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Searching for supermassive Cahn-Glashow particles

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A radiochemical technique, based on the $^{14}N(n,p)^{14}C$ reaction, has failed to detect the presence of supermassive negatively charged particles (X^-) in terrestrial carbon. The limits of X^- per nucleon of $\sim 2\times 10^{-15}$ are applicable to masses less than $\sim 10^5$ amu. These limits are orders of magnitude lower than previously reported for X^- particles of this mass range bound to light nuclei in terrestrial samples. Examination of the geochemical and nucleosynthetic history of such particles suggests that, if present, they should be depleted in light elements and concentrated in elements of the iron region, possibly in meteorites.

I. INTRODUCTION

Cahn and Glashow[1] have pointed out the possibility that superheavy isotopes ($10-10^5$ amu) might be present in ordinary matter in amounts small enough ($<10^{-9}$ per nucleon) to have escaped detection. One source of such isotopes might be the result of the attachment of a very heavy negative particle X^- to an ordinary nucleus. If such a particle had *only* electromagnetic interactions with ordinary matter (no hadronic or weak interactions) and itself had long enough lifetime for decay, there would be present today superheavy isotopes of an element Z of ordinary matter arising from the attachment of the X^- to a nucleus of an element $Z+1$. In view of the present uncertainties in theoretical predictions for particles with masses greater than 10^2 amu ($\sim 10^2$ GeV), experimental searches would appear to be warranted to supplement the extensive theoretical investigations currently under way.

The abundance of supermassive isotopes is clearly very low. Presumably most of any X^- present at an early high-temperature phase of the Universe would have annihilated during the dense cool-down period. There still would be a residue of particles that never found an antiparticle. In addition, there are possibilities of asymmetries of the type that resulted in the baryon excess in the Universe (the baryon-to-photon-number ratio is $\sim 10^{-9}$). Estimates of the primordial (big bang) residues of such particles have been in the 10^{-12} concentration range for masses between 10 and 10^4 amu.[2,3]

In addition to changing the nuclear charge by one unit, the addition of such an X^- to a nucleus of charge Z, by changing the Coulomb energy of the nuclear system, will change the β-stability relationships with its ordinary isobars. For example, $_5(^{14}_{6}CX^-)$ can be expected to have the chemistry of boron and to have a larger β^- decay energy and shorter half-life than ^{14}C.

Although the basis for predicting the presence of such particles is very speculative, their existence would have such importance, both theoretical and even possibly practical, that very sensitive searches for them are warranted.

II. PREVIOUS SEARCHES

Most previous attempts to find heavy isotopes in nature have concentrated on the element hydrogen. Alvager and Naumann[4] set a limit of 3×10^{-18} for the presence of isotopes with masses between 6 and 16 amu. Muller *et al.*[5] used a cyclotron to set a limit of 2×10^{-19} for isotopes with masses below 8.2 amu. However, the most sensitive search reported to date for heavy-mass particles in nature is that of Smith *et al.*,[6] who searched for anomalously heavy hydrogen (M) in water. Their limits of 10^{-28} to 10^{-29} for M/H in terrestrial water are applicable to a mass range of up to 350 times the proton mass, with slightly poorer sensitivity up to 10^3 amu. In terms of the framework of this report, these searches in hydrogen were sensitive to $M=X^+$ or $M=He^{++}X^-$. These limits are many orders of magnitude lower than the predicted values for this mass range (if there were any stable particles) having been left over from the early hot stages of the Universe.[2,3]

Middleton and coworkers[7] have reported a search for isotopes of oxygen with masses up to 54 amu. Their limits are $10^{-16}-10^{-18}$ of such heavy oxygens ($_7NX^-$) relative to ordinary oxygen.

Dick, Greenlees, and Kaufman[8] have searched for anomalous mass (>100 amu) sodium atoms, using expected changes in the hyperfine splitting of the optical spectrum of sodium. Their limits are $<5\times 10^{-12}$ per nucleon of such species. In terms of the concepts of the present work, these limits would apply to an X^- bound to magnesium nuclei.

