In recent years, the cheapest way to accomplish vertical MPS for MX has been by a scheme referred to as *Carryhard*. In this scheme, the MX missile is placed in a hardened cannister containing the required operational support systems, and the missile in its cannister is moved between relatively austere and cheap vertical shelters.

In order to compare Carryhead MX basing with Midgetman missiles on hard mobile launchers based in the Southwest, it is necessary to calculate that number of independent aim points required to extract an attack price of attacking Soviet warheads, which is equal to the attack size the Soviets would need to barrage the area occupied by the mobile Midgetman force. The cost of 50 MX missiles based in Carryhard with the required number of shelters is $21 billion. This is an analytically respectable alternative to consider because it provides adequate survivability at an appreciable cost savings.

Most knowledgable observers place significant emphasis on the greater flexibility provided by a small missile. There is also considerable sympathy for adopting as the nation's new ICBM a small missile, which, over time, could and should be adopted by both the United States and the Soviet Union as the central element of a land-based ICBM force. How the Bush Administration and Congress answer the ICBM modernization question will be a significant indicator of our government to resolve difficult national security issues in the foreseeable future.

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8. Other cost differences should also be considered. For example, the Air Force is correctly concerned with the larger need for uniformed personnel to support the mobile Midgetman.
9. *Soviet Military Power* (Department of Defense, Washington, DC, 1988).
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11. A viewpoint is presented in (*2*) and more recently by Brent Scowcroft and R. James Woolsey, in *American Agenda: Report to the Fifty-First President of the United States of America* (Washington, DC, 1988).
12. Here and elsewhere in the paper, cost estimates are obtained from sources the author believes to be reliable. The estimates depend on many assumptions and should be considered to give an impression of relative, not absolute, costs. System cost estimates are given in fiscal year 1988 dollars, undiscounted, and include remaining research and development, procurement, and 15-year operating expenses.
13. The difference in cost arises largely because of the higher operations and maintenance expenses of mobile basing compared with silo basing.

Optimum Chemical Sites and Techniques for Searches for Negatively Charged Rare Particles

R. N. BOYD, K. TAKAHASHI, ROBERT J. PERRY, TERRY A. MILLER

Supersymmetric particle theories have predicted the existence of massive, negatively charged, nonstrongly interacting particles, denoted as X^- particles. If stable X^- particles existed at the onset of primordial nucleosynthesis, they would have been bound initially to the primordial nuclides. However, subsequent stellar processing, as part of the chemical evolution of the galaxy, is shown to have produced considerable rearrangement of the relative chemical abundances of X^- particles subsequent to their binding to primordial nuclei. Optimal chemical environments in which to search for X^- particles are found to be boron and fluorine. A mass-independent search for X^- particles bound to heavy nuclei that utilizes laser spectroscopy of rotational bands in diatomic molecules is estimated to have an unusually high relative sensitivity to possible X^- particles.

ONE OF THE FASCINATING PREDICTIONS RESULTING FROM attempts of particle theorists to unify the forces of nature is the possible existence of supersymmetric (SS) particles (*1*). Although SS theories have generated considerable attention, their ultimate reality depends on the existence and observation of at least some of the predicted particles. Attempts (*2, 3*) to produce these particles in high-energy interactions have thus far been unsuccessful, but they have set an apparent lower limit on the masses of such particles of about 25 GeV (masses are given as mc^2, where c is the speed of light, with the proton mass being 0.938 GeV in these units).

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Another possibility is that such particles might be either stable or sufficiently long-lived that they could be found, albeit at low abundance levels, in nature. The experiments that have been performed to search for them have not yet found any indication that they exist but have imposed stringent abundance limits for them in some chemical environments. Positively charged SS particles would behave chemically like protons; a mass spectroscopic search (*4*) using hydrogen suggests that such particles having masses between about 6 and 350 GeV could not exist in nature above an abundance of 1×10^{-21} particle per proton. Another experiment (*5*) has set even tighter limits, as low as 1×10^{-29} particle per proton, for some masses, and better than 1×10^{-27} from 12 to 1200 GeV. Although considerations of source material (*6*) and mass-dependent effects of source emission (*7*) weaken these limits considerably, those experiments imply a very low abundance of stable, positively charged SS particles.

The possibility that negatively charged SS particles, or any rare, heavy, negatively charged particles, denoted as X^- particles, could exist as stable entities in nature was discussed by Cahn and Glashow (*8*), and searches for them have been performed by Turkevich, Wielgoz, and Economou (*6*) and by Norman, Gazes, and Bennett (*9*). All three of these studies, together with searches (*4, 5*) performed on H, have examined chemical environments in which special circumstances could make detection of nuclear species with a bound X^- feasible, even at very low abundance levels. For example, the searches in H would be sensitive to a heavy X^- bound to He, because such a system, plus an electron, would chemically resemble H. However, as shown below, interpretation of possible X^- searches requires that additional refinements, specifically effects of stellar processing and galactic chemical evolution subsequent to Big Bang nucleosynthesis, be considered.

We also describe a class of experiments that could be used to search for X^- particles having masses roughly that of the proton and greater that are bound to a variety of nuclides. These experiments would use laser spectroscopy of rotational spectra of molecules, first to seek the X^- particles that could exist in electron-like atomic orbits, and then to obtain an accurate determination of the mass of the X^-.

Interaction and Binding Between X^- Particles and Nuclei

The interaction between the X^- and the nucleus is crucial to our considerations. In this article, we assume that the interaction is electromagnetic; strong interactions would result in profoundly different conclusions. The binding energy of an X^- to a nucleus through an electromagnetic interaction may be calculated with a Hamiltonian (*8*)

$$H = -\frac{\hbar^2}{2\mu}\nabla^2 + V(r) \qquad (1)$$

with

$$V(r) = \begin{cases} -3Ze^2/2R + Ze^2r^2/2R^3 & \text{for } r < R \\ -Ze^2/r & \text{for } r > R \end{cases} \qquad (2)$$

where V is the potential energy, r is the distance between the X^- and the nucleus, μ is the reduced mass, $\hbar$ is Planck's constant divided by 2π, Ze is the nuclear charge, and R is the nuclear radius taken to be $1.2\ A^{1/3}$ femtometers with A being the mass number of the nucleus. The X^- binding energies (in megaelectron volts) for an X^- of mass m_x in nucleus ${}^AZ$, $B_x({}^AZ^x)$, are shown in Fig. 1.

