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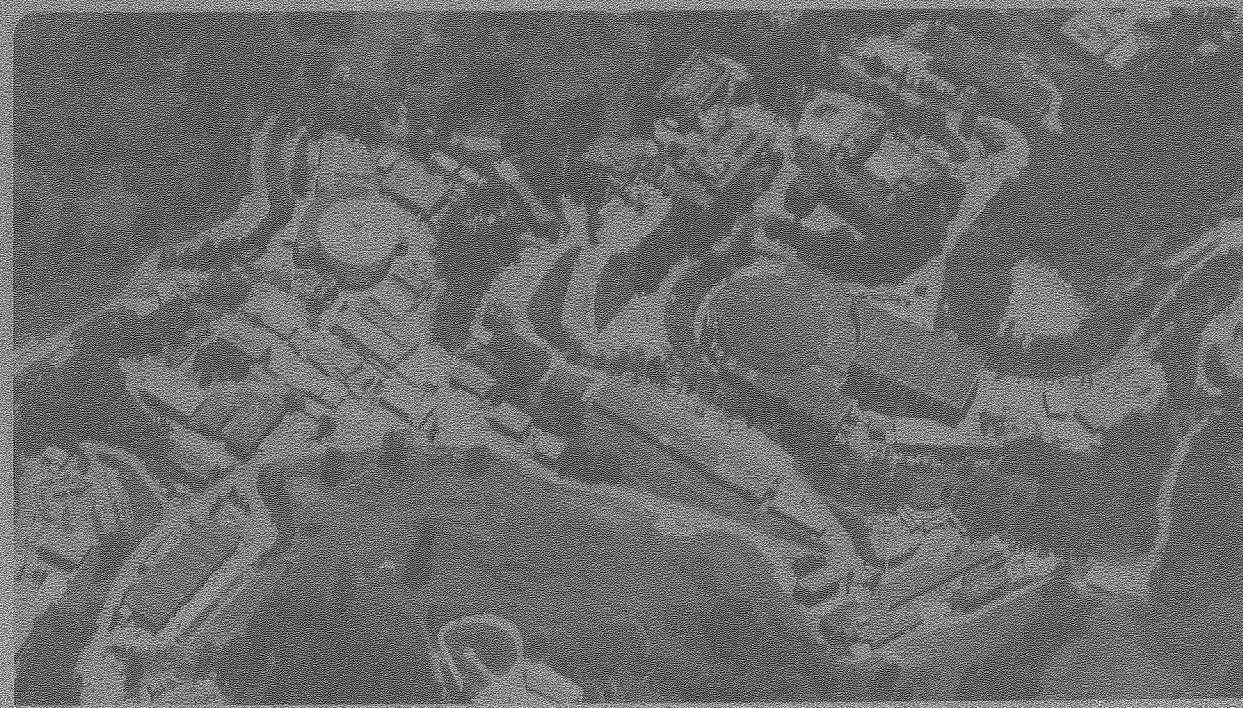
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December 1980



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CHEMICAL SIGNATURES FOR SUPERHEAVY ELEMENTARY PARTICLES

Robert N. Cahn
Lawrence Berkeley Laboratory, University of California
Berkeley, California 94720

and

Sheldon L. Glashow*
Center for Particle Theory
Massachusetts Institute of Technology
Cambridge, Massachusetts 02139

ABSTRACT

Models of dynamical symmetry breaking suggest the existence of many particles in the 10 GeV to 100 TeV mass range. Among these may be charged particles, $X^\pm$, which are stable or nearly so. The X^+ 's would form superheavy hydrogen, while the X^- 's would bind to nuclei. Chemical isolation of naturally occurring technetium, promethium, actinium, protactinium, neptunium or americium would indicate the presence of superheavy particles in the forms RuX^- , SmX^- , $^{232}ThX^-$, $^{235,236,238}Ux^-$, $^{244}PuX^-$, or $^{247}CmX^-$. Other substances worth searching for include superheavy elements with the chemical properties of B, F, Mn, Be, Sc, V, Li, Ne, and Tl.