A CERN internal report[9] indicates that a search is being started for anomalous fission events in very heavy elements as an indication of the presence of superheavy isotopes. A sensitivity of 10^{-11} for the concentration of such isotopes is expected.

Another search, also with negative results, has been reported by Brandt *et al.*,[10] who looked for $_{95}(^{247}CmX^-)$ in ocean hot brines. They obtained a limit of $<10^{-12}$ atoms per gram of dried precipitate. If a stable americium

($Z=95$) nuclide existed, its abundance in the dried precipitate might have been as high as 100 ppm, since the brines are enriched in elements forming chloride complexes [the lead concentration is 800 ppm (Ref. 11)]. In that case the Brandt limit corresponds to an abundance (at element formation time) of 10^{-6} for the supermassive isotopes or an initial abundance of 10^{-8} for X^- per nucleon in americium.

III. PRINCIPLE OF SEARCH

The principle of this search for supermassive isotopes was evolved in discussions with Sugarman.[12] It is based on the expectation that the $_6(^{14}NX^-)$ system, if present in a sample of carbon, would undergo (n,p) reactions to form $_5(^{14}CX^-)$ in a manner relatable to the way ordinary ^{14}N reacts with neutrons to form ^{14}C.

The $_5(^{14}CX^-)$ species would have the chemistry of boron, but be β^- radioactive with a decay energy of ~ 0.8 MeV, increased from that of ordinary ^{14}C by the Coulomb effects discussed by Cahn and Glashow.[1] The expected half-life is calculated to be ~ 15 yr, assuming that the f_0t value is unchanged[13] from that of ^{14}C. Since normal boron does not have any long-lived radioactive isotopes, the presence of a long-lived β^- radioactivity following the chemistry of boron in neutron irradiated carbon would be evidence for the presence of $_6(^{14}NX^-)$ in the original carbon. The energy relationships of the $^{14}_{7}N+n$ systems and those postulated for the $_6(^{14}NX^-)+n$ systems are illustrated in Fig. 1.

It is seen that the (n,p) reaction in this case is almost thermoneutral. Thus, the reactions that would form

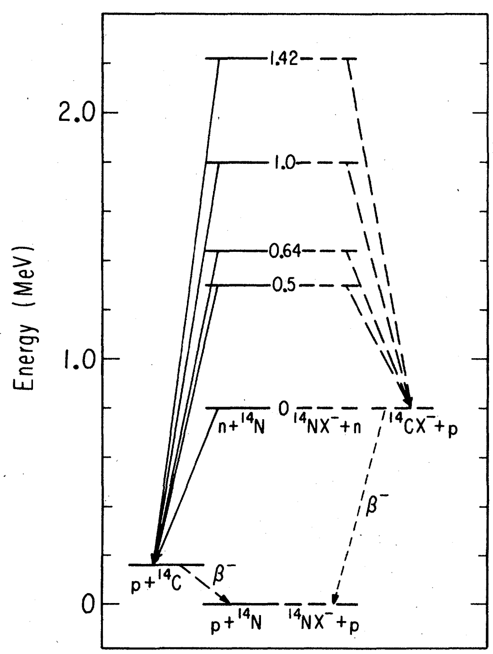

FIG. 1. Energy relations in the $A=14$ plus 1 nucleon system (left side) and those in the same system with a massive X^- attached to the ^{14}N or ^{14}C (right side). It is assumed that only Coulomb forces are involved. The ground states of the two systems are shown at the same energy level.

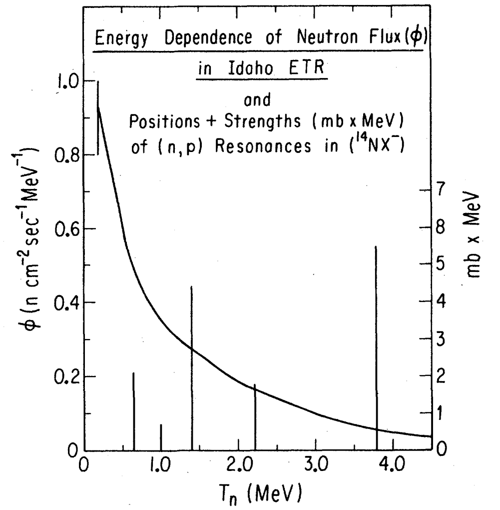

FIG. 2. Energy dependence of the neutron flux (ϕ) in the Idaho Engineering Test Reactor (left scale) and positions and strengths (mb MeV) of (n,p) resonances in the $^{14}NX^-$ system (right scale).

