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STABILITY OF SUPERHEAVY NUCLEI AND THEIR POSSIBLE OCCURRENCE IN NATURE *

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Alpha and spontaneous fission half-lives were estimated theoretically in the region of beta-stable nuclei with $Z \approx 114$, $N \approx 184$. The possible existence of an island of stability is strongly supported. A preliminary search for surviving superheavy elements in nature gave negative results.

The hope for the possibility of the existence of an island of stability connected with nuclear magic numbers somewhat beyond the presently accessible regions of stable nuclei was recently discussed by Myers and Swiatecki [1]. Shell model calculations [2] indicated that such an island may be associated with the doubly closed shell nucleus $^{298}114$. Ground state masses and potential energies of nuclei as a function of deformation have recently been calculated [3,4] for the region in the vicinity of the nucleus $^{298}114$. These calculations are based on a simple deformable shell model potential and are normalized to reproduce the average trends of the liquid drop model. From this we have been able to estimate the alpha and spontaneous fission half-lives and to test for beta stability of these superheavy nuclei. Within the uncertainty of the calculations as discussed below we can make certain statements about the possibility of finding such superheavy nuclei in earthly matter and in cosmic radiation.

The calculation makes use of a generalized harmonic oscillator potential with quadrupole (P_2) and hexadecapole (P_4) deformations. In addition to a spin-orbit force, one also includes a coupling proportional to $l^2 - \langle l^2 \rangle_N$, where the last term represents the average over a given N-shell. There are two adjustable parameters, which are fitted to reproduce optimally the observed level order in the actinide ($A \approx 242$) and the rare earth ($A \approx 165$) regions. We have assumed these parameters to have a linear A-dependence, and extrapolated them to the superheavy region ($A \approx 300$). Pairing energy contributions are calculated on the basis of the single particle levels from the shell model calculations. The pairing strength is assumed to be isospin dependent and proportional to the surface area of the nucleus.

The usually employed volume conservation condition is complemented by a generalized Strutinski method [3,5] of normalization. The latter ensures that *on the average* the behavior of deformation energies at large distortions is that of a charged liquid drop. This is effected by subtracting from the total single particle energy the corresponding value for the case of smeared energy levels according to a well-defined prescription [3,5]. What remains is a fluctuating shell structure contribution, which to a high degree of accuracy is independent of the prescription for smearing the levels. The subtracted term is then replaced by the corresponding liquid drop energy terms (volume + symmetry + surface + surface symmetry + Coulomb). The liquid drop parameters are taken, without readjustment, from Myers and Swiatecki [1], who determine their parameters using a simulated shell structure correction term in their mass formula.

In this way, the potential energy surface in the space of P_2 and P_4 deformations can be studied for each nucleus. The ground state mass corresponds to the lowest minimum in this surface. Empirical masses in the region $150 < A < 250$ are reproduced within approximately 1 MeV (fig. 1). Based on these masses, alpha decay half-lives and beta stabilities are determined.

The spontaneous fission half-lives may also be found from the potential energy surface if one knows B, the inertial parameter associated with the barrier penetration. This parameter has been

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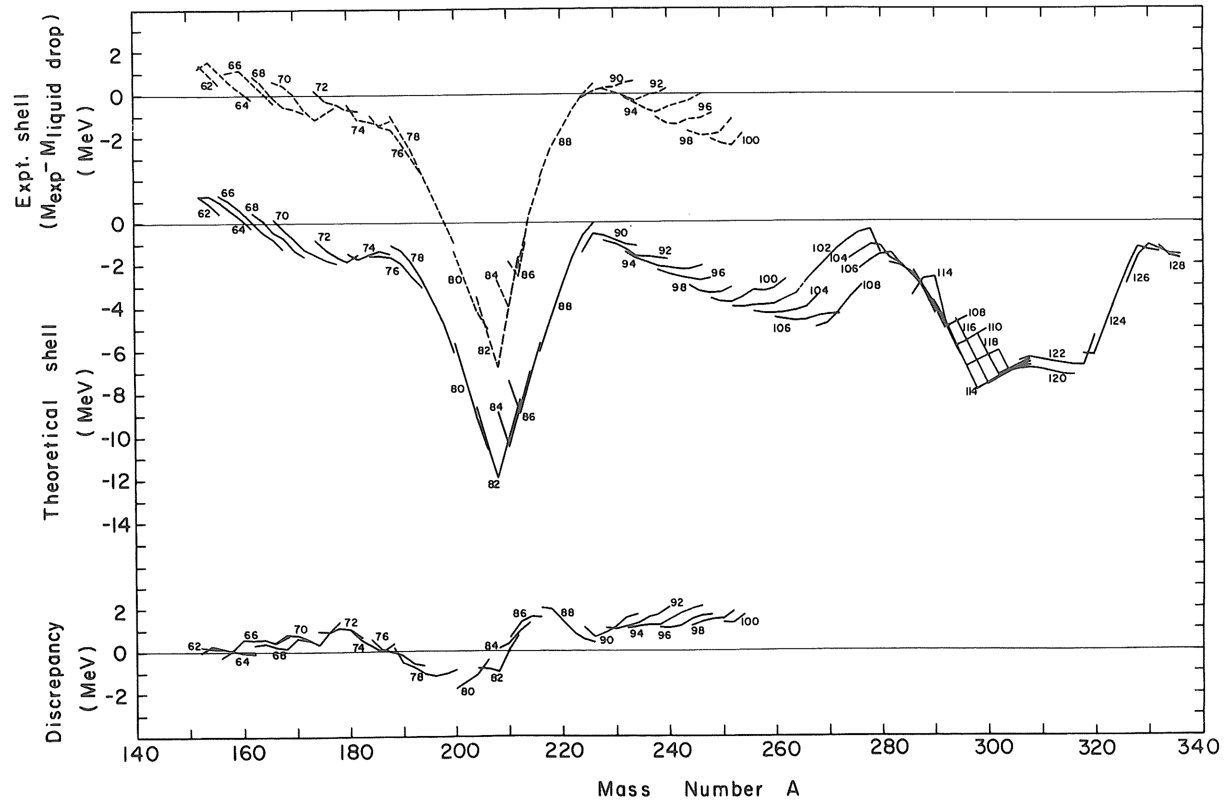

