ON NUCLEAR MOLECULES BUILT UP FROM ^{132}Sn COMPONENTS

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Dedicated to Adam Sobiczewski in honour of his 70th birthday

The possible existence of nuclear quasi-molecules built up from ^{132}Sn components is investigated. The crucial question is whether the extra stability of the doubly magic ^{132}Sn nuclei makes them sufficiently rigid to be able to withstand the strains imposed by their mutual interactions. It is argued that if the simplest quasi-molecular dumbbell configuration were found to be (meta-)stable, then triangular and even tetrahedral structures might have comparable barriers against disintegration and comparable spontaneous fission lifetimes. These are estimated using simplifying assumptions. As regards the dumbbell's stability, one may relate this to the existence of a potential energy pocket in the deformation energy landscape of a fissioning ^{264}Fm nucleus, and to the presence of 'bimodal' fission in heavy Fm isotopes. Further experimental and theoretical studies of such systems may be relevant for answering the question concerning nuclear quasi-molecules.

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1. Introduction

It is a pleasure to dedicate this note to Adam Sobiczewski, whose work I admire and whose friendship I cherish. Adam is widely recognized as having provided us with a reliable aid in the exploration of the tantalizing territory of very heavy elements. Together with his co-workers he was in the forefront of charting the properties of superheavy elements around proton and neutron numbers Z=114, N=184. Originally this was done with quite limited computer technology, but the calculations have stood the test of time to a remarkable degree. This is surely due to the uncompromising integrity and attention to detail that characterize Adam's work.

In my contribution to this volume I would like to take a leap into the unknown by speculating on the possible existence of quasi-molecular configurations consisting of a number of ^{132}Sn nuclei in contact. This speculation is motivated by the circumstance that a doubly magic nucleus like ^{132}Sn is expected to exhibit solid-like properties [1–3], and that solid components can be glued together in quasi-molecular clusters by nuclear proximity interactions [4,5]. (In fact, the earliest application of the proximity technique was described in a 1934 paper on the coagulation of aerosols [6].)

2. Qualitative considerations

The simplest such quasi-molecule would correspond to two ^{132}Sn nuclei in contact. This configuration could also be regarded as a very deformed ^{264}Fm nucleus on the way to fission. (More specifically, to fission of the compact, high kinetic energy type [7].) Whether such a quasi-molecular pocket in the deformation energy map of a fissioning ^{264}Fm nucleus exists, and what its lifetime would be has, as far as I know, not been answered with any degree of certainty. But if a quasi-molecular dumbbell were found to be (meta-) stable, then one can plausibly argue that a triangular quasi-molecule consisting of three ^{132}Sn nuclei in contact would also be (meta-)stable. This is because for the triangle the Coulomb-plus-proximity interaction energy, in its dependence on separation and necking, is approximately just a multiple (three) of the interaction energy for the dumbbell. (I am assuming that the energies of the nuclear proximity bonds are approximately additive.) The argument continues to hold for a four-component tetrahedral quasi-molecule, but fails for more than four components. (In that case the number of proximity bonds becomes less than the number of pairwise electrostatic repulsions.) The cheapest way to disrupt a triangular molecule is to break a single bond in a 'scissor' mode, *i.e.*, by increasing the length of one of its sides, leaving the other two unchanged. It then follows that the spontaneous fission lifetime of the triangle — a system with $Z = 150$, $A = 396$ — could be approximately the same as of the dumbbell. [The logarithm of the lifetime would actually be increased by approximately $\sqrt{(10/9)}$. This factor takes account of the effective mass associated with the scissor mode, which is slightly different than the effective (reduced) mass for the separating dumbbell.] Note also that after the first bond is broken, and the triangle stretches out into a linear configuration, the resulting linear molecule, even if metastable, is unlikely to present a further barrier to the spontaneous fission of the original triangle.

In addition to the spontaneous lifetime for fission of the hypothetical $Z = 150$ nucleus one needs to estimate the lifetimes for alpha and beta decay (or electron capture). These lifetimes will tend to be lengthened by

the magicity of the ^{132}Sn components, and by the fact that the electrostatic energy of the triangle is appreciably less than that of the equivalent sphere (see Section 4). A further factor that might reduce instabilities is the screening of the nuclear electrostatic energy by the atomic electrons. For the very high atomic number in question a fraction of the atomic electrons would become relativistic, would orbit the nucleus at relatively small distances and would thus tend to neutralize to some extent the nuclear electrostatic energy [8]. One should also investigate the effect on the (pseudo-chemical) binding properties of the molecules of adding or subtracting nucleons from the ^{132}Sn components. But the primary question to be answered concerns the existence and lifetime against spontaneous fission of a hypothetical quasi-molecular dumbbell.