All tangible matter appears to be composed of protons, neutrons, and electrons. The presence, however, of trace amounts of super-heavy (say 100 GeV to 100 TeV) stable particles has not been excluded. Stringent limits have been set on hydrogen isotopes up to 17 GeV¹ and oxygen isotopes up to about 50 GeV.² We insist that there is substantial motivation for extending mass searches to much greater masses both on general grounds and on indications from some models of unified weak, electromagnetic, and strong interactions.³

Historically, the chemical search for new varieties of stable matter on Earth terminated with the development of a successful model of the atomic nucleus in the 1930's. Under the constraint that all matter is composed of electrons and nucleons, there is little incentive to search for such things as naturally occurring "isotopes" of lithium or americium with atomic weights of thousands. The dogma of recent decades has it that new particles are to be found only at large accelerators or in cosmic rays by particle physicists, and certainly not in mines by chemists. However, our understanding of fundamental theory remains so limited that we dare not exclude the possible existence of very heavy stable particles as rare but natural constituents of atomic nuclei. They may be far too heavy to produce and study with existing accelerators. Perhaps these new particles do exist, are of potential technological significance, and cohabit with us on Earth. It is an important truism that if we do not search for these particles, we will not find them. In this connection, we should recall the belated discovery of argon as a one-percent constituent of the atmosphere. Its discovery, less than a century ago, is the kind of outrageous surprise we may still

* Permanent address: Lyman Laboratory of Physics, Harvard University, Cambridge, MA 02138.

anticipate today.

The remarkable success of the $SU(2) \times U(1)$ theory⁴ of electro-weak interactions has provided the foundation for more ambitious programs incorporating the strong interactions as well.⁵ In their simplest forms, these theories provide no insight into the masses of the W and Z bosons, which are predicted to be about 80 - 90 GeV. However, there are proposals to extend these models so that the mass scale of the W and Z bosons arises dynamically.⁶ In such models there are other particles with masses in the 10 GeV to 100 TeV range.⁷ Indeed, independent of the validity of any of the proposed models, it can be argued that if the W and Z bosons masses are a fraction of a TeV, there may well be other particles in this mass range.

The prospects for the existence of heavy stable particles can be assessed by considering specific models. In current models of dynamical symmetry breaking, an additional exact non-Abelian gauge symmetry - technicolor - is hypothesized.⁷ Let us suppose for the moment that this new interaction is an $SU(4)$ gauge interaction, quite independent of the known $SU(3)_{\text{color}} \times SU(2)_L \times U(1)$ interaction. All known quarks and leptons are technicolor singlets, but there would be additional very heavy fermions, techniquarks (F), which would transform as a $\underline{4}$ of $SU(4)$. The energy scale of the technicolor interactions is determined by the requirement that the masses of the ordinary W and Z bosons agree with the predictions of the usual $SU(2) \times U(1)$ theory. Although certain meson states of bound techniquarks and anti-techniquarks might be as light as a few GeV, a more typical mass scale is 1 TeV. In particular, one would expect technibaryons composed of four techniquarks forming

a technisinglet with integral spin. Unlike the meson composed of techniquarks, these technibaryons could be quite stable.

Whether the technibaryons would be stable cannot be determined in the absence of a rather complete model including both technicolor and grand unification and no such realistic model exists. Nonetheless, building on theoretical studies of mechanisms for ordinary (!) proton decay, we can identify several distinct possibilities for the technibaryons. We assume that there are gauge bosons with masses $M_G \approx 10^{14}$ GeV which can transform techniquarks into anti-techniquarks or into ordinary fermions. The effective four-fermi interaction could turn $FFFF$ into $\overline{FFFF}$ which can be an $SU(4)$ singlet. Extrapolating from proton lifetime predictions, we could expect the technibaryon lifetime to be $(m_p/M_T)^5 \times 10^{31}$ yrs., where $M_T \simeq 1$ TeV is the technibaryon mass. This yields an estimate of 10^{16} yrs. Severe limits can be placed on the existence of technibaryons of this kind, since no such decays have been observed in cosmic ray experiments.⁸

Perhaps technibaryon decay cannot occur through a single exchange of superheavy gauge boson. This would be the case if technicolor were an $SU(5)$ interaction. The technibaryons would have the structure $FFFFF$ and the simplest effective Lagrangian which could contribute to technibaryon decay would be

$$\mathcal{L} \propto \overline{FFFFF} F / M_G^5. \quad \text{This would give a lifetime of roughly } ^9$$

$$T \simeq M_G^{10} / M_T^{11} \simeq 10^{76} \text{ yrs.}$$

In some schemes, the technibaryons could decay into ordinary particles through the exchange of "sideways" gauge bosons which could be much lighter than the grand unification mass. In such schemes, the technibaryons would not be stable.⁹

