A scan was made from $f = 6.4$ to 7.2 MHz with a heavy ion silicon surface barrier detector placed in the beam line. The frequency was changed continuously with several seconds required to cross the width of any one beam. Beams were detected by observing the individual pulses from the detector on an oscilloscope, so beams with rates much below 1 particle/sec may have been missed. Twenty-six beams were extracted and identified. During the scan the electrostatic deflector voltage was set for seventh-harmonic beams, and most of the detected ions were accelerated on that harmonic.

Because of the open geometry at the cyclotron exit, particles accelerated in anomalous ways can pass into the beam line and may reach the detector. Several such spurious beams were observed during the scan. In each case, the beam was discarded because its current was unaffected when the entrance to the deflector was blocked with the dee probe.

For each beam, the harmonic was determined from the effective deflector voltage $V$. This effective voltage was taken as $E V_i^{2} / S_i$,
where the sum covers each of the two segments of the deflector with
\( V_i \) the segment voltage, \( l_i \) the segment length, and \( s_i \) the mean plate separation for that segment. Fig. 1 is a histogram of the ratio \( f/V \) normalized so that seventh-harmonic beams average to seven. Even though the tails of such fifth- and ninth-harmonic beams could be detected with the deflector set for seventh harmonic, the voltages that maximized the beam current for each harmonic appear well separated. The large magnitude of the separation may be due to changes in the centering of the cyclotron orbit pattern for each harmonic.

The ion charge \( q \) was determined from the energy of the beam measured by the silicon surface barrier detector. For a scan at constant magnetic field, the mass may be eliminated from eqs. (1) and (2) to give the charge as \( q \propto Eh/f \). However, pair production and charge collection inefficiencies make the silicon detector pulse height a function of mass\(^{11}\), with less pulse height for greater mass at the same energy. Departures of 36\% from a proportional relationship between pulse height and particle energy were observed when comparing 27.8 MeV \(^{181}\)Ta with 8.79 MeV \(^4\)He (from the decay of \(^{212}\)Po). Such discrepancies could easily lead to an incorrect assignment of \( q \). To circumvent this difficulty, the observed pulse height was compared with pulse height predictions for each charge state calculated from the formulas of ref. 11, and the ion charge was assigned to the charge state providing the best pulse height agreement. The mass and true energy for each charge state were obtained for the formulas of ref. 11 from eqs. (1) and (2). The pulse height was measured with
respect to the energy deposited by \( \alpha \)-particles from the decay of \( ^{212}\text{Po} \), as prescribed in ref. 11. The locations of the 22 seventh-harmonic beams are shown in fig. 2 as a function of apparent energy and cyclotron frequency. The lines are calculations of the apparent energy using the pulse height defect formula of Kaufman et al.\(^{11}\)). For fig. 2, the \( A \)-parameter of eq. (5) in ref. 11 was fit to our measurements by the value 15.99. The separation of charge states is excellent. The Kaufman formula predicts a slightly greater energy separation than exists in our measurements. Since the energy of each beam is known from the cyclotron parameters to better than 0.5\%, measurements such as these are being used to investigate the pulse height defect of silicon detectors for other experiments\(^{12}\)).

Once the charge and harmonic for an unknown beam were measured, the mass of the ions in the beam was calculated from eq. (1). To obtain atomic masses, corrections were made for missing electrons and the relativistic mass increase of the beam. The resulting atomic masses were very close to the accepted masses for stable isotopes\(^{14}\)), and no ambiguity resulted in the assignment of mass number (shown for the points in fig. 2). Using accepted mass values\(^{14}\)) consistent with the \( Z \)-assignments discussed below, the rms deviation of our measured masses from the accepted values was \( \Delta m/m = 5.8 \times 10^{-5} \). The quality of the agreement for mass serves in most cases as a confirmation of the assignment of charge and harmonic, since a change in those assignments usually leads to a mass that is in marked disagreement with any accepted mass value.
When conducting a continuous mass scan, it is useful to measure the FWHM of the beam intensity as a function of frequency. The ratio of the width in frequency to the scanning speed gives the dwell time at each mass value. This dwell time, together with the beam intensity, can be used to calculate the sensitivity of the scan, as will be discussed later. The fractional width $\Delta f/f$ is determined by the phase acceptance for the beam at extraction radius, and varies as the reciprocal of the number of dee voltage cycles required to accelerate the beam. Since the dee voltage is typically near 60 kV for many beams, the width is given approximately by $\Delta f/f \approx 0.02 q/Eh$, with the beam energy $E$ in MeV. The coefficient in this expression was obtained from measurements made on third harmonic\(^2\). Scans may be conducted close to intense beams because the intensity of the tail is less than 1 particle/sec within $\Delta f/f = 5 \times 10^{-5}$ of the beam center frequency.