3. Fission barriers and lifetimes

As I mentioned, I am not aware of reliable calculations of the dumbbell configuration's stability, so I shall attempt here some kind of rough estimate. In order to obtain a first orientation, plot the Coulomb energy of the two approaching ^{132}Sn nuclei, reduced by the nuclear proximity attraction calculated for frozen density distributions of the fragments [4]. (Unfreezing the neck degree of freedom will be considered presently.) The resulting energy plot, E(approach), is found to have a maximum of 262.4 MeV at an overall elongation L of the system of 24.1 fm, and a minimum of 251.6 MeV at $L = 22.3$ fm [9]. Taken with respect to the theoretical ground state energy of ^{264}Fm, equal to 259.9 MeV [10,11], these numbers become 2.5 MeV and -8.3 MeV, respectively. The difference gives a barrier against disintegration of 10.8 MeV. One also finds that if this barrier were to be penetrated by spontaneous fission, the "exit point" of the penetrability integral would be at an overall elongation of 25.4 fm, resulting in a total width w of the barrier of 3.1 fm. The penetrability exponent I of a cubic barrier of height B (in MeV) and width w (in fm) is given by

$$I = 2.4626\sqrt{10/9}\; w\sqrt{B}\,, \tag{1}$$

where the factor $10/9$ is appropriate in the case of the triangular molecule, and 1 in the case of the dumbbell. I have neglected zero-point energy effects. This gives a lifetime of the order of $10^{-21+11.48}$s, or about 0.31 ns.

I believe this is likely to be an overestimate for the following reason. The experimental masses of the fragments at infinity include a total shell effect correction of $2(-11.75\ \text{MeV}) = -23.5$ MeV [10,11], and this correction will be progressively attenuated as the fragments approach contact. The same is expected of the change in the Congruence energy of [12], which is

$2(-3.61$ MeV) for the separate fragments, and -3.61 MeV for the compound nucleus. In [13,14] it was estimated that the shell correction at contact is attenuated by 10%, to 90% of its initial value. Assuming that the attenuation of the Congruence energy change is similar, we should increase the contact energy (which is close to the energy at the minimum) by some $0.1(23.5 + 3.61) = 0.1(27.11) = 2.71$ MeV, but by less at the position of the maximum. This will lower the barrier against disintegration. To estimate by how much, note that the attenuation is expected to be related to the degree of communication between the two fragments. A measure of this communication is the Proximity interaction itself, so I shall take the attenuation of the shell-plus-congruence energy to be proportional to the proximity potential. With this prescription the new interaction energy curve exhibits a minimum of -4.95 MeV at 22.35 fm, a maximum of 3.45 MeV at 24.05 fm and an exit point at 25.15 fm [9]. The result is a barrier of 8.4 MeV with a width of 2.8 fm. This leads to an estimated lifetime of $10^{-21+9.15}$ s, or about 1.4 ps. (Note that the energy of the minimum is negative, which makes the collapse of the molecule to the ground state of ^{264}Fm energetically forbidden.) Repeating the calculation with an assumed attenuation of the shell-plus-congruence energy at contact of 20%, we find a lifetime of the order of $10^{-21+6.91}$ s, or about 8.0 fms.

The above estimates are based on the frozen density idealization. What happens when this restriction is removed and a neck is allowed to grow between the fragments? There are two opposing tendencies that will decide the outcome. The macroscopic energy, say M, can be reduced by filling the space between the fragments with a neck, which drastically reduces the surface energy. The reduction can be estimated by comparing the *macroscopic* energy of the approaching fragments *i.e.*, the energy E(approach) used above augmented by 27.11 MeV, with the macroscopic energy of the ^{264}Fm nucleus along its symmetric fission valley. I find that at the locations of the minimum and maximum of the energy pocket under study, the latter energy is 3.5 MeV and 1.8 MeV above the ground state of ^{264}Fm. It follows that the macroscopic energy would benefit by 15.3 MeV and 27.8 MeV, respectively, by the filling of the neck region. Opposed to this is the loss of the (negative) shell-plus-congruence energy, which will be denoted by S. The price of this loss would be 27.11 MeV (both at the elongation of the minimum and of the maximum) if no account were taken of the attenuation of S with decreasing separation of the fragments. This changes to 23.75 MeV at the minimum and 26.15 MeV at the maximum in the "10% scenario" (reduction of S by 10% at contact). In the 20% scenario the corresponding numbers are 20.4 MeV and 25.2 MeV. The above numbers show that the competing energies have roughly similar magnitudes, with the shell-plus-congruence resistance to neck growth having the upper hand near the minimum, but the