We shall not attempt to estimate the concentration of superheavy particles,¹⁰ but simply assume that some are created in the big bang and that some fraction remain after particle - anti-particle annihilation. We shall assume also that the surviving superheavy particles are integrally charged: $X^\pm$. These particles will be assumed to have only electromagnetic interactions with conventional matter, but this assumption is probably not too important and most of our expectations would be unaffected by including some conventional hadronic interactions. For our purposes, it will not matter whether the X 's are fermions or bosons. Of course, if there are stable neutral X 's the presence or absence of hadronic interactions would be of primary importance for them.

The X^+ particles would behave as protons, (possibly) lacking hadronic interactions. They would thus be found as superheavy hydrogen. The searches for such isotopes would not have found the X^+ 's if their mass is greater than about 17 GeV.

The X^- particles should they exist, would be distributed among the various nuclei. The binding of the X^- 's to nuclei can be estimated using a simple model in which the nucleus is regarded as a sphere with uniform charge density, and in which the mass of the X^- is assumed to be much larger than that of the nucleus. The Hamiltonian

is then

$$H = \frac{p^2}{2M_N} - \frac{3Z\alpha}{2r_0} + \frac{Z\alpha}{2r_0} \left(\frac{r}{r_0} \right)^2, \quad r < r_0. \quad (1a)$$

$$= \frac{p^2}{2M_N} - \frac{Z\alpha}{r}, \quad r > r_0. \quad (1b)$$

where $r_0 \approx 1.2 A^{1/3}$ F. is the nuclear radius and where M_N is the mass of the nucleus. For large nuclei, the X^- is always inside r_0 and the problem is just that of a simple harmonic oscillator. The binding energy is

$$E_b = \frac{3}{2} \frac{Z\alpha}{r_0} - \frac{3}{2} \left(\frac{Z\alpha}{M_N r_0^3} \right)^{1/2} \\ = (1.8ZA^{-1/3} - 8.8Z^{1/2}A^{-1}) \text{ MeV.} \quad (2)$$

For fixed A , this pushes stability towards higher Z :

$$\frac{\partial E_b}{\partial Z} = (1.8A^{-1/3} - 4.4Z^{-1/2}A^{-1}) \text{ MeV.} \quad (3)$$

$$\text{For } A \approx 100, \quad \frac{\partial E_b}{\partial Z} \approx 0.4 \text{ MeV.}$$

In the limit of small nuclei, the problem is Coulombic and the binding energy is simply

$$E_b = \frac{1}{2} (Z\alpha)^2 M_N. \quad (4)$$

In between these extremes it suffices to treat the problem variationally, using a wave function of the form $\psi \sim e^{-\gamma r/r_0}$.

The result is that

$$E_b = \left(M_N r_0^2 \right)^{-1} \lambda(Z\alpha M_N r_0), \quad (5)$$

where the function $\lambda(a)$ has the limits

$$\lambda(a) \rightarrow \frac{1}{2} a^2 - \frac{2}{5} a^4 \quad (a \rightarrow 0), \quad (6a)$$

$$\lambda(a) \rightarrow \frac{3}{2} a - \sqrt{3}a \quad (a \rightarrow \infty). \quad (6b)$$

The $a \rightarrow 0$ limit gives the correct first order perturbation since the trial wave function is exact for the Coulomb problem. The $a \rightarrow \infty$ limit is very nearly correct $\left(\sqrt{3} \approx \frac{3}{2}\right)$. The function $\lambda(a)$ is shown in Fig. 1. The electrostatic binding energies for a few low A nuclides are shown in Table I.

As a general rule, the X^- will bind to the highest Z nuclei accessible. During the big bang, X^- 's would have bound to ${}^4\text{He}$ as soon as it was formed. Moreover, while

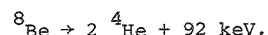
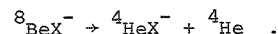
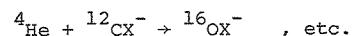
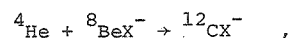
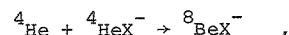
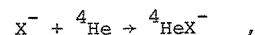


Table I shows that the supernucleus ${}^8\text{Be}X^-$ would be stable against the decay



Thus, even in the big bang the X^- 's would be processed into high Z nuclei:



Of course, the X^- 's, would be processed into heavy nuclei as well in the other processes which are responsible for ordinary nucleosynthesis.

