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EVIDENCE FOR SUBSHELL AT N = 152

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Large breaks in the plots of alpha particle energy vs. mass number at constant Z give striking evidence[1] for the closing of a major shell at 126 neutrons (N = 126). We have applied this sensitive criterion to look for the much smaller "subshell" effects which might be found in the region N > 126 and find good evidence for a subshell at N = 152.

The plot of alpha particle energies[2] vs. mass number for the isotopes of californium (Z = 98) shown in Fig. 1 shows a definite break in the region of Cf^{250} to Cf^{252} indicating a subshell at 152 neutrons. This break is of the order of a few percent as large as that corresponding to the major shell at N = 126 in the analogous curve for polonium.[1]

Since the postulated existence of a subshell at 152 neutrons depends entirely on the mass assignments of the nuclides involved, it appears worthwhile to review and expand the evidence on which these mass assignments are based.

<u>Cf^{244} and Cf^{246}</u>. These two isotopes of californium have been well established and identified by their respective curium daughters, Cm^{240} and Cm^{242}.

<u>Cf^{248}</u>. This isotope has been produced by He^{++} bombardment of curium containing Cm^{245} as its heaviest constituent. The yield of this activity was consistent with the reaction $Cm^{245}(n,\gamma)Cf^{248}$. The half-life observed agrees very well with that predicted for an even neutron isotope of

californium with the alpha energy observed. Additional proof of its even neutron nature is afforded by the fact that this isotope has been found to undergo spontaneous fission with a half-life comparable to that of Cf^{246}.

Cf^{249}. Recent measurements[3] show the alpha spectrum of this nuclide to consist of two prominent alpha groups: 6.00 Mev (10 percent) and 5.82 Mev (90 percent); this is characteristic of many even proton-odd neutron nuclides but not of even-even alpha emitters. This isotope of californium has been observed to grow in carefully purified berkelium samples produced by multiple neutron capture on plutonium; the mass number of this isotope has been shown to be 249 by mass spectrometer measurements.[4] That the point in Fig. 1 for Cf^{249} (N = 151) is lower than that for Cf^{250} (N = 152) is not surprising since the 6.00 Mev energy may not represent the ground state transition for this odd neutron nuclide and, in any event, in the case of the major shell at N = 126 we have an analogous situation with such pairs as Po^{209} (N = 125) 4.877 Mev, Po^{210} (N = 126) 5.298 Mev and Em^{211} (N = 125) 5.847 Mev, Em^{212} (N = 126) 6.262 Mev.[5]

Cf^{250}. Short neutron bombardments[3] of Bk^{249} have produced a 3.13 hour beta-activity which can be assigned to Bk^{250}. The decay of this activity produces a 6.05 Mev alpha-emitter with a half-life of about 12 years which is thus assigned to Cf^{250}. The relationship between alpha half-life and energy for even isotopes of californium is satisfied by these data. This isotope has been found to undergo spontaneous fission at a sufficiently high rate to indicate an even number of neutrons.

Cf^{252}. The weight of the argument that there is a subshell at 152 neutrons falls chiefly on the conclusion that the 6.15 Mev californium is heavier than that emitting 6.05 Mev alpha particles and that both of these activities are due to even neutron isotopes. The following experiments seem to bear this out quite conclusively. (1) Pure californium (Cf^{249}) which had been chemically separated after growing into a carefully purified berkelium (Bk^{249}) was subjected to the maximum MTR neutron flux for one week. The resultant californium was found to consist almost entirely of 6.05 Mev alpha activity; less than 2 percent of the activity had an energy of 6.15 Mev. (2) Longer neutron bombardments of Bk^{249} have produced both the 6.15 Mev and the 6.05 Mev californium alpha activities. The ratio of the 6.15 to 6.05 Mev activities was found to increase as the bombardment time was lengthened, as would be the case if the 6.15 Mev activity were produced by a reaction rate of higher order than that for the 6.05 Mev californium. (3) Direct decay measurements[3] made over a period of several months on relatively pure samples of the 6.15 Mev activity indicate that the half-life of this activity is about two years. This is shorter than would be expected for an odd neutron californium isotope with this energy and these data coupled with the information that this isotope exhibits a three percent branching for spontaneous fission leads to the conclusion that its mass number is probably 252 rather than 251. So far no radioactivity has been found that might be associated with Cf^{251}; this is not surprising since a combination of a high neutron capture cross section and a long alpha half-life as expected for an odd mass isotope would make it difficult to make such an identification. Recent mass spectrometer

measurements[4] on californium isotopes are in agreement with the above assignments of the 6.15 and 6.05 Mev alpha activities to Cf^{252} and Cf^{250}, respectively.