The equation describing the rate of change of the number abundance of the X^--bearing nucleus $n({}^AZ^x)$ in a high-temperature (T) environment is

$$\frac{dn({}^AZ^x)}{dt} = -\lambda_I\, n({}^AZ^x) + \lambda_C\, n(X^-) + (\text{nuclear reactions}) \qquad (3)$$

where λ_I and λ_C are the rates of photoionization (${}^AZ^x \rightarrow {}^AZ + X^-$) and capture (${}^AZ + X^- \rightarrow {}^AZ^x$), respectively, and $n(X^-)$ is the number abundance of free X^-. The last term in Eq. 3 represents the effects of nuclear reactions. The rates λ_I and λ_C could be computed with the Hamiltonian (Eqs. 1 and 2) (folding with the Planck and Boltzmann distributions, respectively). For simplicity, however, we set up an "equivalent" Coulomb potential to mimic Eq. 1, so that well-known analytic formulae (*10*) for the cross sections can be used. (This approximation is better for light than for heavy nuclei but should suffice for the purposes of the present study.) Figure 2 displays the ionization and capture rates calculated for some species versus temperature. Combination of those rates with primordial abundances (*11–13*) of nuclei shows that all of the X^- capture reactions occur in times (*11–13*) much greater than that for primordial nucleosynthesis.

Because we were primarily interested in the gross features of the distribution of X^- throughout the periodic table, we did not consider detailed nuclear reaction rates for nuclei with a bound X^-. For the most part, such details will not affect our conclusions. One exception, that involving ${}^8Be^x$, is worth noting, however. The added X^- would increase the binding energy enough so that ${}^8Be^x$, unlike ${}^8Be$, would be bound. Furthermore, ${}^8Be^x$ would be formed (*6*) from the reaction ${}^4He^x + {}^4He \rightarrow {}^8Be^x + \gamma$ (γ denotes a gamma ray), which, because ${}^4He^x$ and ${}^4He$ would have a small Coulomb barrier, would be expected to proceed readily under stellar H burning conditions. We have calculated the rate for that reaction, using the (standard astrophysical) expression (*14*) for the total cross section for radiative capture σ at center-of-mass energy E

$$\sigma(E) = (S/E)\exp(-2\pi Z_1 Z_2 e^2/\hbar v) \qquad (4)$$

where Z_1e and Z_2e are the charges of the two interacting nuclei, and v is their relative velocity. S, the "astrophysical S-factor," is a number that usually varies slowly with energy; we have assumed a typical nonresonant (*14*) value for S of 1.0 keV-barns. The result of this estimate is indicated in Fig. 2. Turkevich *et al.* (*6*) also listed reactions that would convert ${}^8Be^x$ to heavier nuclei. Because these comprise a fairly complicated nuclear reaction network and estimates of the appropriate reaction rates would necessarily be quite

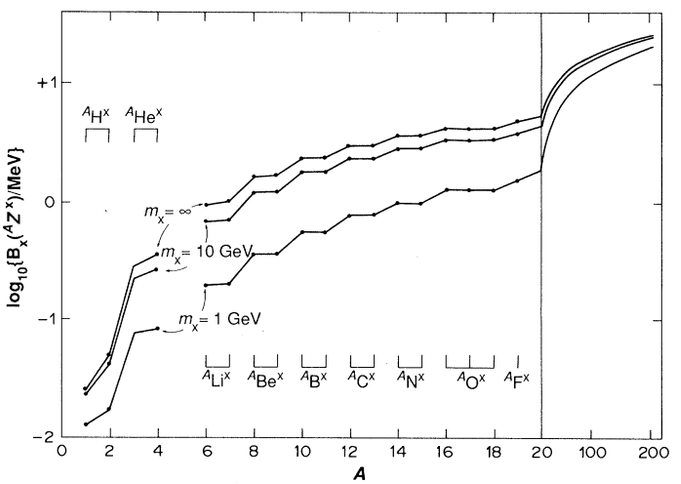

Fig. 1. Binding energy of an X^- to a nucleus as a function of the mass of the nuclide. Three masses of X^- were assumed: 1 GeV, 10 GeV, and an infinite mass.

uncertain, we did not believe that a detailed study of these effects was justified. Qualitatively, we would expect that these reactions would proceed fairly rapidly to promote most of the lower mass, X^--bearing nuclides to at least C^x.

Big Bang Nucleosynthesis and X^- Sites

The environment in which Big Bang nucleosynthesis occurred could have determined the nuclei to which the X^- particles, if they are stable, are now bound, at least in matter that has not been processed in stars. Big Bang nucleosynthesis is thought to have begun at a sufficiently high temperature (10^9 K) that appreciable combination of X^- particles with protons to form $^1H^x$ during that period would not have occurred. This reflects the relatively small binding energy of an X^- to ^1H, that is, approaching 25 keV as the X^- mass is taken to infinity. Even after most of the ^4He was formed, the temperature would still have been high enough that the probability of forming even $^4He^x$ would have been small.