Through nucleosynthesis the X^- 's would be distributed among the nuclides. The combinations $\text{Ru}X^-$ and $\text{Sm}X^-$ are of special interest because they have the charges of the nuclei Tc and Pm, which are not found in nature since their most stable isotopes have half-lives of only 4.2×10^6 yrs. (${}^{98}\text{Tc}$) and 17.7 yrs. (${}^{145}\text{Pm}$). Thus any technetium or promethium found by chemical separation techniques should be suspected of being in fact $\text{Ru}X$ or $\text{Sm}X$ respectively. If a sufficiently large amount were isolated chemically, it would be possible to demonstrate the presence of superheavy material simply by density measurements. With lesser amounts, neutron activation could provide a definitive indication: the superheavy technetium would have the chemistry of technetium but not its nuclear chemistry.

The best procedure for searching for technetium may be to examine material containing rhenium, which most resembles it chemically. This was the procedure which led to the spurious discovery of natural technetium by Noddack, Tanka and Berg who named it "masurium".¹¹ For promethium, the best place to look may be where the neighboring lanthanides, neodymium and samarium are found. This, again, once led to the spurious discovery of naturally occurring promethium by Hopkins who called it "illinium".¹¹

It is necessary to check that the superheavy nuclides $\text{Ru}X^-$ and $\text{Sm}X^-$ would be stable against β decay since the presence of the X^- shifts stability towards higher Z. The $\text{Ru}X^-$ would have about 0.4 MeV less electrostatic binding than the $\text{Rh}X^-$ isobars. A check of binding energies shows that the A = 96, 98, 99, 100, 101, 102 and 104 isotopes would indeed remain stable even with the addition of the X^- . For Sm,

the electrostatic shift is about 0.34 MeV. The $A = 144, 149, 150, 152$ and 154 isotopes would remain stable. The $A = 146, 147$ and 148 isotopes of Sm are α -emitters with half-lives of 1×10^8 yrs., 1×10^{11} yrs. and 8×10^{15} yrs. respectively. The superheavy analogs $^{146,147,148}\text{SmX}^-$ would also be α -emitters, but would release about 0.5 MeV less energy and would thus have considerably longer half-lives. We thus conclude that the superheavy analogues of the stable forms of Ru and Sm would themselves be stable.

A situation analogous to that of technetium and promethium occurs for actinium and protactinium. Although no isotope of uranium or thorium is absolutely stable, ^{238}U and ^{232}Th have half-lives of 4.5×10^9 yrs., and 1.4×10^{10} yrs. respectively. Thus the superheavy nuclides $^{238}\text{UX}^-$ and $^{232}\text{ThX}^-$ should both be present if the stable X^- exists. These nuclei would form atoms with the chemistry of protactinium and actinium, respectively. Neither of these elements occurs in nature since their longest lived isotope is ^{231}Pa , with a half-life of 3.3×10^4 yrs. In fact, the half-lives of $^{238}\text{UX}^-$ and $^{232}\text{ThX}^-$ would far exceed 10^{10} yrs. because the presence of the X^- gives added stability to the parent nucleus relative to the daughter in the alpha decay. Using Eq. (2), we find that the energy of the alpha emitted in the decay is 0.4 MeV less than if the X^- is not present. Using the Geiger-Nuttall law and the known half-lives of various uranium and thorium isotopes, we estimate that the half-lives of the superheavy isotopes will be about 10^3 times longer.¹² This would be enough to make the half-lives of $^{235}\text{UX}^-$ and $^{236}\text{UX}^-$ comparable to the age of the universe, as well. Indeed, even $^{247}\text{CmX}^-$ and $^{244}\text{PuX}^-$ would have half-lives of

the order of 10^{10} yrs. See Fig. 2. These would have the chemistry of americium and neptunium, respectively. In summary, a search for naturally occurring actinium, protactinium, americium and neptunium would be an important means of searching for superheavy, charged, stable particles.

