Superheavy Elements in Nature ?

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This note comments on recent estimates of alpha and spontaneous fission half-lives near the conjectured magic proton and neutron numbers $Z=114$ and $N=184$, emphasizing

1) a possible non-existence of a significant $N=184$ shell,
2) the expected effects of odd proton and neutron numbers on the stability in this region, and
3) possible relations to unexplained natural radionuclides reported in the literature.

In contrast to other estimates, our considerations yield isotopes of the elements 111 and 113 as the candidates for superheavy nuclei with the relatively longest halfives.

Until now, the theory of superheavy element stabilities is not more than a crude phenomenology. A recently performed careful study[1,2] of alpha and spontaneous fission half-lives could nonetheless give some important hints. Such studies, see also Refs.[3–5], have led to speculations that isotopes of the elements 110 to 114, eka-platinum to eka-lead[3,4], may exist in nature. So far, attempts to detect such elements in platinum[1,6,7] and lead samples[7,8] gave negative[1,6,7] or inconclusive results[8]. In view of the qualitative uncertainties in both the method[9] and the extrapolations of Refs.[1,2], we will confine ourselves to the purely qualitative use of their results.

Inspection of the second figure in Ref.[1] reveals a strong sensitivity of their half-life estimates to the assumption of a significant $N=184$ shell: Our Fig. 1 shows in heavy lines their contours for regions of "good chances" for survival in earthly matter, i.e., for alpha and spontaneous fission half-lives of 10^8 years. It should be noted that only even-even nuclei were considered in Refs.[1,2] with regard to spontaneous fission half-lives. Decreasing the influence of the $N=184$ shell to a point where it produces in the contours a kink half as pronounced as in Refs.[1,2] — as shown by thin lines in Fig. 1 here — is what we call in the following "50% 184-influence". Even when the alpha contour is unchanged, such a 50% decrease of the 184-influence would no longer leave the overlap between the contours of sufficiently long alpha and spontaneous fission half-lives. As was emphasized, for instance, in Refs.[9] and [10], the extrapolations of Ref.[1] have an additional uncertainty due to the lack of self-consistency. It was only the $Z=114$ shell that has been confirmed by *self-consistent* calculations[10] whereas no significant $N=184$ shell resulted from that investigation. Therefore, the extrapolations of the latter work give less support to the possibility of finding superheavy elements in nature.

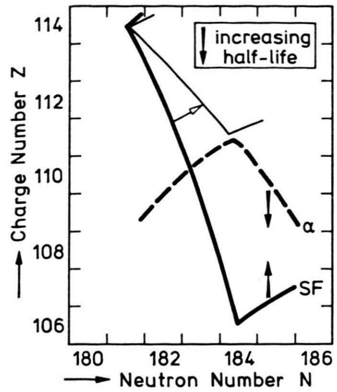

Fig. 1. Estimated half-life contours in the $N-Z$-plane for even-even isotopes of superheavy elements. The heavy lines are the 10^8 year contours for spontaneous fission (solid) and alpha half-life (dashed) estimated in Refs.[1,2]. Thin lines show an example of how spontaneous fission contours will shift when the $N=184$ shell is less pronounced.

Nonetheless, let us speculate about mechanisms that might contribute to increased stability in this region. For instance, the self-consistent results[10] suggest a somewhat stronger $Z=114$ influence. In addition, a mechanism widening the long half-life regions perpendicular to this (Z-)direction should be the odd-odd effect. In the transuranium region, nuclei having odd proton and neutron numbers are observed to have systematically longer half-lives than those with even-even composition. Whereas in alpha decay the increase in half-life amounts, on the average, only to a factor of about 10^2, cf.[11], inspection of spontaneous fission half-lives[3,4] indicates that the values for odd-even and even-odd nuclei increase by a factor of 10^4 to 10^5 compared to their even-even neighbours. For odd-odd nuclei, the hindrance may even be stronger, as the half-life

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of $^{254}_{99}Es$ shows which is the only odd-odd nucleus of known spontaneous fission half-life. Such hindrance effects are qualitatively understood in the framework of the adiabatic fission theory[12–14].