Fig. 1. Experimental and theoretical mass values for $150 < A < 310$ plotted relative to the spherical liquid drop values as of ref. 1.

estimated [6] in two alternate ways. First, from an analysis of experimental fission half-lives of actinides ($Z = 90 - 100$) with the ground state fitted to the empirical mass, Moretto and Swiatecki [7] have determined that the values $BA^{-5/3}$ lie within a small range independent of A. In view of the difference in saddle point shapes between actinides and superheavy elements it may be a somewhat unsatisfactory assumption to use the same $BA^{-5/3}$ for the superheavy nuclei. Alternatively B has been determined at the saddle point from "microscopic" theory by Sobiczewski, Szymanski and Wycech [6,8]. The B values so obtained for the superheavy nuclei are constant within 30% and lie within 30% of the first estimate. We have used an average of the B values determined in these two alternative ways in order to estimate the fission half-lives.

The results are summarized in the half-life contour diagram of fig. 2. The great uncertainty associated with the numbers obtained must be emphasized. First of all there is the uncertainty of the extrapolation of the shell model potential and the liquid drop parameters to an unknown mass region. Furthermore, a deviation of 30% in the estimate of the inertia parameter B corre-

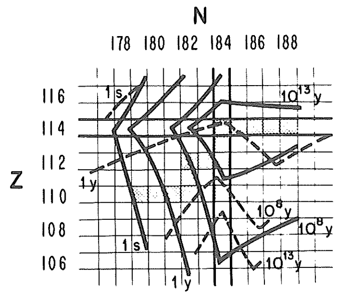

Fig. 2. Contours of theoretical half-lives in the vicinity of $Z = 114$ and $N = 184$. The thick dark lines are contours of spontaneous fission half-lives. The broken lines are contours of alpha half-lives. Beta stable nuclei are shaded.

sponds roughly to a factor of 10^6 in the spontaneous fission half-lives, while a 1 MeV deviation in alpha energy corresponds to a factor 10^6 difference in alpha half-lives. An underestimate of a given nuclear mass due to a local shell effect leads normally to an overestimate of the fission half-life. On the other hand the alpha half-life depends on the relative masses of two nuclei two units of N and Z from each other. Hence so long as an under- or over-estimation of masses affects these two nuclei to a similar degree, the error in alpha energy is comparatively small. For the actinide region (where we do not have the uncertainty due to extrapolation of parameters) our alpha energies are within 5% of the experimental values corresponding to half-lives agreeing within a factor of ten, but our fission half-lives for some isotopes can be wrong by a factor as large as 10^6 either way.

All these uncertainties may move the contours of half-lives in fig. 2, but the general pattern should remain the same so long as $Z = 114$, $N = 184$ is a well-developed magic nucleus. Thus, the main use of the figure is as a guide in the search for relatively long-lived nuclei in this region.

Some general features of fig. 2 may be pointed out. The longest fission half-lives center rather symmetrically around ($Z = 114$, $N = 184$). It must be emphasized here that any stability against spontaneous fission in this region is due to the extra binding resulting from the shell effect so that as one goes away from $^{298}114$, the fission half-lives decrease rapidly. Without the shell effect, the alpha half-lives depend on the inclination of the $\Delta N = \Delta Z$ line (which is the direction of alpha decay) with respect to the direction of the beta stability valley. The shell effect essentially increases the alpha half-lives for nuclei with $Z < 114$ and $N < 184$ and decreases those for nuclei $Z > 114$ and $N > 184$. It also causes the kink shown in fig. 2.