$_5(^{14}CX^-)$ require energetic neutrons which interact primarily in the resonance region. The calculated proton-emission widths of the individual resonances were modified[14] from those of ordinary ^{14}N by assuming that the main factors were the lower probabilities for Coulomb-barrier tunneling. The positions of the important resonances and the estimated cross-section integrals are indicated in Fig. 2.

The special advantage of this approach (as well as that of Brandt *et al.*) relative to mass spectrometric searches is that the former has a much higher upper mass limit. Thus, although the sensitivities cannot approach those achieved by Smith *et al.*,[6] their results are applicable to masses less than 10^3 amu, whereas the present search should be sensitive to masses as high as 10^5 amu.

IV. EXPERIMENTAL DETAILS

The samples of carbon used were reactor-grade graphite obtained from Battelle Northwest Laboratories. They had been irradiated in the Engineering Test Reactor[15] at Idaho Falls, Idaho. The neutron spectrum in the reactor, calculated from the data of Morgan,[16] is shown in Fig. 2. Using this flux distribution and the modified resonance integrals (those at 0.64 and 1.4 MeV contribute most) leads to an effective cross section of 3.2 mb for the (n,p) reaction with ($^{14}NX^-$) in this neutron flux.

Two samples of graphite were processed. Their characteristics and those of the irradiations are shown in Table I. A third sample was accidentally lost in processing.

The graphite samples were crushed and a known amount of boric acid (~ 1.5 mg boron) was added together with 0.3 g of $Ca(OH)_2$ suspended in water. The carbon was burned off in oxygen and the residue put through chemical operations designed to remove any natural radioactivity or activity induced in the impurities in the graphite by the high neutron flux. This procedure started by

TABLE I. Results of search for supermassive carbon.

Sample	Mass (g)	Fluence[a]	Recovery[b]	Activity limit[c]	Concentration limit Isotopic	Nucleonic[d]
I	2.2	2.0×10^{22}	0.23	<0.01	$<2.6\times10^{-13}$	$<1.8\times10^{-14}$
II	30	1.7×10^{22}	0.15	<0.01	$<3.1\times10^{-14}$	$<2.2\times10^{-15}$

[a]The fluence is in integrated neutrons per cm^2, with energies greater than 0.18 MeV.
[b]The recovery is based on the fraction of added inert boron that was recovered.
[c]The activity limit is in events min^{-1} above background in a measurement of $\sim25\%$ efficiency for the postulated β^- particles of 0.8 MeV maximum energy.
[d]This is the limit of X^- to nucleons (neutrons and protons).

fusing the residue, after CO_2 volatilization, with Na_2O_2 and Na_2CO_3. The melt was then dissolved in perchloric acid. Several scavenging precipitations were performed with hydroxides and carbonates. These were followed by a distillation of methyl borate from perchloric acid. The basic glycerol solution containing the distillate was dried and ignited with Na_2O_2 and, after a $Fe(OH)_3$ scavenge, the boron was precipitated as barium borotartrate. This sample usually showed traces of radioactivity, so it was dissolved, ignited to destroy organic matter, scavenged again with $Fe(OH)_3$, and finally precipitated as barium borotartrate. This was then filtered and the final sample was measured on a low-background β^- counter. An aluminum absorber of ~7 mg cm^{-2} was used to remove any natural ^{14}C activity. The efficiency of this counter for the predicted energy β rays was estimated to be about 25%.