We can identify other chemical substances where the concentration of X^- 's is likely to be enhanced. Let us indicate by $N_X^0(Z,A)$ the fraction of the nuclei with X^- 's in them which have Z protons and $A - Z$ neutrons. By the superscript zero we mean that this is an initial distribution, let us say representing the distribution at the time of the formation of the solar system. We indicate the fraction of ordinary nuclei at the same time with Z protons and $A - Z$ neutrons by $N^0(Z,A)$. We define the function $a(Z,A)$ by

$$N_X^0(Z,A) = a(Z,A) \cdot N^0(Z,A) \quad (7)$$

A reasonable hypothesis is that the X^- 's are distributed approximately as the nucleons so that $a \propto A$ and

$$a(Z,A) = \frac{A}{\sum_{Z,A} N^0(Z,A)} = \frac{A}{\bar{A}} \quad (8)$$

Certainly this is a very crude approximation, but it will suffice except for some special cases to be considered below.

Let us denote by $P(Z)$ the probability that a nucleus with a charge of Z actually has $Z + 1$ protons and one X^- (we shall suppress dependences on A). This probability, in the initial distribution, is

$$P(Z) = R a(Z + 1, A) N^0(Z + 1)/N^0(Z) \quad (9)$$

where R is the ratio of the number of X^- 's to the total number of nuclei.

This probability, $P(Z)$, may not represent the present terrestrial probability because of processes which occurred during or since the formation of the earth. However, let us begin by ignoring this correction. Then using our simple ansatz, Eq. (7), we can select good candidates for the search for superheavy particles by listing for various elements the quantity $A N^0(Z+1)/N^0(Z)$. Some selected values are shown in Table II. The odd-Z nuclei are seen to have the largest values. This simply reflects the greater nuclear stability of the even-Z nuclei which are consequently produced in greater numbers in nucleosynthesis. An exception to this rule is Be, whose even-even nucleus ^8Be is unstable against decay in two alpha particles.

From Table II, we see there are several other elements which are particularly attractive candidates for a search for superheavy matter: B, F, Mn, Be, Sc, and V. In these cases, neutron activation might be used to identify a component whose nuclear structure was not those of the element being studied.

An exception to the approximation given by Eq. (8) is $^8\text{BeX}^-$ which we would expect to be made rather easily. Limits on this supernucleus should be obtained by studying lithium, perhaps by the search for isotope-shifted visible lines.

Another exception to Eq. (8) would be Pb. Since the presence of an X^- pushes stability towards higher Z, it is likely that the X^- 's would be found preferentially in high Z nuclei. In addition to favoring the nearly stable actinides, this would lead to a concentration in Pb. The result would be PbX^- , with chemical properties identical with those of Tl.

The probabilities described by Eq. (9) do not take into account any effects which might have occurred during or since the formation of the Earth. In particular, most of the volatile elements originally present were lost from the initial atmosphere. Thus neon is quite rare (18 ppm by volume) in the atmosphere although it is a primary component of nucleosynthesis (see Table II). It is possible that NaX^- , which would be chemically a noble gas, might have survived the process which resulted in the loss of volatile elements. A list of suggested searches is given in Table III.

Present limits on the concentrations of superheavy nuclei of the sort discussed here are rather weak. The agreement between masses measured chemically and using mass spectrometers is about one part in $10^{5.13}$. Thus the concentration of 100 TeV X^- 's in matter with $A \sim 100$ must be less than about one part in 10^8 . Dover, Gaisser, and Steigman suggest that one part in 10^{10} is a likely scale for the concentration of superheavy particles. Of course it is desirable to push far beyond this scale if possible. Searches in hydrogen¹⁴ might be sensitive to a level of a part in $10^{18} - 10^{20}$. Searches using neutron activation in heavier elements would require finding $\sim 10^{11}$ abnormal nuclei.¹⁵ Thus chemical separation of 1 kg of rhenium ($6 \cdot 10^{26}$ nucleons) could produce a limit of about a part in $10^{15} - 10^{16}$.

We have outlined a number of chemical searches which might lead to the discovery of new, very heavy, stable, charged particles. Any of them has the potential to find these new objects, but which search is the most promising we cannot say. Not only does that depend on the applicability of specialized techniques for isolating

and identifying the new substances, but it depends as well on geophysical considerations which determine where the new substances might actually be found terrestrially. With such large atomic weights, these new chemicals might have undergone extensive fractionation so they may not be distributed in the same manner as their lighter analogs.