The curve for californium in Fig. 1 is sketched in beyond N = 154 on the assumption that the behavior is analogous to that for polonium[1] in the region of the major shell at N = 126. In the same spirit, analogous curves based on a subshell at N = 152 are sketched in for elements 99 and 100 using the experimental data available.[6] Similar curves for Z < 98 might also be expected due to closed subshells at Bk^{249}, Cm^{248}, etc.

So definite a subshell at this point does not seem to be predictable from a simple consideration of the shell model of Mayer[7] and Haxel, Jensen, and Suess.[8] The major shell of 126 neutrons is attained upon completion of the $i_{13/2}$ level and the simple order of levels following this, which of course is not necessarily the actual order,[9] might be $i_{11/2}$, $g_{9/2}$, $g_{7/2}$, $d_{5/2}$, $d_{3/2}$, $s_{1/2}$, $j_{15/2}$. A possible order of filling to give a break at N = 152 might be $g_{9/2}$, $s_{1/2}$, $g_{7/2}$, $d_{5/2}$ with the following levels $i_{11/2}$, $d_{3/2}$, and $j_{15/2}$ at a definitely higher energy. Another possibility is that the break comes after the filling of the $g_{9/2}$ and $j_{15/2}$ shells. However, an energy break of this sort interpreted simply on the basis of subshell completion in the process of the filling of levels in the simple single particle shell model may be an oversimplification because this is just the region where the strong surface coupling caused by large spheroidal distortion[10-13] or configuration interaction[14-18] of several nucleons may be important. In this connection one might expect on the basis of either the Bohr-Mottelson[12]

strong surface coupling model or the deShalit-Goldhaber[15] configuration interaction arguments regarding trends of first excited state energies that if the nucleon configuration at N = 152 involves only completely filled levels, the first excited state energies should approach a maximum as is observed in the closing of other shells;[19-21] the experimental evidence so far indicates that this is not the case.[4,22] Thus it seems that the 152 neutron subshell may be of a fundamentally different nature than the major closed shells.

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1. Perlman, Ghiorso, and Seaborg, Phys. Rev. 77, 26 (1950).
2. The following alpha energies with corresponding references were used: Cf^{244}, 7.15 Mev [Thompson, Street, Ghiorso, and Seaborg, Phys. Rev. 80, 790 (1950)]; Cf^{246}, 6.75 Mev [Ghiorso, Thompson, Street and Seaborg, Phys. Rev. 81, 154 (1951)]; Cf^{248}, 6.26 Mev [E. K. Hulet, Ph.D. Thesis, University of California Radiation Laboratory Unclassified Report UCRL-2283 (July, 1953)]; Cf^{249}, 6.00 Mev [Ghiorso, Thompson, Choppin, and Harvey, Phys. Rev. (May 15, 1954 issue)]; Cf^{250}, 6.05 Mev [Ghiorso, Thompson, Choppin, and Harvey, Phys. Rev. (May 15, 1954 issue)]; Cf^{252}, 6.15 Mev [Thompson, Ghiorso, Harvey, and Choppin, Phys. Rev. 93, 908 (1954)].
3. Ghiorso, Thompson, Choppin, and Harvey, Phys. Rev. (May 15, 1954 issue).
4. H. Diamond, et al., Phys. Rev., in press.
5. Hollander, Perlman, and Seaborg, Revs. Modern Phys. 25, 469 (1953).

6. The following alpha energies with corresponding references were used: 99^{247}, 7.3 Mev [Ghiorso, Rossi, Harvey, and Thompson, Phys. Rev. 93, 257 (1954)]; and 99^{253}, 6.63 Mev, 100^{254}, 7.22 Mev, 100^{255}, 7.1 Mev [Choppin, Thompson, Ghiorso, and Harvey, Phys. Rev. (May 15, 1954 issue); see also Fields, Studier, Mech, Diamond, Friedman, Magnusson, and Huizenga, Phys. Rev., in press]. The alpha energy for 99^{254} has been calculated from the closed cycle data involving Bk^{250} and 99^{254} beta energies and the 100^{254} alpha energy [Ghiorso, Thompson, Choppin, and Harvey, Phys. Rev. (May 15, 1954 issue); and Choppin, Thompson, Ghiorso, and Harvey, Phys. Rev. (May 15, 1954 issue)].

7. M. G. Mayer, Phys. Rev. 75, 1969 (1949); ibid., 78, 16, 22 (1950).

8. Haxel, Jensen, and Suess, Phys. Rev. 75, 1766 (1949); Z. Physik 128, 295 (1950).

9. G. T. Seaborg, Phys. Rev. 92, 1074 (1953).

10. J. Rainwater, Phys. Rev. 79, 432 (1950).

11. D. L. Hill and J. A. Wheeler, Phys. Rev. 89, 1102 (1953).

12. A. Bohr and B. R. Mottelson, Dan. Mat.-fys. Medd. 27, No. 16 (1953); Phys. Rev. 89, 316 (1953); Phys. Rev. 90, 717 (1953).