V. RESULTS AND DISCUSSION

The results of the measurements of the β radioactivity of the purified boron samples are summarized in Table I. There is no evidence for the production of a new long-lived β^- radioactivity following the chemistry of boron in the samples of highly neutron-irradiated graphite. For example, measurements on sample II gave a mean of 0.114 counts/min ($N=20$, $S=0.0066$). Interspersed measurements on a background sample gave 0.119 counts/min ($N=6$, $S=0.011$). From this we conclude that the sample had less than 0.01 counts/min above background (95% confidence). Assuming that the hypothesis proposed is valid for the neutron conversion of ($^{14}NX^-$) to ($^{14}CX^-$), limits can be calculated on the concentration of ($^{14}NX^-$) in the original graphite. These are based on a 15-yr half-life for ($^{14}CX^-$), a 400—500-day irradiation over a period of 4—5 yr, and a 12-yr decay period before analysis. They are shown in the next to last column of Table I. These limits, in terms of X^- per nucleon, are 14 times lower, or $\sim3\times10^{-15}$ in the case of the 30-g graphite sample.

These limits are not as low as those of Smith *et al.*[6] for hydrogen or even those of Middleton *et al.*[7] for oxygen. On the other hand, they are lower than the limits of Brandt *et al.*[10] and of Dick, Greenlees, and Kaufman. In addition, like these last two searches, the applicable mass ranges are up to about 10^5 amu (estimated from the possible mass effects during the chemical operations—centrifugation and distillation), and thus much larger than the more sensitive searches.

Questions can be raised whether terrestrial water (the ultimate source of the samples in the hydrogen and oxygen searches) may have a significantly lower concentration of the ultraheavy isotopes than primordial hydrogen because the water was probably originally derived from dehydration and diffusion out of hydrated minerals (the earth probably has lost any original gaseous water). The present work suffers from similar disadvantages in being subject to possible biases due to the geochemical history of the samples. The graphite probably derives from living material that had incorporated atmospheric carbon dioxide into its structure. The possibility of significant mass fractionation of such supermassive isotopes of carbon, in the diffusion out of rocks of the CO_2 and subsequent life processing, is hard to exclude.

Thus, the present work suggests that further searches for such particles concentrate on samples that are least likely to have been depleted in supermassive isotopes. Iron meteorites are good candidates from this standpoint.

VI. NUCLEOSYNTHETIC HISTORY

In addition to the questions about the geochemical history of the samples used in the present and other searches

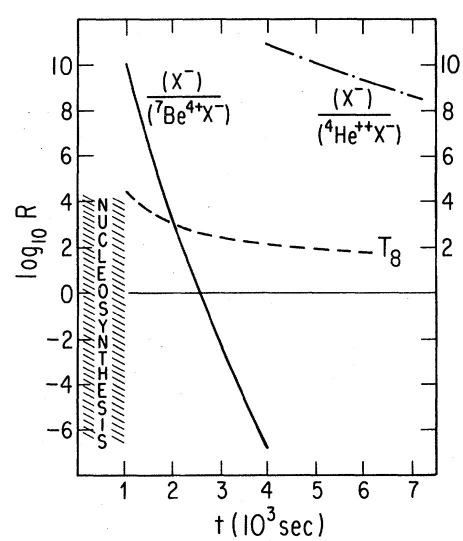

FIG. 3. Logarithm of the equilibrium ratios (R) of the amounts of X^- particles that are free relative to the amounts attached to $^7Be^{4+}$ and $^4He^{++}$ as a function of time in the early universe (left scale). The temperature T_8 in units of 10^8 K, as a function of time is shown on the right scale.

for supermassive isotopes, there are questions concerning the nucleosynthetic history of the particles responsible for their existence. In the case of Cahn-Glashow-type particles (i.e., long-lived, massive, negatively charged particles having only electromagnetic interactions with ordinary matter), these questions can be discussed concretely in terms of presently accepted scenarios of nucleosynthesis.

Cahn and Glashow[1] have calculated the binding energies of such particles to ordinary light nuclei. These binding energies are low enough (25 keV to $^1H^+$, 311 keV to $^4He^{++}$, and 1.49 MeV to $^7Be^{4+}$) that the X^- particles will not affect the course of standard model nucleosynthesis during the first few minutes of the Universe[17] because the temperatures are too high. However, as the temperature drops, they will become bound to nuclei before electrons do. Figure 3 shows the equilibrium ratios of $(X^-)/(^7Be^{4+}X^-)$ and $(X^-)/(^4He^{2+}X^-)$ in the period 1000 to 7000 sec. These calculations suggest that all the X^- will end up bound to $^7Be^{4+}$ even though the abundance of this species is only $\sim 3\times 10^{-10}$. There should be less than 10^{-16} of the nucleons left in the form of superheavy hydrogen $(^4_2He^{++}X^-)$ by the time electronic recombination screens the nuclei from exchanging their X^- particles. This might help explain the lack of heavy hydrogen found by Smith *et al.*[6]