The mass numbers and beam intensities observed in this scan were compared with the known isotopic abundances\(^15\) to form a self-consistent list of the elements observed. This list was expected to reflect the composition of the source and insert, which contained light rare earths, metals from the iron family, and various common materials\(^16\). In most cases, at least one isotope of each element could be assigned unambiguously. On seventh harmonic, the $q = 1$ beams were magnesium, aluminum, and silicon, and the $q = 2$ beams were chromium, manganese, iron, and nickel. The $q = 3$ beams were predominantly isotopes of krypton, probably from contamination in the discharge support gas, along with $^{89}$Y and possibly $^{87}$Sr. The missing $q = 4$
beams fell between ruthenium and tin, and were not expected to be
part of the doped source. The $q = 5$ beams were assigned to barium,
lanthanum, praseodymium, and neodymium, with possibly some cerium.
Some barium isotopes were not sufficiently intense to be detected
at our scanning speed. The isotope $^{144}\text{Nd}$ was accelerated on fifth
harmonic. The gap in the scan (see fig. 2) at 6.66 MHz was necessary
because intense beams, most likely $^{56}\text{Fe}^{2+}$ and $^{84}\text{Kr}^{3+}$, prevented leaving
the silicon detector exposed in the beam line. Such gaps could be
covered, with reduced sensitivity, by observing scattered beam or by
collimating or dispersing the beam before it reaches the detector.
The scan was made from lower to higher frequencies, and at 7.1 MHz
(marked by a cross in fig. 2) it was necessary to change the ion source.
It is likely that this change is related to the appearance of $^{131}\text{Xe}$
when other isotopes of Xe were not seen. The intensities of the seventh-
harmonic beams were consistent with the known abundances to within
a factor of 2. Since the scan took about 5 hr, these discrepancies
may have arisen in part from changes in the ion source output with time.
The beams accelerated on other harmonics included $^{146}\text{Nd}^{5+}$ ($h = 5$),
$^{40}\text{Ar}^{2+}$ ($h = 5$), $^{58}\text{Ni}^{3+}$ ($h = 5$), and $^{181}\text{Ta}^{5+}$ ($h = 9$).

Beam intensities may be compared more accurately if the frequency
change from one mass to the next is made quickly. Fig. 3 shows
the relative intensities for Kr beams compared with the known
abundances$^{15}$). These measurements were taken$^{17}$ during a search for
$^{81}\text{Kr}$ as part of the radioisotope dating project$^{3}$). The beam
intensities were normalized so that the sum of the beam intensities
and the abundances would be the same. The agreement is good except
that the lighter elements were seen with more intensity. This reflects a general characteristic of the cyclotron to accelerate lighter ions more efficiently, since their higher velocity makes center region charge exchange losses less severe. If such trends can be measured in advance, beam intensities can be compared with an accuracy of a few percent.

The sensitivity of an analysis for a particular mass is defined here as the fraction of that mass contained in the source feed which would give one count in the detector during the dwell time. The sensitivity is given by the reciprocal of the product of dwell time (seconds) and beam current (particles per second) for that mass if it were to constitute 100% of the source feed material or sample. For trace elements, this beam current must be inferred from beam currents for nearby masses whose concentration in the sample is known. Such interpolations must include the mass dependence of the acceleration efficiency, as demonstrated in fig. 3 for the krypton isotopes.

The general trend of sensitivity with mass can be obtained from typical cyclotron beam currents\(^{18}\). This sensitivity is plotted in fig. 4 for a dwell time of 1 sec, approximately the dwell time in the scan reported here. The line is a guide to the eye. Points from this scan were not included because the beam intensity was not optimized. The value indicated by the open circle for \(^{232}\)Th was measured with poor vacuum and source feed, and does not represent good cyclotron operation. When the trend of fig. 4 is extrapolated to a mass of 300 amu, it appears that the sensitivity of a search there for superheavy elements is about \(10^{-4}\). Better sensitivity for the heaviest masses
has been achieved with tandem accelerators\textsuperscript{19}) even though their sensitivity is limited by background radiation from the accelerator. It is likely that the cyclotron's sensitivity will improve with new machines\textsuperscript{20}) designed with cryopumping in the dees.

