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A SEARCH FOR ANOMALOUSLY HEAVY ISOTOPES OF LOW Z NUCLEI

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We present the results of a search for anomalously heavy isotopes of light elements using an electrostatic charged particle spectrometer in conjunction with the MP tandem accelerator facility at the Nuclear Structure Research Laboratory of the University of Rochester. New limits for the abundance of anomalously heavy isotopes (100–10000 amu) in ordinary terrestrial H, Li, Be, B, and F samples and enriched ^{2}H, ^{13}C, and ^{18}O samples are reported.

1. Introduction

It is a curious fact that, in spite of the large variety of elementary particles that have been observed and hypothesized during the past 40 years, it appears that all stable matter can be explained as various combinations of neutrons, protons, and electrons – particles well known to physicists since the 1930s. Big-Bang cosmology implies that all types of particles were present in large numbers during the earliest moments of creation. Thus, particles of virtually any mass that have lifetimes comparable to the age of the universe ($\approx 10^{10}$ years) should exist today as remnants of the Big-Bang. Various calculations have been performed [1–3] which yield estimates of 10^{-10}–10^{-12} for the concentration of anomalously heavy isotopes in nature. There are several potential candidates among the particles that are commonly considered in high energy physics possessing the required level of stability. In technicolor theories [4], for example, the lightest "techni-baryon", a technicolor singlet state of three technicolor quarks, is expected to have a lifetime of $\approx 10^{16}$ years. Supersymmetric theories [5] predict that all fermions have bosonic partners and vice versa; the lightest of these is expected to be stable. Particles of these types, if charged, should be observable in matter. Positively charged particles would have similar chemical properties to hydrogen and would appear in nature as an isotope of hydrogen with anomalous mass. Negatively charged particles would bind to ordinary nuclei, changing a nucleus of atomic number Z into one with atomic number $Z-1$ and anomalous mass.

We have constructed an all-electrostatic beam line for the University of Rochester Nuclear Structure Research Laboratory (NSRL) MP tandem electrostatic accelerator to search systematically for such components of matter. An electrostatic beam line transports ions independently of their masses, an essential feature since the masses of the ions for which we are searching are not known. We can enhance the selection of these ions by appropriately tuning the beam and configuring the detector.

Searches for massive isotopes of hydrogen have been reported [6–8], the most sensitive being that by Smith

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and co-workers [9]. Using electrolysis followed by analysis in a time-of-flight mass spectrometer, they were able to establish concentration limits of $< 10^{-28}$ per nucleon in ordinary water for isotopes in the mass range 8 – 1200 amu. Searches for anomalous isotopes of heavier nuclei have had less sensitivity. One of the most sensitive to date is that of Turkevich et al. [10], which places limits on the natural abundance of > 100 amu carbon-like nuclei at less than 1 per 10^{15} nucleons. Other less sensitive searches covering more limited mass regions have been reported for helium [11], lithium [11], beryllium [11], oxygen [12], sodium [13], and iron [14].

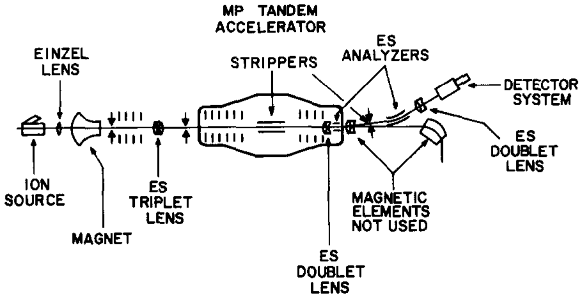

Fig. 1. A plan view of the apparatus including the injection system into the accelerator.

2. The electrostatic spectrometer

A diagram of the accelerator, spectrometer, and detection system is shown in fig. 1. It has been described in detail in ref. [15]. A cesium negative ion source, which can be used to sample virtually any type of solid material and many gases, feeds into the accelerator directly through electrostatic lenses. The sample beam is accelerated to 5 MeV in charge state 1^- to the terminal where it is stripped to charge state 1^+ in a 5 μg/cm^2 carbon foil. At the high energy end of the accelerator, the 10 MeV beam is focused with an electrostatic quadrupole doublet and deflected 1.3° off axis by a pair of electrostatic plates within the accelerator pressure vessel. Outside the accelerator, the beam passes through a removable 5 μg/cm^2 foil stripper and a set of adjustable slits at the focus of the quadrupole. This is followed by a high resolution 20° electrostatic analyzer, an electrostatic quadrupole doublet, and a small magnetic steerer which can be adjusted to sweep out low mass (< 100 amu) ions. A slit located at the focus of the second quadrupole system is followed by a microchannel plate time-of-flight transmission detector, a gas pressure cell to range out high Z ions, and a gas ionization counter to measure $\delta E/\delta x$, range and total energy of the ions.