The subsequent history of the X^- particles, after formation by electron capture of $(^7Li^{3+}X^-)$, will depend on the prehistory of the earliest stars. As matter agglomerates and temperatures rise in the prestellar stage, the following reactions are expected to occur:

$$^1H^+ + (^7Li^{3+}X^-) \to {^4He^{++}X^-} + {^4He^{++}} \quad (16.76)\,, \qquad (1)$$

$$(^4He^{++}X^-) + {^4He^{++}} \to (^8Be^{4+}X^-) + \gamma \quad (1.15)\,, \qquad (2)$$

$$(^8Be^{4+}X^-) + {^1H^+} \to (^9B^{5+}X^-) + \gamma \quad (0.40)\,, \qquad (3)$$

$$(^9B^{5+}X^-) + e^- \to (^9Be^{4+}X^-) + \nu \quad (0.68)\,, \qquad (4)$$

$$(^9Be^{4+}X^-) + {^1H^+} \to (^{10}B^{5+}X^-) + \gamma \quad (7.19)\,, \qquad (5)$$

$$(^{10}B^{5+}X^-) + {^1H^+} \to (^{11}C^{6+}X^-) + \gamma \quad (9.38)\,, \qquad (6)$$

$$(^{11}C^{6+}X^-) \to (^{11}B^{5+}X^-) + \beta^+ + \nu \quad (1.35)\,, \qquad (7)$$

$$(^{11}B^{5+}X^-) + {^1H^+} \to \begin{cases} (^8Be^{4+}X^-) + {^4He^{++}} \quad (7.90), & (8a) \\ (^4He^{++}X^-) + 2\,{^4He^{++}} \quad (6.75), & (8b) \\ (^{12}C^{6+}X^-) + \gamma \quad (16.63). & (8c) \end{cases}$$

(The numbers in parentheses on the right are the predicted Q values for the reactions in MeV.) Note that the presence of X^- makes $^8BeX^-$ and $^9BX^-$ particle-stable.

Reactions (2) through (8a) and (8b) represent an X^- catalyzed conversion of protons into ^4He. If the X^- should be liberated, for example, in reactions (1) or (8b), it would probably get reattached quickly to an abundant $^4He^{++}$ nucleus. However, reaction (8c) will drain off X^- particles into $^{12}CX^-$, stopping the cycle after some 10^3 to 10^4 conversions. These reactions should proceed at a lower temperature and density than the polymerization of ^4He into ^{12}C.

Similarly, the X^- can enable the standard CNO cycle to proceed at a lower temperature than that at which it usually occurs, but again the catalysis will be stopped after 10^3–10^4 conversions, and the X^- will end up as $(^{16}OX^-)$. This can further capture protons and build up heavier elements. Because of the lower Coulomb barriers, this incorporation into elements in the region of iron is likely to go to a larger extent than with nuclei not having an X^-.

Thus, although Cahn-Glashow-type particles can catalyze prestellar and stellar nucleosynthesis, the extent of this, for concentrations of X^- per baryon of less than 10^{-12}, is expected to be negligible before the X^- particles are incorporated into elements heavier than even oxygen. Whereas Coulomb effects should enhance nuclear processing up until the iron region, such effects should be less important in the neutron processing of material into heavier elements.

VII. SUMMARY

A radiochemical technique has failed to detect the presence of supermassive negatively charged particles (X^-) in terrestrial carbon. The limit of X^- per nucleon of 3×10^{-15} is applicable to masses less than $\sim 10^5$ amu. This limit is lower than those reported in previous work for this mass range.

Examination of the geochemical and nucleosynthetic history of such particles suggests that they may be depleted in samples of the lightest elements and further searches should be directed at samples of heavier elements such as those near iron.

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