Even if some of the X^- particles did bind to ^4He, the increase in binding energy appears insufficient to allow a stable or even long-lived $^5Li^x$ or $^5He^x$ unless the X^- particles interacted through the strong interaction. Thus the usual reactions $^4He + {}^3H \rightarrow {}^7Li + \gamma$ and $^3He + {}^4He \rightarrow {}^7Be + \gamma$ would still be required to bridge this "mass 5 bottleneck" of nucleosynthesis. However, if the X^- particles interacted through the strong interaction, $^5He^x$ and $^5Li^x$ would be stable to baryon emission. Then the strongly interacting X^- particles could have an enormous impact (*15*) on some astrophysical processes, including stellar burning, even at extremely low abundance levels. Furthermore, an unstable, strongly interacting X^- could have drastic effects on Big Bang nucleosynthesis without leaving directly observable signatures.

Because the binding energy of an X^- to a nucleus increases with nuclear charge [hence with mass (see Fig. 1)] and because temperature decreased with time after the Big Bang, the X^- particles that were captured by the heaviest nuclei would tend to remain with those nuclei while those captured by lighter nuclei would be more readily reionized. As the universe expanded with the passage of time, the rapidly decreasing density would have made it progressively harder, despite the decreasing temperature, for nuclei to combine with X^- particles. This would be more so for heavier nuclei than for lighter nuclei, as λ_C is (see Fig. 2) smaller for massive nuclei because of their lower thermal (kinetic) energy. These factors would have resulted in a distribution of X^- particles throughout the primordial nuclei. Although the ratio of abundances of X^--bearing nuclei to their normal counterparts, $[A^x]/[A]$, would be somewhat higher for the heaviest nuclei produced, it would probably not be

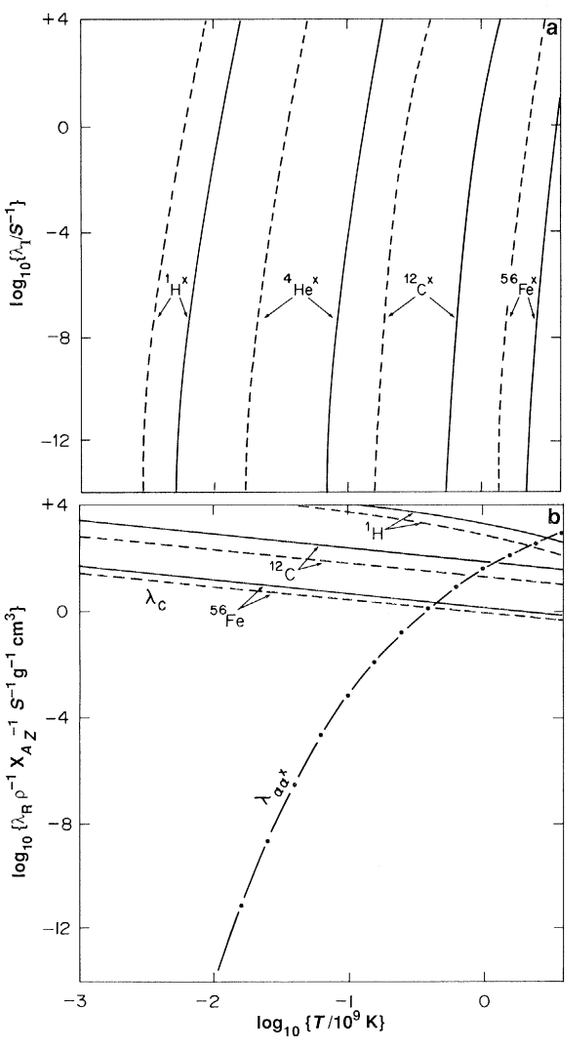

Fig. 2. (**a**) Photoionization rates for various X^- nuclei as a function of the temperature of the environment. The solid curves assume an infinite X^- mass, the dashed curves assume an X^- mass of 1 GeV. (**b**) Rates for capture of an X^- on a variety of nuclei. The solid curves assume an infinite X^- mass; the dashed curves assume an X^- mass of 1 GeV. The dashed-dotted curve is the estimated rate for the reaction $^4He^x + {}^4He \rightarrow {}^8Be^x + \gamma$. The density is denoted by ρ.

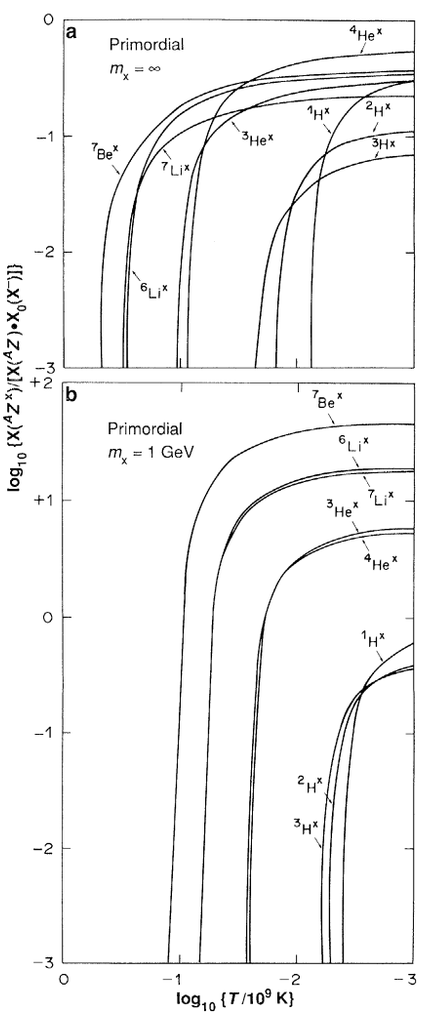

Fig. 3. Ratios of abundances of X^- nuclei to those of the normal nuclei divided by the abundance of X^-, as a function of the temperature of the environment (**a**) for an infinite X^- mass and (**b**) for an X^- mass of 1 GeV. The values for an X^- mass of 10 GeV are close to those for the infinite mass.

greatly reduced for the lightest nuclei. These abundances (Fig. 3) were obtained with the above rates combined with the standard model (*11, 13*) of primordial nucleosynthesis.