Applying a "survival-of-the-fittest" test with respect to fission, alpha decay, and beta decay in the above region and taking the calculated numbers at their face value, one ends up with *one* possible candidate for survival in earthly matter, namely $^{294}110$, which should be chemically closely related to platinum. However, the uncertainty of our numbers as discussed above may indicate that, instead, a nucleus close-by may have a better chance of survival. However, if the total half-life falls below $\sim 2 \times 10^8$ years, its detection in earthly matter is beyond the capabilities of our present techniques *.

A question may be asked whether such a long-lived superheavy element may be produced in nature in the first place. The indication ** is that such a superheavy element could well be formed by the r-process [9] in which a nucleus absorbs a large number of neutrons very rapidly and then undergoes successive beta decays to much heavier relatively stable nuclei ***. Most very neutron rich isotopes seem to be sufficiently fission stable for this process, which also requires a condition of huge neutron flux and very high temperatures. This condition may have prevailed at some point in the history of the universe and may also exist in some massive stars and quasi-stellar radio objects at this present time. This at one raises the possibility of detecting superheavy nuclei in the primary cosmic radiation. According to the most optimistic estimates, the nuclei of interest in the primary cosmic radiation may have been produced 10^5 years ago, while elements in the solar system have an age of $\sim 5 \times \times 10^9$ years. If we take fig. 2 at its face value, we see that in the study of the primary cosmic rays, one might be able to find a few more nuclei which live longer than 10^5 years.

So far, the experimental search for an indication of an element with Z near to 114 has followed three essentially independent approaches.

A. The first [10,11] involved the attempt to observe the spontaneous fission of short-lived nuclei produced by the reaction:

$$^{248}\mathrm{Cm} + {}^{40}\mathrm{Ar} \rightarrow {}^{284}114 + 4\mathrm{n}\ .$$

Negative results were obtained corresponding to an upper limit of $\sim 10^{-32}$ cm^2 for the cross section. This is not surprising because the experimental techniques used can detect half-lives only down to $\sim 10^{-8}$ sec and the predicted half-life is less than 10^{-15} sec. The obvious difficulty with the heavy ion approach at present is that nuclei so produced are much too neutron deficient. However, the availability in the future of projectiles of larger neutron excess such as ^{48}Ca, ^{64}Ni and ^{50}Ti might improve the picture.

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* We would like to thank Dr. Luciano Moretto for drawing our attention to this point.

** We are grateful to Dr. P. A. Seeger for a helpful discussion of the r-process.

*** Our estimate of masses along the prospective r-process path is, however, sensitive to the value assumed for the coefficient of the surface symmetry energy. Conceivably the value of this coefficient, after readjustment of all the liquid drop parameters, might be such as to make the generation of superheavy elements impossible. The isotopic trends of actinide fission half-lives, which we fail to reproduce adequately [3], may be indicative of this.

B. The second approach [12] took the view that $^{294}110$ might have been produced and still be present in nature. This element is expected to be chemically similar to platinum. Therefore natural platinum ores were used in the following experiments. The search followed two lines. First, on the assumption that the half-life was very long ($>10^{14}$ years), analyses were made using X-ray fluorescence, mass spectrometry, and finally activation with heavy ions as well as protons at various energies. The results indicated that the upper limit on the presence of $^{294}110$ in platinum is <1 part in 10^9. The second mode of investigation assumed the half-life to be less than 10^{14} years. Low background counting of natural platinum ore for neutrons, gamma rays and spontaneous fissions detected no activity above background. This suggests on the basis of the present sensitivity that the longest half-lives are less than 2×10^8 years, or that the element is present in undetectable amounts.

Even if the longest half-life in this region of elements is less than 2×10^8 years, it may be possible to obtain information concerning superheavy nuclei existing at some time in the past by searching for neutron rich products of spontaneous fission in meteorites or in natural ores of platinum and its neighboring elements.

C. Extensive nuclear emulsion studies of very heavy particles in the cosmic rays at high altitudes have been made by P. H. Fowler and collaborators. They have reported evidence for uranium and its neighbors [13]. Continuation of their work indicates [14] one event with $Z = 103 \pm 4$. They have accumulated much more data recently which are under analysis.

We have profited very much from discussions with Dr. W. J. Swiatecki, who provided the original stimulus for the undertaking of these investigations. Most useful advice by Drs. A. Bohr and B. R. Mottelson is gratefully acknowledged. We also wish to thank Dr. L. G. Moretto for helpful discussions.

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