macroscopic tendency for neck growth having a slight edge near the maximum. This is a danger signal that the frozen density approximation may be failing near the maximum, and that the estimates of the barrier height may have to be revised.

In order to get a better idea of the possible outcomes of the competition between the above two tendencies I shall make a rough interpolation of the energy between the entrance channel fusion valley of two approaching fragments (assigned a neck parameter $\nu = 0$) and the fission valley (assigned a neck parameter $\nu = 1$). I shall denote the shell-plus-congruence energy in its dependence on ν by $S(\nu)$, with $S(0)$ denoted by S_0 and $S(1)$ by S_1. The macroscopic energy will be written as $M(\nu)$, with $M(0)$ denoted by M_0 and $M(1)$ by M_1. I shall interpolate between M_0 and M_1 by a parabola with a minimum at $\nu = 1$ as follows:

$$M(\nu) = M_1 + (M_0 - M_1)(1 - \nu)^2 \,. \tag{2}$$

To represent the attenuation of the shell-plus-congruence correction with increasing ν I shall also use a parabola (inverted) up to its maximum at some neck coordinate $\nu = \nu_1$, where $S(\nu_1) = 0$, followed by zero for $\nu > \nu_1$. Thus:

$$S(\nu) \;= \begin{cases} \frac{S_0(\nu_1 - \nu)^2}{\nu_1^2} \,, & \text{for } \nu < \nu_1 \,, \\ 0 \,, & \text{for } \nu > \nu_1 \;\; . \end{cases} \tag{3}$$

The reason for introducing ν_1, which determines the range of the damping function $S(\nu)$, is that this range may well be different (smaller) than the range of the macroscopic interpolation function $M(\nu)$.

The condition for the neck growth to be inhibited is that the total energy $E(\nu) = M(\nu) + S(\nu)$ should have a positive slope at $\nu = 0$. This will be satisfied if

$$M_1 - M_0 - \frac{S_0}{\nu_1} > 0 \,. \tag{4}$$

From what was said before, the quantities entering Eq. (4) have the following values (in MeV). At the minimum: $M_1 = 3.5$, $M_0 = 18.8$, $S_0 = -23.75$ (10% scenario), $S_0 = -20.4$ (20% scenario). At the maximum: $M_1 = 1.8$, $M_0 = 29.6$, $S_0 = -26.15$ (10% scenario), $S_0 = -25.2$ (20% scenario).

If $M_1 - M_0 - S_0 > 0$ (this is the case at the minimum of the potential pocket) Eq. (4) is satisfied for any value of ν in the range 0 to 1. But if $M_1 - M_0 - S_0 < 0$ (this is the situation at the maximum) then ν_1 must be less than $-S_0/(M_0 - M_1)$ to prevent neck growth. This translates into $\nu_1 < 0.94$ or $\nu_1 < 0.91$ in the 10% and 20% scenarios, respectively. Thus, with $\nu_1 = 1$, there would be a slight tendency for the neck to open around

the location of the maximum, and the barrier against disintegration and the deduced lifetime would be a little smaller.

A striking consequence of assuming that ν_1 is less than 1, is the appearance of 'bimodal' fission in the topography of the potential energy surface in the space of the two variables L and ν. *Two* fission valleys are now present, one along the conventional valley with a filled-in neck ($\nu = 1$), and the other corresponding to more compact shapes ($\nu = 0$). The valleys are separated by a ridge (a maximum along the ν variable). Such bimodal fission in Fm isotopes had been identified experimentally [7]. Calculations, such as those in [13–15], provide a plausible interpretation of the experimental findings. In order for our model, represented by Eqs. (2), (3), to be in qualitative agreement with experiments on bimodal fission (and with the above calculations) we are required to assume $\nu_1 < 1$, for example $\nu_1 = 0.7$. Such a value of ν_1 would imply that also at the maximum of the pocket the growth of the neck is inhibited, and there would be no need to revise the lifetime estimates on that score.