Even though these considerations would suggest that H^x should be abundant immediately after primordial nucleosynthesis, stellar processing of the primordial material subsequent to the Big Bang would heat much of the primordial H^x to a sufficiently high temperature to reionize it, thereby reducing its abundance. If H^x did exist, it would behave chemically like a neutron, complicating selection of a chemical environment favorable to a search for H^x. Thus it was deleted from our considerations of likely candidates in the present universe for stable X^- particles.

Sites of X^- Particles at the Time of Formation of the Solar System

Although ionization and capture processes for X^- particles occurred much less rapidly in the early universe than in the seconds over which appreciable temperature changes occurred, the opposite situation exists in stars. The millions to billions of years over which most stellar burning stages operate, depending primarily on the stage, the total mass of the star, and the fraction of its mass that is in nuclei heavier than ^{4}He, are responsible for this reversal. Figure 2 shows some relevant reaction rates for stellar burning conditions. Table 1 lists various stages of stellar burning, along with some of their characteristic properties (*14*). At a temperature typical of any of them, tens of millions of kelvins for H burning to several billion kelvins for the final stages of stellar collapse, any X^- in an environment containing only H would be ionized and recaptured many times. If heavier nuclei also were present, however, some probability for X^- capture by them would exist. Subsequent reionization would not occur if the capturing nucleus were heavy enough, because of the large resulting binding energy. If the heavier nuclei were subsequently involved in a higher temperature phase, however, the X^- might be ionized and a net removal of the X^- from those nuclei would result. Thus it is important, in determining the present sites of X^- particles that have resulted from stellar processing, to know what fraction of any star completes each stellar burning phase and what conditions, for example, temperature and nuclidic abundances, prevail in each phase.

Because we are concerned with present terrestrial sites of X^- particles and because Earth was formed from the interstellar medium (ISM), we must understand how material is processed in stars and what part of it is expelled at the end of each star's life. After a star has converted most of the nuclear fuel in its interior to more massive nuclei, it will, as it assumes its ultimate stellar state, return some of the nuclei it synthesized to the ISM. However, stars of different masses end their lives in different ways; some may return little of their material to the ISM (low-mass stars, which collapse to white dwarfs), whereas others may undergo an explosion of their entire mass into the ISM (several solar mass stars, which become "C deflagration supernovae"). More massive stars have shorter lives than less massive ones. All of these factors, plus the abundance of the stars as a function of their mass, are important in determining the interstellar abundance of the nuclides.

In addition, the probabilities of having X^- particles bound to various nuclei will be different in each zone of the stellar interior, both because of the variation in temperature and because of the differing nuclidic abundances (see Table 1). For example, the temperature in the periphery of a star probably never becomes high enough even to ionize He^x. Thus, any estimate of the extent to which stellar processing has redistributed the X^- particles from their primordial binding sites requires not only the information required for predicting nuclidic abundances but the information on X^- reaction rates discussed above as well. Thus we have applied a model (*16*) for galactic chemical evolution that we have modified to include X^-; although it is a fairly simple model, it should give an adequate estimate of the optimum X^- binding environments. We will be trying to describe the ISM about 10 billion years after the Big Bang, because that is thought to be the material from which Earth was formed.

Table 1. Stages of stellar burning.

Stage	Typical temperature (K)	Typical denisty (g/cm^3)	Abundant elements
H burning	2×10^7	1×10^2	H, He, C, N, O
He burning	2×10^8	1×10^4	He, C, N, O
C burning	5×10^8	1×10^5	C, O, Ne
Ne burning	1×10^9	1×10^6	O, Ne, Si
O burning	2×10^9	1×10^6	O, Si
Si burning	4×10^9	1×10^7	Si, Fe

Any model of chemical evolution requires an "initial mass function," that is, a description of the distribution of stars as a function of mass. A mass-dependent model of stellar evolution must be invoked to describe the features of stellar processing discussed above. Some function that describes the rate of star formation out of the ISM is also required. Finally, the rate at which gas from the galactic halo is falling into the plane of the galaxy must also be included in the equation. Therefore, the simplest equation that might describe the rate of change of the total amount of interstellar gas, m_g, is

$$dm_g(t)/dt = -Q(t) + R(t)Q(t) + f(t) \quad (5)$$

where $Q(t)$ is the (mass-integrated) stellar birth rate, $R(t)Q(t)$ is the total ejection rate at time t from those stars that had formed before, and $f(t)$ is the rate of mass infall from the galactic halo. Included in $Q(t)$ is a vast amount of information about stellar evolution. That subject, however, is beyond the scope of this study; more detailed descriptions can be found in (*16*).

The equation describing the rate of change of the amount of gas of the ISM that is in nuclide i, m_i, is

$$\begin{aligned} dm_i(t)/dt = &-\lambda_i{}^g m_i(t) \\ &- [m_i(t)/m_g(t)]Q(t) + [m_i(t)/m_g(t)]R(t)Q(t) \\ &+ \gamma_i{}^{net}(t)[1 - R(t)]Q(t) + I_i f(t) \end{aligned} \quad (6)$$

where $\lambda_i{}^g$ is the decay rate of nuclide i in the ISM if nuclide i is radioactive, $m_i(t)$ is the mass of nuclide i in the ISM (and so m_i/m_g is the fraction of the mass in nuclide i), $\gamma_i{}^{net}(1 - R)Q$ is the net stellar ejection rate of nuclide i, and I_i is the fraction of the infalling extragalactic gas that is in nuclide i. If nuclide i could be produced by radioactive decay from other nuclides in the ISM, a term representing that effect should also be included in Eq. 6.