We periodically verified the mass independence of the system by measuring the transmissions of the three elements produced by a specially prepared BeCuAu alloy. Small variations in the optimum beam tunes for these three elements were identified as being due to small differences in the energy losses in the two stripper foils and to small residual magnetic fields at the low energy end of the accelerator. These effects, while significant for masses of a few amu, have little effect for the high mass, low Z ions that are the subject of the search reported here.

For the hydrogen search we introduced a small magnetic field just downstream of the ion source. This eliminated light particles from the beam but had negligible effects on particles with masses $\gtrsim 100$ amu.

For other searches, we exploit the fact that heavy ions, in passing through a thin foil, do not have as many orbital electrons stripped as do light ions of the same energy. As a general rule, only those electrons that have orbital velocities less than the ion's velocity are stripped. This means, for example, that a 10 MeV 1000 amu "isotope" of beryllium will strip to charge state 1^+ half of the time, while normal ^{9}Be will usually strip completely to charge state 4^+, emerging in charge state 1^+ with a probability of 10^{-4}. By tuning for charge state 1^+ after both the terminal and high energy stripper foils, we achieve a rejection factor of $\approx 10^7$ for normal ions. Normal ions that do pass through the system are rejected in the detector, since a heavy version of any particular element will have a distinctly different $\delta E/\delta x$ and range.

3. Results

We measured a variety of samples, including normal lithium, beryllium, boron, and fluorine, and specially enhanced samples of oxygen, carbon, and hydrogen. We used a commercially available enriched oxygen sample which was enhanced in ^{18}O by a factor of 416. The carbon sample was prepared at the ^{13}C separation facility at Los Alamos and had an enhancement factor of $\approx 10^5$.

In our searches for heavy hydrogen, we used several samples: sea water from a depth of 3 km, commercially available deuterium (enhancement factor ≈ 20000), and samples of heavy water originally manufactured from lake water (enrichment 2×10^4) and further enriched by the UKAEA and Rutherford-Appleton Laboratory to give overall enhancement factors of 10^6–10^9 for heavy hydrogen-like isotopes. Each water sample was chemically processed to form H_2.

The concentration limit per nucleon, C, of heavy isotopes is calculated using the formula:

$$C = \frac{n_0}{n_s A} \frac{1}{\gamma_s} F.$$

Here n_0 is the number of heavy isotope events required for a 90% confidence level, n_s is the number of monatomic negative ions of the measured element produced by the ion source, A is the atomic mass number of the sample element, γ_s is the pre-enrichment factor, and F includes all mass dependent factors. n_s was measured using the relation:

$$n_s = If\Delta t,$$

where I is the average negative ion current from the ion source, f is the ratio of the monatomic ion current of the element of interest to the total current from the sample, and Δt is the total live time of the measurement.

The mass dependent term is given by the expression:

$$F = \frac{1}{y_1 y_2 \epsilon_t \epsilon_m} \frac{N^-(m_1)}{N^-(m_h)},$$

where y_1 and y_2 are the stripping yields for the heavy isotopes into charge state 1^+ at the first and second stripping foils, ϵ_t is the transmission of the heavy isotope, ϵ_m is the mass dependent correction for magnetic sweeping, and $N^-(m_1)/N^-(m_h)$ is the ratio of the negative ion emission rates for light and heavy isotopes in the ion source.

Transmission losses through the accelerator are due primarily to the stripping of negative ions in the residual gas at the low energy end of the accelerator. These losses have been measured by Lund [16] to range from about 10% for O^- up to 75% for Au^-. Theoretical calculations of charge changing cross sections indicate that they can be written as $\sigma = F(Z)g(v/v')$, where $v' = e^2/h = 0.007c$ is a velocity typical for the outermost electrons in an atom [17]. Data summarized by Allison and Garcia-Munoz [18] indicate that $g(v/v')$ has a broad maximum at $v/v' = 1$ and drops by about a factor of 10 at $v/v' = 10$, which is a characteristic velocity for light ions such as oxygen at the low energy end of the accelerator. At $v/v' = 0.1$, $g(v/v')$ falls below its peak value by about a factor of 3. This indicates that for any given Z, the variation in stripping cross section is less than a factor of 10. Thus for low Z ions, we expect a maximum variation in the transmission efficiency for heavy isotopes relative to normal ions to be a factor of three. We have taken the heavy isotope transmission to be 10%, which is more than a factor of 3 lower than that measured for normal ions, and have considered only the masses ≥ 100 amu where the effect of the magnetic sweeping is insignificant.

