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Search for an Anomalously Heavy Isotope of Oxygen

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A search has been made for an anomalously heavy isotope of oxygen spanning the mass range 20 to 54 amu with use of a tandem accelerator as an ultrasensitive mass spectrometer. An upper limit of heavy hadron to nucleon ratios was established at 10^{-16} over the entire mass region and over a large portion of it at 10^{-18}. This ratio is considerably lower than the predictions of cosmological models, namely $\sim 10^{-10}$ to 10^{-11}.

In a recent Letter, Dover, Gaisser, and Steigman[1] suggest the possibility that new stable heavy hadrons, proposed by several different elementary-particle theories,[2] should be present in $Z > 1$ nuclei at levels of at least $\sim 10^{-10}$. In several specific models, these authors[1] further note that the stable hadron is electrically neutral and isoscalar and its mass is likely to be $\gtrsim 10$ GeV and most probably would be nonintegral (in amu). Since the particle is not anticipated to bind with a single proton to form a heavy isotope of hydrogen, the high sensitivity ($\sim 10^{-18}$ to 10^{-19}) searches of Muller, Alvarez, Holley, and Stephenson[3] and Alvager and Naumann[4] would, assuming its existence, have failed to detect it. The limits on anomalous nuclei of arbitrary mass in elements with $Z > 1$ are much poorer, typically 10^{-6}.[5]

In view of the foregoing it was decided to use our FN tandem accelerator as an ultrasensitive mass spectrometer and to search for an anomalously heavy isotope of some element with $Z > 1$. Although several elements were considered oxygen was chosen because (1) it readily forms a negative ion, (2) the dimer and trimer negative-ion beams are very weak, and (3) by using ^{18}O that had been enriched by gaseous diffusion one might expect to gain a factor of about at least 500 in sensitivity.

Experimental procedure.—Figure 1 shows schematically the modifications that have been made over the past year to our tandem accelerator for such studies. These include the addition of a second sputter source followed by a 90° analyzing magnet, a crossed-field velocity selector, and a removable detector located immediately prior to the image slits of the tandem's 90° analyzing magnet. The source magnet is double focusing with a radius of 30 cm and has a mass resolution $\Delta M/M$ of about 1/50 when the entrance and exit slits are adjusted to accept 90% of the available negative-ion current. The velocity selector is 50 cm long and is located immediately after the high-energy quadrupole magnet about 200 cm before the object slits. The $^{18}O^-$ beam was generated in the sputter source by spraying ^{18}O gas onto the surface of a high-purity titanium cone. The negative-ion current throughout the experiment was about 32 μA.

Preliminary measurements revealed that the most severe experimental difficulty was the extremely high count rate near the integer masses of 27 and above (frequently $> 10^6$ counts/sec). Since calculations based on the tables of North-

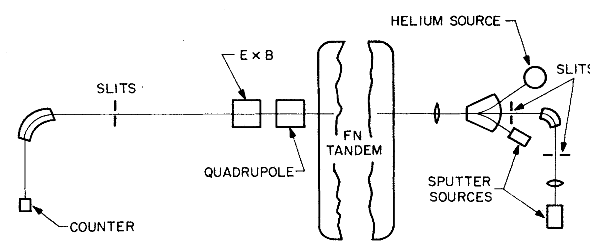

FIG. 1. General plan of FN tandem accelerator as used for supersensitive mass spectroscopy.

cliffe and Schilling[6] revealed that the rate of energy loss and range of oxygen ions, at our energy of 42 MeV (7 MV on terminal, $^{18}O^{5+}$), had a surprisingly low dependence on mass, we decided to use nickel absorber foils to stop the higher-Z ions. This procedure dramatically reduced the background but did not completely eliminate it. Further study of the particles that were detected in our counter revealed that they were frequently oxygen isotopes and consequently the additional complication of an ionization chamber in front of the silicon detector would not be worthwhile. The final detecting system consisted of a foil wheel containing five different nickel foils ranging in thickness from 2.7 to 6.9 mg/cm^2 followed by a 6-mm-diam silicon detector. The size of the latter determined the mass resolution of the tandem's 90° analyzing magnet to be $\Delta M/M \sim 1/300$.

Measurements were begun at mass 20 and between 20 and 30 amu the negative-ion and positive-ion magnets were simultaneously incremented in steps of 0.05 mass units. During each step the total number of counts within a window extending ± 3 MeV of the calculated energy of the anomalous mass oxygen after traversing the nickel foil was integrated over a 48-sec period. In general the crossed-field analyzer was not used since previous studies showed that it introduced some aberrations and attenuated known beams by about 30%. However, whenever a peak was observed with a few hundred counts, the region was rescanned with the crossed-field analyzer adjusted to transmit the appropriate mass. A particularly useful contaminant peak in this region was ^{23}Na—providing about 10^4 counts/sec—and frequent use was made of it to check the proper functioning of the many components of experimental equipment. This peak also proved invaluable for checking the integrity of the tandem's generating voltmeter which was used to hold the terminal potential at precisely 7 MV. During the scan, periodic calculated adjustments were made to the high-energy quadrupole magnet.

The region between mass 30 and 40 was scanned in a similar fashion but, due to a reduction in mass resolution of the 90° positive-ion magnet, the mass step was increased to 0.067 amu. Mass 40 to 50 was likewise scanned but in steps of 0.1 and 50 to 54 in steps of 0.2 amu. Measurements were curtailed at this point because of the onset of saturation in the positive-ion magnet but a crude continuous search to 60 amu provided an upper limit of 10^{-13} for the heavy hadron to nucleon ratio.