We have performed calculations using this chemical evolution model to predict the most probable environment for X^- particles in the ISM after 10 billion years of galactic evolution. The results of these calculations are presented under the "Primordial infall" columns in Table 2. The ratios displayed are our predicted abundances of the X^--bearing nuclides in the first column (having charge number Z) to the solar abundances (*17*) of normal nuclides (having charge number $Z - 1$) in the second column. These are relative ratios, as an overall normalization factor, related to the total abundance of stable X^-, must be applied to the ratios. This ratio gives the basic indicator of the feasibility of an X^- search in any chemical environment; one seeks a nuclide that is found to be likely, after consideration of stellar processing, to have embedded X^-

particles, but that is situated in the periodic table one charge unit above a nuclide with a very low abundance. We have included in Table 2 only those X^--bearing nuclides that are fairly abundant, because those are well reproduced, at least up to Fe (to about 30%) by the chemical evolution model (*16*). Other criteria, such as the inherent sensitivity of any type of search, can also be important. From the principal criterion, however, B appears to be the most favorable candidate by several orders of magnitude, primarily because its abundance is very small, and C appears to be a good "sink" for X^- particles. The infinite X^- mass case suggests that a search in H would have to be better than one in B by seven orders of magnitude, if the infalling material is primordial, to be competitive. Another good case is provided by F; an experiment in that chemical environment would have to be more sensitive by three orders of magnitude than that in B to be competitive.

Because the infall rate assumed for this model (*16*) is generally thought to be large, its results indicate a kind of upper limit on the amount of unprocessed material that presently exists in the galaxy. For the "Primordial infall" results shown in Table 2, this matter was assumed to contain the primordial distribution of X^-. In order to see how the X^- distribution would change if the infalling material had been processed (but preserving the success of the model in reproducing present-day galactic parameters), we also calculated abundances assuming that the infalling gas had all its primordial $^4He^x$ converted to $^{12}C^x$. [This would also accommodate the assertion, by Corbelli and Salpeter (*18*), that the infalling gas might not be primordial.] These results, given under "Processed infall" in Table 2, indicate a reduced $^4He^x$ abundance and an increased $^{12}C^x$ abundance, but other abundances change very little. The resulting ranges are indicative of the uncertainties in the results of this calculation; it is encouraging that there is no qualitative change. However, these ranges also indicate that the search in B for X^- particles may be inherently ten orders of magnitude better suited to this search than that performed in H.

Of course, these results could be affected by events not included in the model. For example, supermassive Population III (Pop III) stars (*19, 20*), sometimes discussed as precursors to present-day stars, might have existed before galaxy formation. The high temperature they would attain over almost all of their mass would lower the He^x abundance, first because of attrition of He^x from ionization of the X^- particles followed by subsequent recapture to heavier nuclides, and second because of conversion by nuclear reactions of He^x to $^8Be^x$ or heavier nuclides. The trend of the X^- abundances from this scenario is suggested by our "Processed infall" results, although the abundance of He^x would decrease greatly, and the abundances of C^x and heavier X^--bearing nuclides would increase, from even the "Processed infall" values. Because Pop III stars with X^- particles would produce C (or C^x) very efficiently, this process could be responsible for the observation that even the oldest stars in the universe seem to have considerably more C than is generally thought to have been produced by Big Bang nucleosynthesis.

Searches for X^- Particles

A number of experiments have been performed on terrestrial material to search for X^- particles. We will discuss the implications of those that appear to set the tightest limits on the terrestrial abundance of X^- particles. Of particular interest is $^4He^x$, because its abundance in H has been found (*4, 5*) to be extremely small. Although our predictions do not suggest that H is a particularly good place to look for X^- particles (see Table 2), the extraordinary sensitivity of that experiment overcomes that deficiency to some extent. Our galactic, chemical evolution model suggests that the abundance of $^4He^x$ would decrease only a small amount from its primordial value as a result of stellar ionization and burning, the latter because of the $^4He^x + {}^4He \rightarrow {}^8Be^x + \gamma$ reaction. However, the possibility of processing scenarios, for example, in Pop III stars, which could have reduced the abundance of He^x by both mechanisms before formation of present-day galaxies, could lower the abundance from our predicted values for He^x by many orders of magnitude. Furthermore, it has recently been noted (*21*) that a dilution of the published abundance from the "anomalous H" experiments should be invoked, reducing their sensitivity by several orders of magnitude for the heaviest masses examined, because of a slower diffusion rate of more massive particles from the source material (*7*). Thus the extremely tight abundance limits (*4, 5*) for H of anomalous mass do not necessarily provide as stringent a limit on the possible abundance of X^- particles as they indicate. Although the results from those experiments are very impressive, the caveats peculiar to He^x render the meaning of those limits, as they apply to X^- particles, quite uncertain.

Turkevich *et al.* (*6*) conducted an ingenious search beginning with a C sample, which, when radiated with neutrons, would have had its $^{14}N^x$ converted via (n,p) reactions to $^{14}C^x$. Selection of anything chemically like B from their sample, followed by observation of possible beta decays of $^{14}C^x$, produced their limit. The $^{14}C^x$ would have undergone beta decay with a sufficiently long half-life that no competition would be expected from other reaction products. Turkevich *et al.* found no evidence for X^- particles at a level of 2×10^{-15} per nucleon in C nuclei with a sensitivity that goes to X^- masses as high as 10^5 GeV but that has a decreasing sensitivity at lower masses.

An accelerator mass spectrosocpy search (*21*) that examined a variety of elements, Li, Be, B, C, O, and F, for nuclei of anomalous mass (which would include nuclei with embedded X^- particles) with masses above about 100 GeV, also did not find any evidence for such, at abundance limits from 1×10^{-13} (for Be) to 1×10^{-20} (for C) anomalous nuclei per normal nucleus. The B case is worthy of special comment. Because, as our calculations have shown, a significant amount of C^x would be produced in stellar processing, C^x is chemically similar to B, and B is a rather rare element, a search of $[C^x]/[B]$ should afford the best possibility for finding X^- particles. Furthermore, the processes discussed above that could deplete He^x from its primordial abundance would increase that of C^x and so would not dilute any limit obtained for B. Thus the abundance limit (*21*) for that sample, 1×10^{-12} to 1×10^{-14} anomalous particle per B nucleus (the limit depends on the X^- mass), may represent the best test yet, particularly in a model-independent sense, of the existence of X^- particles. It should be noted, however, that the experiment was sensitive only to masses above 100 GeV.