We would like to thank L.W. Alvarez, F. Asaro, R. Hagstrom, R. Muller, and R. Wagonner for their assistance and advice. One of us (R.N.C.) was supported in part by an A.P. Sloan Fellowship and expresses his thanks to the Aspen Center for Physics where a portion of this work was completed. The other acknowledges the kind hospitality of the Lawrence Berkeley Laboratory where this work was begun. He also thanks Prof. C.K. Jorgensen for several enlightening conversations and communications.

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13. See, for example, R.J. Holt, et al, Phys. Rev. Lett. 36, 183 (1976), where limits for Bi are discussed. Their experiment set very stringent limits on the occurrence of abnormal nuclei of a particular kind, but was insensitive to the sort of matter we are discussing.
14. Ingenious techniques for such searches have already been proposed by R. Muller and by R. Hagstrom. We thank Drs. Muller and Hagstrom for discussing with us the possibilities of searching for superheavy hydrogen.
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TABLE I

Electrostatic binding energies of X^- to various nuclides. The X^- is taken to be much more massive than the nucleus. See Eqs. 4 - 6.

Nucleus	E_b (MeV)
1_H	0.025
2_H	0.050
3_H	0.075
$^3_{He}$	0.270
$^4_{He}$	0.311
$^5_{Li}$	0.842
$^5_{He}$	0.431
$^6_{Li}$	0.914
$^7_{Li}$	0.952
$^7_{Be}$	1.49
$^8_{Be}$	1.55

TABLE II

Possible sites for enhanced X^- concentration. The final column is a rough guide to the enhancement. The concentrations, N^0 , are relative to $Si = 10^6$ and are taken from Ref. 16. The abundances are not for the present terrestrial composition, but for the solar system.

Z	$N^0(Z)$	$N^0(Z+1)$	A	$A N^0(Z+1)/N^0(Z)$
3 (Li)	5×10^1	8×10^{-1}	7	1×10^{-1}
4 (Be)	8×10^{-1}	4×10^2	9	3×10^3
5 (B)	4×10^2	1×10^7	11	3×10^5
9 (F)	2×10^3	3×10^6	19	3×10^4
10 (Ne)	3×10^6	6×10^4	20	3×10^{-1}
11 (Na)	6×10^4	1×10^6	23	4×10^2
13 (Al)	8×10^4	1×10^6	27	3×10^2
15 (P)	1×10^4	5×10^5	31	2×10^3
17 (Cl)	6×10^3	1×10^5	35	7×10^2
18 (Ar)	1×10^5	4×10^3	40	1×10^0
19 (K)	4×10^3	7×10^4	39	7×10^2
21 (Sc)	4×10^1	3×10^3	45	4×10^3
23 (V)	3×10^2	1×10^4	51	3×10^3
25 (Mn)	9×10^3	8×10^5	55	5×10^3
27 (Co)	2×10^3	5×10^4	59	1×10^3
33 (As)	7×10^0	7×10^1	75	7×10^2
36 (Kr)	5×10^1	6×10^0	84	1×10^1
54 (Xe)	5×10^0	4×10^{-1}	131	1×10^1
81 (Th)	2×10^{-1}	4×10^0	204	4×10^3

TABLE III

Suggested chemical searches for superheavy matter.

Superheavy Material	Chemical Behavior	Motivation for Search
1. $X^+e^-, ({}^4\text{He}X^-)e^-$	H	Site of all X^+ ; large ${}^4\text{He}$ abundance.
2. $\text{Ru}X^-$	Tc	No stable Tc isotope.
3. $\text{Sm}X^-$	Pm	No stable Pm isotope.
4. ${}^{232}\text{Th}X^-$	Ac	No stable Ac isotope.
5. ${}^{235}, {}^{236}, {}^{238}\text{U}X^-$	Pa	No stable Pa isotope.
6. ${}^{244}\text{Pu}X^-$	Np	No stable Np isotope.
7. ${}^{247}\text{Cm}X^-$	Am	No stable Am isotope.
8. $\text{C}X^-$	B	B much scarcer than C.
9. $\text{Ne}X^-$	F	F much scarcer than Ne in cosmic abundance.
10. $\text{Fe}X^-$	Mn	Mn much scarcer than Fe.
11. $\text{B}X^-$	Be	Be much scarcer than B.
12. $\text{Ti}X^-$	Sc	Sc much scarcer than Ti.
13. $\text{Pb}X^-$	Th	Th much scarcer than Pb.
14. $\text{Cr}X^-$	V	V much scarcer than Cr.
15. $\text{Be}X^-$	Li	$\text{Be}X^-$ copiously produced in ${}^4\text{He}X^- + {}^4\text{He} \rightarrow {}^8\text{Be}X^-$.
16. $\text{Na}X^-$	Ne	Initial Ne lost from atmosphere.