Note that Fig. 10(c) in [14] confirms the existence, in the potential energy landscape of ^{264}Fm, of a pocket that might be regarded as the sought-for quasi-molecular state. But the hollow is extremely shallow, with a barrier of the order of an MeV. Whether those calculations are sufficiently realistic in the relevant region of the deformation-energy space to constitute a serious argument against a quasi-molecule is not clear. In particular, it may be relevant that in [14] the Wigner term, together with the A-independent contribution to nuclear masses, were taken positive, whereas in the present estimates I consider the congruence energy (which replaces the Wigner term in the theory of nuclear masses used here) to be negative — see [12]. A more exhaustive investigation of the existence of a pocket, using for example a self-consistent Hartree–Fock scheme, would be indicated. But it would be essential to make sure that such calculations use parameters very accurately fitted to many ground-state *and* fission saddle-point masses, and that they are capable of describing correctly the transition of a single nucleus into two symmetric fragments.

4. Alpha and beta decay

I am able to report only the roughest kind of estimates concerning the possible stability against alpha and beta decay of a molecular configuration consisting of three ^{132}Sn nuclei in contact. By comparing the mass of such a configuration with the mass of a system in which one of the Sn nuclei has been replaced by ^{128}Cd (without changing the distance between the centers of the components, taken to be $2(1.14)132^{1/3}$ fm) one deduces that the energy of the emitted alpha particle would be 13.05 MeV. This implies

a relatively short lifetime for alpha decay, but a closer analysis would be required to decide whether it would be the controlling lifetime factor, in view of the very short spontaneous fission lifetimes estimated in the previous section. (Assuming instead that the residual system consists of a ^{132}Sn nucleus and two ^{130}In nuclei implies an alpha particle energy of 8.81 MeV, *i.e.*, decay to an excited state.)

As regards beta decay, the situation is more clear-cut. Comparing the masses of triangular systems in which the mass numbers of the three components are held fixed at 132, but the atomic number of one of them is varied from $Z = 45$ to $Z = 52$, one finds the lowest mass at $Z = 48$. With respect to this mass, the sequence of masses from $Z = 45$ to $Z = 52$ is given (in MeV) by: 12.64, 5.07, 4.05, 0.00, 2.19, 1.00, 10.20, 17.20. (Other assignments of neutrons and protons to the three components do not lower the above total masses.) Thus the choice of $Z = 50$ (*i.e.*, three ^{132}Sn components) corresponds to a system stable both against beta decay and electron capture. This system is, in fact, not far from the bottom of the valley of beta stability smoothed over shell and even–odd effects. This was unexpected, since an extrapolation of the conventional valley of beta stability would make a nucleus with $Z = 150$ and $A = 396$ very unstable against electron capture. The result is explained by the lowering of the Coulomb energy of the triangle with respect to the Coulomb energy of the equivalent sphere.

5. Summary and conclusions

I have explored the possibility of making nuclear molecules from ^{132}Sn components. Whether such a possibility exists hinges on the answer to the question whether the shell-plus-congruence energy of these doubly magic nuclei is sufficiently strong to preserve their solid-like characteristics in the face of the strains imposed by the interactions between them. I formulated this question in terms of the competition between the macroscopic and the shell-plus-congruence energies. In view of the approximations made, the answer is not clear-cut, but not entirely discouraging. It would be premature to dismiss out of hand the possibility that a triangular quasi-molecular state with atomic number Z=150 and mass number $A = 396$ might have a lifetime several orders of magnitude longer than a conventional estimate for a spherical configuration would suggest. The stability and lifetime of a tetrahedral molecule with Z=200 and $A = 528$ is still more difficult to estimate reliably.

Information concerning the crucial question of the dumbbell's stability might emerge from refined analyses of the spontaneous fission properties of heavy Fm isotopes. In the distant future, experiments on the scattering on each other of heavy Sn isotopes, ideally ^{132}Sn on ^{132}Sn, might throw

light on the existence of a quasi-molecular state. On the theoretical side, a more incisive study of the dumbbell and triangle configurations would be illuminating. In such an effort, guidance from Adam Sobiczewski would, as always, be invaluable.

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