Norman *et al.* (*9*) conducted an X^- search in the Fe region; they found, by means of a search for gamma rays of anomalous energy resulting from bombardment of their sample, no evidence for X^- bound to nuclei at an abundance level of 1.2×10^{-12} X^- per nucleon in Fe nuclei. Their experiment was sensitive to large X^- mass; their lower mass limit was about 100 GeV. The Si burning phase of stellar evolution synthesizes nuclei up to Fe and Ni through a complicated coupled set of nuclear reactions, some of which liberate ^4He nuclei from some nuclei that are subsequently captured by heavier nuclei. This process operates at a temperature of several billion kelvin but for a very short time, so there would be a very small probability of ionizing the X^- particles bound to any nuclei as massive as Fe. However, these particles would be liberated by the lighter nuclei in that stellar region and would tend to be captured by the nuclei close in mass to Fe being formed. Even so, our chemical evolution results suggest that the Fe region is not as good a place to look for X^- particles bound to nuclei as some of the lighter nuclei,

because most of the nuclei of which the stars are composed do not progress as far as Si burning. However, because Fe is considerably more abundant than Mn, the element that would be chemically most like Fe^x, the Fe experiment (*9*) did have some chance of detecting X^- particles bound to nuclei.

Another nucleus, Tc, was observed by Cahn and Glashow (*8*) to be a good place to search for X^- nuclei because of the nonexistence of stable Tc isotopes. The "natural abundance" of Tc on Earth results from several rare processes, for example, cosmic-ray interactions, reactions induced by light fission products, or fission fragments, and so is very small. Ru^x would have the chemical properties of Tc, and Ru is reasonably abundant for its region of the periodic table. Experiments (*22*) are presently under way that ultimately will make it possible to search for Ru^x in Tc.

Spectrosocpic Detection of X^- Particles

The desirability of using any atom for an X^- search depends both on its probability of containing an X^- and on its amenability to high-accuracy experiments. As noted above, the presence of an (assumed massive) X^- in a nucleus with charge Z would change the chemistry of that atom (with corresponding loss of an electron) to that of the atom with charge $Z - 1$. A search for nuclei containing X^- particles poses two major experimental challenges. First, because the mass of the X^- is unknown, it would be difficult to distinguish the signature of an X^--containing ion from an "impurity" ion. Secondly, X^- particles are unlikely to be very abundant. Thus, in any successful search for them we must use a technique with enough sensitivity to mass differences to identify nuclei containing X^- particles but with sufficient limitation of that sensitivity to restrict the search to an acceptable region.

Mass Dependence of the Spectrum

Thus we propose to observe the electronic spectrum of a diatomic molecule containing one heavy and one light nucleus. We can detect its spectrum with good sensitivity by using resonantly enhanced, multiphoton ionization spectroscopy (REMPI) with a tunable laser source. The molecule should be easily obtainable and it should be possible to maintain it at equilibrium in the gas phase at pressures of at least 1 torr at ambient temperature. We believe that the hydrogen halides (HF, HCl, HBr, and HI) come very close to satisfying these criteria. Furthermore, HF (with HNe^x being the X^--bearing molecule) appears to contain the second most favorable nuclide F for containing X^- particles.

In REMPI, laser photons having insufficient energy to ionize the molecules in the sample, but with energy such that one or more of them is tuned to the energy difference between the ground state and a well-defined excited state, promote the molecule to that excited state. An additional laser photon (or photons) can then promote an electron to a continuum state, producing an ion. Detection of the ions thus produced provides the signature of the desired process.

On the basis of the known properties of hydrogen halide spectra, we will examine, for a variety of possible X^- masses, the expected spectral shifts. Generally we can write the expected shift, δS, of the lines of the REMPI spectrum as

$$\delta S = \delta E + \delta W + \delta B f(J) \quad (7)$$

The δE term denotes the shift in the molecular electronic energy levels upon a change in mass, but not in chemical identity, of the constituent atoms. Roughly speaking, it arises from the fact that the electrons are much lighter than nuclei. The δW term indicates the mass dependence of the vibrational frequencies. The final term indicates the mass dependence of the rotational frequencies; $f(J)$ is a function of the rotational quantum number J. The most likely spectral transition to probe in HF is the two-photon, excited transition between the ground $X\ ^1\Sigma^+$ electronic state and the excited $B\ ^1\Sigma^+$ state, for which the rotational selection rules allow $\Delta J = 0, \pm 2$ transitions. Because the $\Delta J = 0$ transition is simplest and is likely to be the most intense, we shall confine our attention to it. In this case $f(J) = 2(J + 1)$.

Van Vleck (*23*) was the first to consider isotopic corrections to electronic spectra, and his work suffices for our present purposes. He recognized that one could express δE in Eq. 7 as a sum of three terms

$$\delta E = P + Q + U \quad (8)$$

where P and Q result from the coupling of the electronic and nuclear degrees of freedom in a molecule, that is, they are (some of) the terms neglected in the Born-Oppenheimer approximation. Howev-

Table 2. Predicted relative abundances of X^--bearing nuclides in the ISM at the time of formation of the solar system. Abun denotes abundances: the $Abun_\odot$ are solar nuclidic abundances, and $Abun(X^-)$ is the total abundance of X^- particles. "Primordial infall" assumes that the infalling galactic material has the primordial X^- distribution among the nuclides. "Processed infall" assumes that all He^x in the infalling material has been converted to C^x.