Odd effects may also play a role in the formation of superheavy elements by the *r*-process in which a nucleus absorbs rapidly a large number of neutrons and then undergoes successive beta decays to nuclei of much higher charge number. From the yields of heavy transuranium nuclei formed by multiple neutron capture in thermonuclear explosions, it has been concluded that such capture chains proceed more likely in odd-*Z* elements[15].

To estimate the consequences of odd effects on stability, let us start again with the results of Refs.[1,2] and shift their contours as shown in the lower part of Fig. 2. The weakening of the $N = 184$ shell is done graphically as indicated in

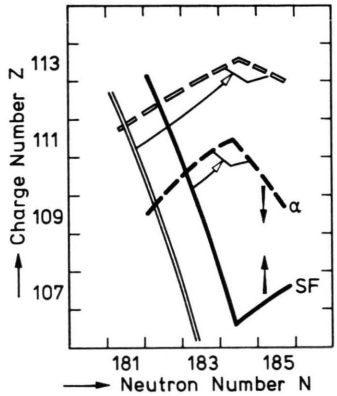

Fig. 2. Contours for odd-odd nuclei (heavy lines) and estimate of the odd-odd effect on spontaneous fission and alpha half-lives (light lines) that is expected to widen the region of relative stability.

Fig. 1. As we turn down the $N = 184$ influence, an odd-odd isotope $^{294}111$ of the element eka-gold appears to be the candidate which survives longest since at that point of the $N-Z$-plane the overlap between alpha and spontaneous fission half-lives finally vanishes. This graphical method also indicates that the survival chance of $^{294}111$ improves significantly if the 114-influence is increased by about 30%. In such a case the strength of the $N = 184$ shell of Refs.[1,2] could be reduced by more than 70%.

Now we switch on an odd-odd effect as follows (Fig. 2): Place the contours for the region of "good survival chances" for both spontaneous fission and alpha decay midway between the 10^0 and 10^8 year contours of Refs.[1,2], corresponding to a hindrance factor of about 10^4 for both decay processes. This is well within the limits of uncertainty given as a factor of 10^6 in Refs.[1,2]. With the *full* strength of the $N = 184$ shell, the region of survival chances would now extend to the elements 107 and 105, eka-iridium and eka-rhenium, whereas element 113, eka-thallium, could stand about 60% *reduction* of the $N = 184$-influence. As far as spontaneous fission is concerned, our shift by a factor of 10^4 for the half-lives of odd-odd nuclei seems quite moderate in view of experimental data[3,4] and theoretical estimates[12]. However, the extension of the alpha stability region has to be quite significant; also we have neglected that the kink in the alpha contour will decrease when the $N = 184$-shell effect is reduced. Nonetheless our considerations would suggest, in contrast to Refs.[1,2], the candidates $^{294}111$ and $^{298}113$ for longest half-lives — the latter having less stability because of alpha decay. However, these odd-odd nuclei might be very beta-unstable* and, hence, their beta-stable[1,2] neighbours $^{293}111$ and $^{297}113$ could be the more favourable candidates for occurence in earthly matter.

From the geochemical point of view, the elements eka-rhenium to eka-lead should most likely occur in sulfide minerals. In addition, the eka-platinum metals and eka-gold may be found in placer deposits of their related elements in resistate sediments whereas eka-thallium may also occur in alkali silicates, as thallium does. It is interesting to note that unexplained alpha emitters were observed in such materials in older[16–20] and more recent studies[21–26]. For example, alpha emitters with 4.5 to 5.0 MeV energy were chemically isolated from sulfides[20,21], osmiridium[22] and other minerals[23,24], and were also observed in mica in form of pleochroic haloes[17–19,25]. This energy is close to that expected[2] for alpha decay of nuclei with $Z = 106$ to 109 and $N \approx 184$. Other haloes found in mica[17–19] fall into the region of alpha energies expected[2] for isotopes of the elements 110 to 114, i.e., 5.2 to 7.8 MeV energy.

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