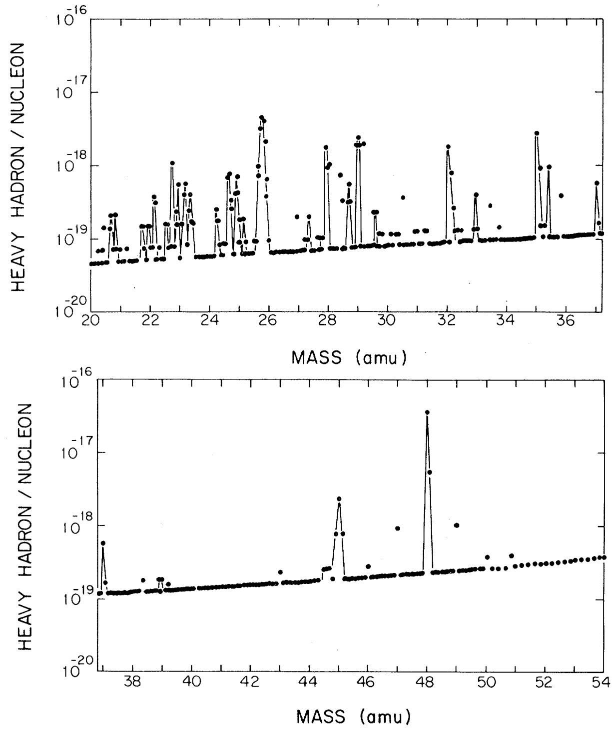

FIG. 2. The upper limit for heavy hadron to nucleon ratio is plotted vs mass. The ratio assumes a 500-fold enrichment and a transmission efficiency which decreases with mass, causing the rising baseline. The "peaks" are principally due to knockout ^{16}O from an oxide layer on the Ni foils. Ten counts at mass 35 correspond to a limit of approximately 10^{-18}.

Results and discussion.—Figure 2 shows a logarithmic plot of our results. The abscissa represents the mass in amu's and the logarithmic ordinate is the upper limit for the ratio of anomalously heavy hadrons to nucleons calculated with the assumptions: (1) an $^{18}O^-$ beam intensity of 30 μA, (2) the ^{18}O gas was enriched at least 500 times in any anomalously heavy oxygen, (3) the transmission through the tandem for $^{16}O^{5+}$ at 7 MV utilizing a carbon-foil stripper is 20%, as experimentally verified, and (4) the stripping efficiency falls off with the velocity with which the ion arrives at the terminal.

It is evident from the figure that the number of counts never exceeded a heavy hadron to nucleon ratio of 10^{-16}, which is well below the level of 10^{-10} suggested by Dover *et al.*,[1] and over much of the region is below 10^{-18}. Many of the "peaks" occur at integral masses and can readily be explained. For example, the strong peak at mass 48 arises because of $^{48}Ti^{5+}$ ions striking the nickek foil and knocking out ^{16}O from a surface oxide layer. The latter pass through the foil and produce a very intense low-energy peak which tails into the ± 3 MeV integrated energy window because of inefficiencies in the pileup-rejection circuitry. Other peaks, such as the one at mass 45 have rather subtle explanations and are, in part, a result of coincidence. In this particular case $^{27}Al^{18}O^-$ ions, selected by the negative-ion magnet, are accelerated to the terminal where some of the ^{18}O are stripped to 5^+ and emerge from the tandem with an energy of 37.8 MeV. A small fraction of these charge exchange to 3^+ ions before the tandem's 90° magnet and have almost exactly the same magnetic rigidity as mass 45 ions of charge 5^+ and energy 42 MeV.

Although we feel that the experiment could be refined and the sensitivity improved to 10^{-19} or better, we doubt that this would be worthwhile. Oxygen, though perhaps a good choice on physical grounds, may be a poor choice chemically since free oxygen on earth has undergone considerable chemical and biological cycling by processes with rates which depend on mass. It is possible that an extremely heavy isotope might be discriminated against so completely that its abundance would be attenuated considerably from that anticipated from cosmological considerations. Additionally, although we find it hard to conceive that the enrichment process could discriminate against anomalously heavy oxygen, it is noteworthy that residual oxygen from the air in the vacuum system of our source results in a current of at least 1 μA of $^{16}O^-$. If atmospheric oxygen had an anomalously heavy isotope we would have readily observed it at a level of $30\times 500 \times 10^{-16}$, that is 1.5×10^{-12}. It is our intention to repeat the experiment in the near future using ^{7}Li, a fraction of which is generally thought to be primordial in origin.

The present results place an upper limit on the existence of a stable heavy hadron with a mass in the range 4 to 38 amu (in oxygen) of 10^{-16}/nucleon and over much of the mass range at $< 10^{-18}$/nucleon. If such a particle had existed at the level of 10^{-10}, as suggested by Dover, Gaisser, and Steigman,[1] in the mass range studied, then we would have expected to observe 10^9 counts—this is to be compared with a maximum observed count of 100. In addition, the results place upper limits of 16 times those quoted above on the abundance of any anomalously heavy isotope of oxygen. It is noteworthy that the experiment is between nine and ten orders of magnitude more sensitive than previous measurements made by comparing physically and chemically measured masses.

We would like to express our thanks to Carl Dover, Tom Gaisser, and Ralph Amado for suggesting the experiment and to them, along with William Stephens, for many helpful discussions and suggestions. Our thanks are also due to Charles Adams for much help and assistance in operating the ion source and the tandem accelerator and also in accumulating much of the data. We also wish to acknowledge the support of the National Science Foundation.

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