Nuclide (Z)	Nuclide ($Z - 1$)	$Abun((Z^x)/[Abun_\odot\ (Z - 1)\cdot Abun(X^-)]$			
		Primordial infall		Processed infall	
		$M_x = \infty$	$M_x = 1$ GeV	$M_x = \infty$	$M_x = 1$ GeV
He^x	H	5.4×10^{-1}	7.7×10^{-1}	4.4×10^{-2}	3.9×10^{-2}
C^x	B	3.2×10^{7}	3.3×10^{7}	6.1×10^{8}	1.3×10^{8}
N^x	C	8.2×10^{-1}	4.0	2.2×10^{-1}	1.0
O^x	N	4.9	4.6×10^{1}	4.4	1.4×10^{1}
Ne^x	F	2.4×10^{3}	3.9×10^{4}	2.1×10^{3}	3.0×10^{4}
Mg^x	Na	9.5	1.5×10^{2}	8.6	1.2×10^{2}
Si^x	Al	1.1×10^{1}	5.2×10^{1}	3.6×10^{1}	3.7×10^{1}
S^x	P	4.5×10^{1}	1.8×10^{2}	1.6×10^{2}	1.3×10^{2}
Ar^x	Cl	1.3×10^{1}	7.1×10^{1}	4.6×10^{1}	6.0×10^{1}
Ca^x	K	1.4×10^{1}	6.4×10^{1}	5.0×10^{1}	5.4×10^{1}
Cr^x	V	1.3×10^{2}	1.0×10^{2}	4.8×10^{2}	9.2×10^{1}
Fe^x	Mn	4.4×10^{2}	3.1×10^{2}	1.6×10^{3}	2.8×10^{2}
Ni^x	Co	4.7×10^{2}	9.8×10^{2}	1.7×10^{3}	9.7×10^{2}
Zn^x	Cu	4.4	5.1×10^{1}	1.7×10^{1}	4.8×10^{1}
Sn^x	In	2.2×10^{1}	6.1×10^{4}	8.2×10^{1}	6.1×10^{4}
Pb^x	Tl	1.5×10^{1}	1.1×10^{4}	5.8×10^{1}	1.2×10^{4}

er, for accurate determination of isotope effects, these terms must not be neglected. The last term in Eq. 8, U, may be thought of as the remaining isotope effect in a molecule even if it were not rotating or vibrating. It is akin to, but more complicated than, the electronic isotope shifts in atoms.

The direct calculation of any of the terms of δE would be formidable; fortunately it is not necessary. Van Vleck has shown that P and Q are proportional to μ^{-1} and U (the smallest of the three parts of δE for the hydrogen halides) is approximately so. Thus we assume that δE is proportional to μ^{-1}, where μ is the molecule's reduced mass, that is,

$$\mu = M_1M_2/(M_1 + M_2) \quad (9)$$

where M_1 and M_2 are the masses of the two nuclei. Note that μ is fairly insensitive to the heavy mass M_2 so long as $M_2 >> M_1$, a condition always satisfied for the hydrogen halides.

Using finite differences, one can express the electronic isotope shift δE_x of an X^--containing hydrogen halide in terms of the known value of the shift, δE_D, observed for the deuterated halide

$$\delta E_x \approx \delta E_D\left[\frac{1 - (\mu_0/\mu_x)}{1 - (\mu_0/\mu_D)}\right] = \delta E_D\left[\frac{1 - \xi_x}{1 - \xi_D}\right] \quad (10)$$

where $\xi_x = \mu_0/\mu_x$, with an analogous definition for ξ_D. We shall assume that the transition observed is a two-photon, resonantly enhanced transition to the vibrationless level of the $B\ ^1\Sigma^+$ state of HF. Using literature values (*24, 25*) for the H and D species, we find $\delta E_D(HF) = 47.4\ cm^{-1}$. These results give the possible values of δE_x for various mass values of X^- that are listed in Table 3.

In addition to the electronic isotope shift, there exists a shift in the vibrational energy levels. The vibrational frequency of a molecule in a given electronic state is given to a reasonable approximation by

$$\omega_o = (2\pi)^{-1}(k/\mu_0)^{1/2} \quad (11)$$

where k is the molecule's force constant. For simplicity, we shall assume harmonically spaced vibrational levels with quantum numbers ν' in the B state and ν'' in the X state. Thus

$$\delta\omega_x = [(\nu'' + 1/2)\omega_o'' - (\nu' + 1/2)\omega_o'](1 - \xi^{1/2}) \quad (12)$$

The values of ω_o' and ω_o'' (the single and double primes indicate the upper and lower states, respectively) are obtained from the known frequencies (*24, 25*) of the X and B states of HF. The values of $\delta\omega_x$ thus obtained are tabulated in Table 3.

The final isotopic correction pertains to the rotational energy levels. The simplest formula for them is

$$E = BJ(J + 1) \quad (13)$$

with B the rotational constant of HF, given by a constant times μ^{-1}. Using Eq. 9, one can show that the coefficient of the last term of Eq. 7 is given by

$$\delta B_x = (B'' - B')(1 - \xi_x) \quad (14)$$

The rotational constants in the two states are known (*24, 25*) from the HF spectrum. The values of δB_x obtained from Eq. 14 are listed in Table 3.

The last column of Table 3 gives the total shift to be expected for an HF-like species containing an X^- of a given mass. The calculation is made for the $J = 4$ transition, one of the stronger rotational transitions within the spectrum.

Table 3. Mass-dependent shifts (in reciprocal centimeters) for the 0_0^0 band* of the $B\ ^1\Sigma^+ \rightarrow X\ ^1\Sigma^+$ transition in HF. The frequency of the 0_0^0 band at a one-photon wavelength of 240 nm is tabulated for convenience; however, this transition is likely to be very weak because of poor Frank-Condon overlap. A higher ν' level provides a higher transition probability; for example, the 14-0 band at 208 nm was easily observed in the absorption spectrum of HF. The appropriate $\delta\omega_x$ (the only term to change appreciably) can be calculated from Eq. 12. All the qualitative conclusions cited in the text regarding observation and sensitivity will remain unchanged for the various ν' levels. One further caveat about the numbers in this table is appropriate. The harmonic oscillator and rigid rotor approximations are probably suspect to the level of precision calculated. However, inclusion of higher order spectral corrections is straightforward and again would not alter any of the conclusions.