The ratio of the rates of negative ion emission can be written as the product of two ratios:

$$\frac{N^-(m_1)}{N^-(m_h)} = \frac{Y(m_1)}{Y(m_h)} \frac{S^-(v_1)}{S^-(v_h)}.$$

Here $Y(m)$ is the sputtering yield (atoms sputtered per primary ion) and $S^-(v)$ is the negative ion formation probability (fraction of those sputtered atoms which leave the ion source as negative ions). To calculate the sputtering yields we have used the empirical formula of Matsunami et al. [19] which indicates a loss of about a factor of 10 at mass 10000. Although several theories

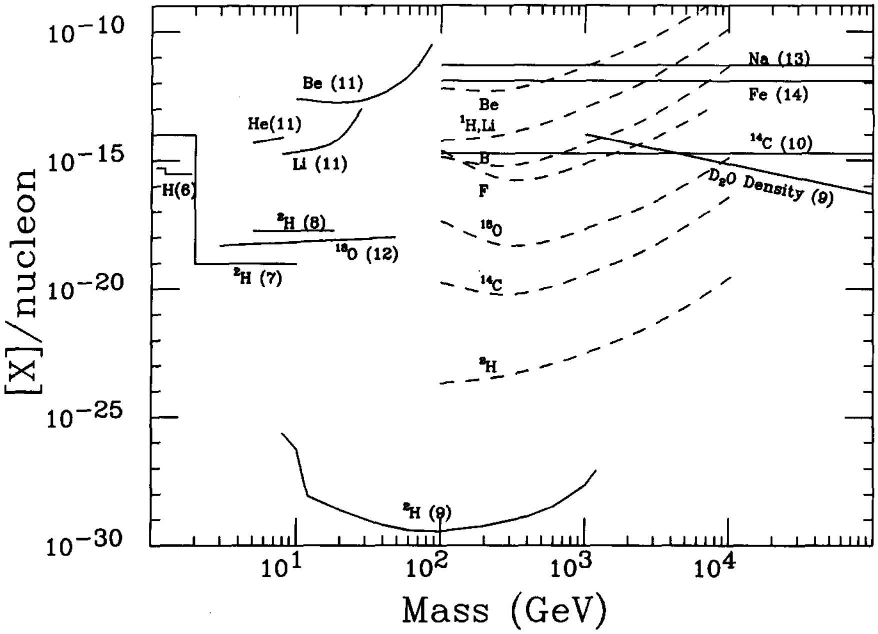

Fig. 2. Concentration limits (90% confidence level) for the existance of heavy isotopes in matter. Our results are shown as dashed lines and previously published results as solid lines. The dashed line labeled ^{1}H is for unenriched sea water from a depth of 3 km; that labeled ^{2}H is for deuterium enriched as described in the text.

for $S^-(v)$ exist [20–23], the theory of Nørskov and Lundqvist [24] is the most consistent with the experimental observations of Yu [25] and Vasile [26]. In this theory, the velocity-dependent ion emission probability is of the form:

$$S^-(v) \propto e^{-\{C_1(\phi - A) + C_2\}/v} \text{ (negative ions)}$$

$$S^+(v) \propto e^{-\{C_1(I - \phi) + C_2\}/v} \text{ (positive ions)}.$$

Here ϕ is the surface work function, A is the electron affinity, I is the ionization potential, and C_1 and C_2 are fitted parameters. Vasile has measured $S^+(v)$ for ions of Cr, Ag, Cu, and Zr with energies in the range of 1.4 to 60 eV, spanning more than 6 orders of magnitude in ion formation probability. Plots of $\log_{10} S^+(v)$ against v^{-1} show linearity in the energy region $4 \leqq E \leqq 30$ eV, with slopes of $-(2-3) \times 10^6$ cm/s, and slopes of $-(3-6) \times 10^6$ cm/s for $E > 30$ eV. No correlation with $(I - \phi)$ is observed. Similarly, Yu determines the slope for negative oxygen ions to be 5×10^6 cm/s. We have used this value in our calculations.

We required $\geqq 1$ event ($n_0 = 2.3$ for 90% confidence level) to define a positive signal in the heavy hydrogen search because the spectra were essentially background free. For the other samples, we required $\geqq 3$ events ($n_0 = 5.3$ for 90% confidence level) within a region taken to be $\geqq 3$ times wider than our fwhm resolution in the total energy measurement and 4 $\delta E/\delta x$ measurements. In fig. 2, we show our preliminary results as dashed lines, and previously published results as solid lines. The curves for previous searches that make use of an ion source have not been corrected for the mass dependence of negative ion formation; in particular the hydrogen curve for ref. [9] should be somewhat higher than that shown.

4. Conclusions

Geological fractionation processes are not thought to dramatically change the concentration of heavy isotopes in normal matter. In light of the large discrepancy between the measurements reported here and the predictions of refs. [1–3], it would appear safe to rule out the existence of stable charged particles in the mass range 100–10 000 amu.

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