The cyclotron is less sensitive for large masses because these slower beams incur greater losses due to charge exchange in the cyclotron center region. Measurements of the internal cyclotron current as a function of cyclotron radius reveal that the $^{40}\text{Ar}^{1+}$ beam intensity decreases by a factor of about 80 during acceleration. These losses vary as $\exp(-x)$, where $x$ is proportional to the product of the charge exchange cross section and the vacuum chamber pressure. The three times larger cross section for thorium\textsuperscript{21}) suggests that the losses for the heaviest masses may be as large as $10^6$. With charge exchange losses accounting for much of the general trend of sensitivity with mass, it is apparent that even small improvements in the vacuum will result in better beam intensity.

4. Conclusions

We have demonstrated that the cyclotron may be operated as a high energy mass spectrometer for all atomic masses. Samples to be analyzed are placed in the ion source discharge, and the cyclotron frequency is adjusted to accelerate sequentially each mass value. The quantity of each mass constituent in the sample is related to the intensity of the emerging beam. Measurements of the energy, harmonic number, and resonant frequency of each beam determine, without ambiguity, the charge state and mass of the beam. A partial mass scan generated a series of beams whose mass numbers and intensities
were consistent with the composition of the ion source and sample and with the isotopic abundance of the elements observed. The accuracy of quantitative measurements of trace element concentrations is limited to a few percent by the stability of the cyclotron and ion source. The mass resolution of the cyclotron is good, permitting scans to be made within 1\% of known stable masses which produce intense beams.

The principal advantage of the cyclotron for mass spectrometry lies in its sensitivity. By using charged particle detectors to count individual ions, light trace elements can be detected at levels as low as 1 part in \(10^{14}\) with only a few seconds of integration time. Because the sensitivity is not limited by background radiation, individual trace elements at levels at least \(10^3\) lower may be detected by using longer integration times. Thus this system is particularly well adapted to finding a few, low-level, exotic species.

This sensitivity is achieved because of the absence of background radiation from the cyclotron. The requirement of isochronous acceleration removes ions created by charge exchange or collisions in the residual gas inside the machine, and the production of molecular ions is discouraged by the use of high ion source arc power. Any remaining background radiation can usually be distinguished from beam by its signature in the charged particle counters.

With the present internal ion source at the Berkeley 88-Inch Cyclotron, the sensitivity for the heaviest masses is limited primarily by beam losses in the center region of the cyclotron. Even small improvements in the vacuum could result in substantial improvements in sensitivity. Additional cryogenic pumping is planned, and an external
ion source with better center region vacuum has been proposed. Sputtering rates for solid samples may be improved by better placement of the sample within the arc or by biasing the sample with a negative voltage. For special problems other ion source configurations favoring greater stability and better accuracy could be built.

The authors are indebted to A. R. Smith for the samples of rare earth ore; to J. B. Moulton and G. J. Wozniak for help with the experiment; to J. T. Walton and F. S. Goulding's instrumentation group for silicon detectors; and to D. L. Hendrie and the staff of the 88-Inch Cyclotron for their support and encouragement.
References


3) R. A. Muller, Science 196 (1977) 489.


5) A. Jain, private communication.


7) K. W. Ehlers, Nucl. Inst. and Meth. 18, 19, (1962) 571.


10) D. J. Clark, R. A. Gough, W. R. Holley, and A. Jain, to be published.


13) Fabricated by ORTEC, Oak Ridge, Tennessee.
17) R. A. Muller, private communication.
18) 88-Inch Cyclotron Available Beam List--1977, distributed by the Nuclear Science Division of Lawrence Berkeley Laboratory (1977).
Figure Captions

Fig. 1. Number of beams as a function of harmonic calculated from the ratio $f/V$ (see text).

Fig. 2. Location of seventh-harmonic beams as a function of apparent energy (pulse height) in the silicon detector and cyclotron frequency. Lines of constant charge were calculated from the formulas of ref. 11. Breaks in the lines indicate areas not covered in the scan. The cross denotes a change of ion source. Atomic mass numbers are noted by most points.

Fig. 3. Relative intensities of Kr beams (crosshatched) compared with the known isotopic abundances (open bars).

Fig. 4. Sensitivity of the cyclotron for mass analysis as a function of mass. Trace elements present at these concentrations could be accelerated into a 1-particle/sec beam. The straight line is a guide to the eye.
Fig. 2
Fig. 3
Fig. 4

The graph shows the relationship between sensitivity and a variable labeled $A$. The data points are plotted on a logarithmic scale, with sensitivity values ranging from $10^{-16}$ to $10^{-4}$, and $A$ values ranging from 0 to 250. The graph includes a best-fit line that illustrates the trend in the data.
This report was done with support from the Department of Energy. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the Department of Energy.