X^- mass increment*	ξ_x	δE_x	$\delta\omega_x$	δB_x†	δS
0	1.000000				
10	0.982628	1.8	13.0	5.9	20.7
25	0.971376	3.0	21.5	9.7	34.2
50	0.963495	3.9	27.4	12.4	43.7
100	0.957667	4.5	31.9	14.3	50.7
200	0.953995	4.9	34.7	15.6	55.2
∞	0.949624	5.6	38.0	17.1	60.7

*Incremental mass increase (in gigaelectron volts) of an Ne^x nucleus compared to ^{19}F. †Calculated for the $\Delta J = 0$ transition with $J = 4$.

Sensitivity

It is easiest to compare the REMPI experiment to a conventional mass spectrometry experiment. The efficacy for producing ions from neutral molecules can be made close to unity in either experiment. Indeed, REMPI has often been touted as a technique for single atom detection. Assuming that a 1-torr sample of volume 0.1 cm^3 can be converted to ions and collected with unit efficiency per laser shot gives a total ion count of 3×10^{15}. Most detector electronics would, of course, be saturated by such a count rate, but the implication is that at least one in 3×10^{15} molecules could be detected for each laser shot. Assuming a modest repetition rate of 33 Hz gives a sensitivity of one in 1×10^{17} per second.

Two factors could diminish this sensitivity. Because we are exchanging a wide scan in mass space for a narrow one in frequency space, we will detect only molecules in a particular rotational, vibrational level. Thus the sensitivity is reduced by the fraction of molecules in a given level. For HF in the most populated rotational level, this would reduce the sensitivity to about 1×10^{16}. However, because this type of experiment spans all masses, it does not have the mass limitations that characterize the other experiments that have been performed. In addition, it has the capability of providing a striking confirmation that X^--bearing nuclei have been observed. Initially all ions would be detected for maximum sensitivity. If a frequency resonance from an X^--containing molecule is suspected, its time-of-flight mass spectrum could be obtained to verify the isotopic distribution of the X^--containing nucleus, which, for the HF sample, should be very similar to that of the Ne isotopes.

The other sensitivity-reducing factor that must be taken into account is background. Barring accidental coincidences, this is mainly the "noise" accompanying the wings of the REMPI lines of the ordinary molecule. If no attempt is made to eliminate Doppler broadening in the experiment, one can expect (in fundamental photon units) a Doppler half width of 0.06 cm^{-1} at 300 K. Assuming a most likely shift of 44 cm^{-1} (approximately 50 GeV mass for X^-) yields a shot noise factor less than 1 count per second from the Gaussian wing of the Doppler-broadened line of the ordinary molecule at the position of the X^- molecule resonance. This clearly has little effect on the ultimate sensitivity of the experiment.

On the other hand, the wing of the Lorentzian component of the actual profile expected will probably contribute more noise. If we assume a half width of the excited state of a few megahertz, the shot noise yield at the line position of the X^--containing molecule suggested above would be of the order $10^4\ sec^{-1}$. This would reduce

the sensitivity in a simple experiment to approximately one in 10^{13} for a 1-s integration time.

Numerous improvements in the sensitivity of this very simple scheme can be envisioned. (i) Overall 100-s counting times are reasonable. (ii) The material in the sample can be enriched in heavy isotopes, which could readily improve the sensitivity by several more orders (at least three) of magnitude. (iii) Gated detection of the ions, following a simple time-of-flight scheme that rejects all ions with masses less than 30 GeV (mass HF + 10 GeV), would greatly reduce wing noise. Rejection ratios for HF should be at least 10^3. Thus a realistic ultimate sensitivity that might be achieved for REMPI searches for HNe^x in HF is roughly 1×10^{-20}.

Conclusions

Our considerations of galactic chemical evolution have provided new insight into the optimum chemical environments in which to search for X^- particles, with B (corresponding to C^x) being the best candidate of the stable nuclides. Other good candidates include F (corresponding to Ne^x) and Tc (corresponding to Ru^x).

Although a variety of experiments have been conducted to search for stable X^- particles, none has successfully located them as yet. The least ambiguous abundance limit appears to be that obtained for a B sample, 1×10^{-12} to 1×10^{-14} for X^- masses from 100 to 10,000 GeV, although the limit obtained from the H sample also infers some interesting information. Laser spectroscopic techniques can be used to search for stable X^- particles. Further searches of the types already performed for such particles should be undertaken, in view of the importance that their discovery would have on particle physics and possibly on astrophysics.

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Research Articles

DNA Looping Generated by DNA Bending Protein IHF and the Two Domains of Lambda Integrase

LINA MOITOSO DE VARGAS, SUNGHOON KIM, ARTHUR LANDY

The multiprotein-DNA complexes that participate in bacteriophage lambda site-specific recombination were used to study the combined effect of protein-induced bending and protein-mediated looping of DNA. The protein integrase (Int) is a monomer with two autonomous DNA binding domains of different sequence specificity. Stimulation of Int binding and cleavage at the low affinity core-type DNA sites required interactions with the high affinity arm-type sites and depended on simultaneous binding of the sequence-specific DNA bending protein IHF (integration host factor). The bivalent DNA binding protein is positioned at high affinity sites and directed, by a DNA bending protein, to interactions with distant lower affinity sites. Assembly of this complex is independent of protein-protein interactions.

PROTEIN-INDUCED DNA BENDING AND PROTEIN-MEDIATED DNA looping contribute to the overall structure of higher-order complexes in several transcription, replication, and

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