FIGURE CAPTIONS

1. The dimensionless function $\lambda(a)$, which gives the binding energy for the hybrid Coulomb - harmonic oscillator problem. The dot-dash curve is the solution to the pure Coulombic problem. The dotted curve solution to the harmonic oscillator problem. The solid curve is the solution to the hybrid problem. See Eqs. 1 - 6.
2. The half-life, $t_{1/2}$ (in yrs.), of Pu isotopes versus Δ , the energy released the α -decay. The half-life of ^{244}Pu is estimated by extrapolating from measured life-times using the Geiger-Nuttall Law and the calculated reduction of 0.4 MeV in Δ .

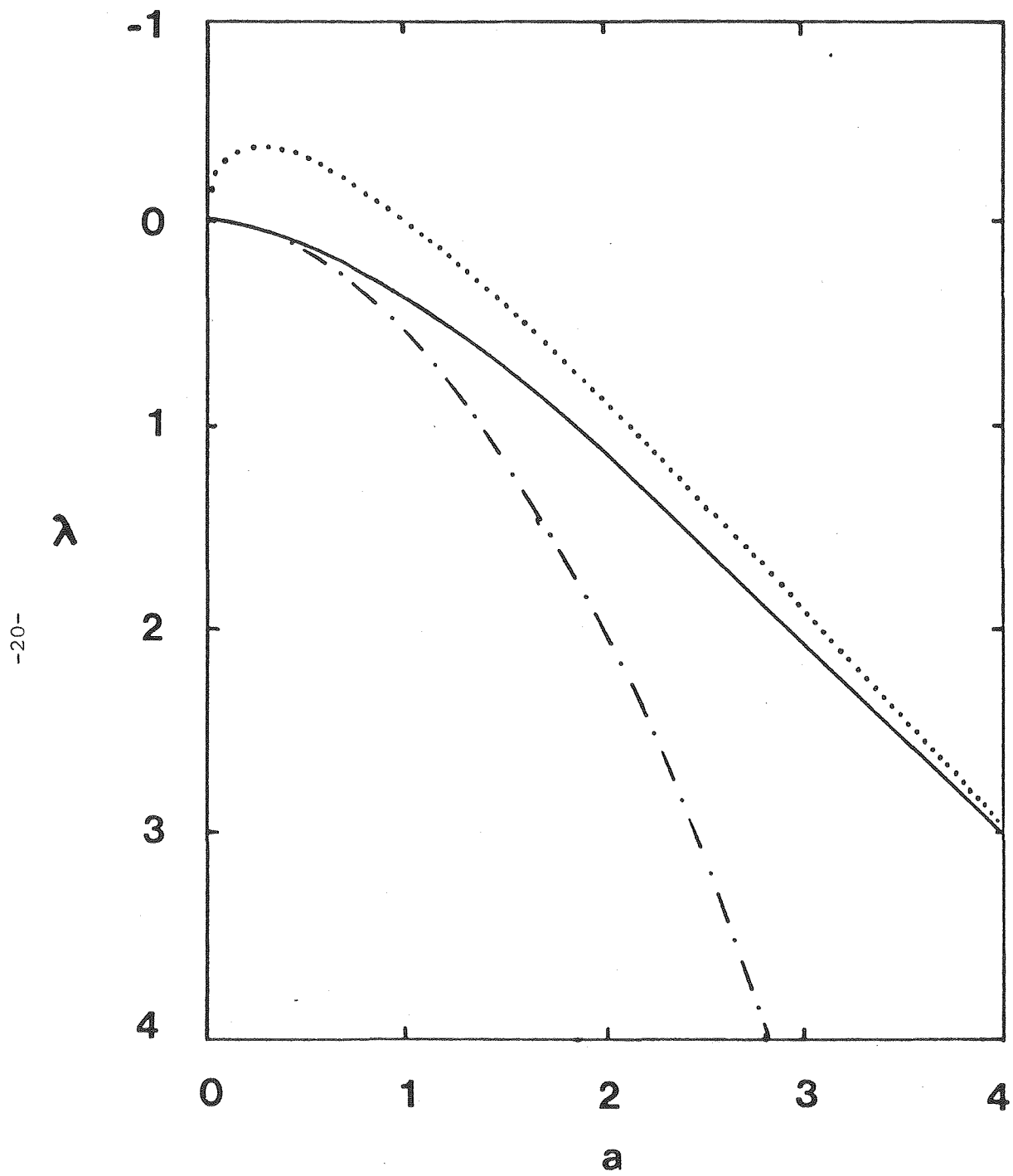


Fig. 1

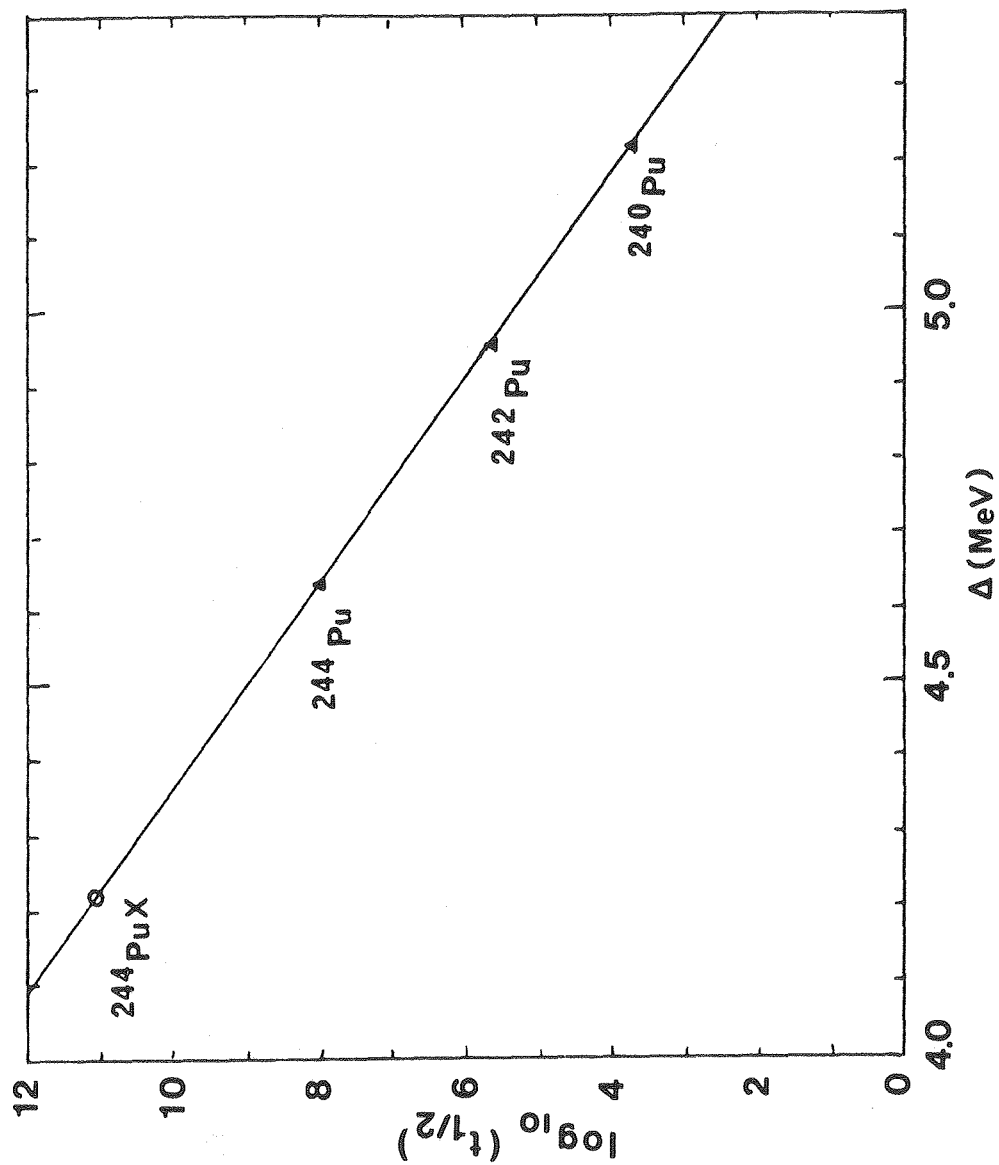


Fig. 2