13. K. W. Ford, Phys. Rev. 90, 29 (1953).

14. D. Kurath, Phys. Rev. 80, 98 (1950).

15. A. deShalit and M. Goldhaber, Phys. Rev. 92, 1211 (1953).

16. A. R. Edmonds and B. H. Flowers, Proc. Roy. Soc. (London) A215, 120 (1952).

17. I. Talmi, Helv. Phys. Acta 25, 185 (1952).

18. B. H. Flowers, Proc. Roy. Soc. (London) A212, 248 (1952); ibid., A215, 398 (1952); Phys. Rev. 86, 254 (1952); Phil. Mag. (7), 43, 1330 (1952).

19. P. Stähelin and P. Preiswerk, Helv. Phys. Acta 24, 623 (1951).

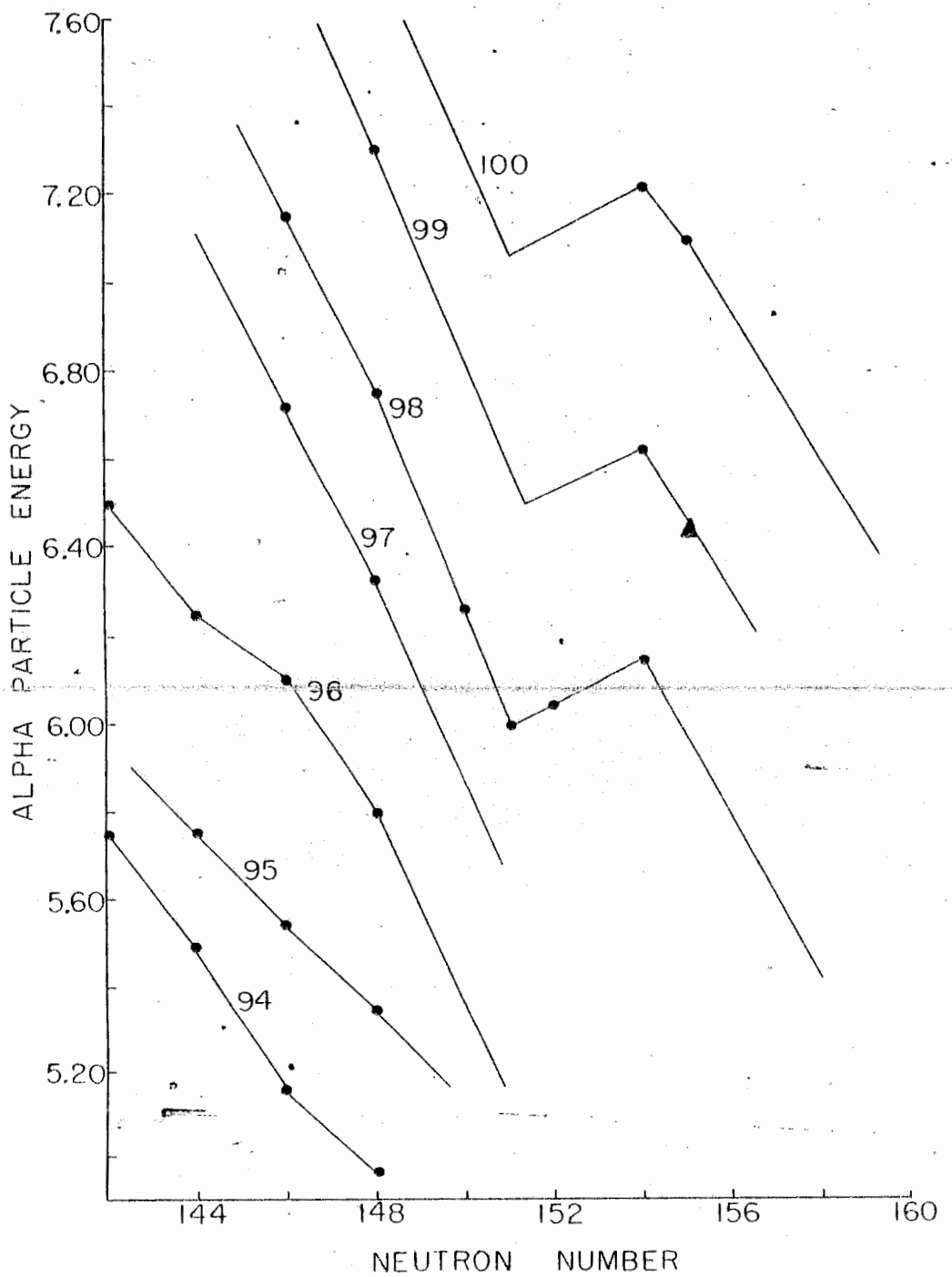
20. G. Scharff-Goldhaber, Phys. Rev. 90, 587 (1953).

21. F. Asaro and I. Perlman, Phys. Rev. 87, 393 (1952).

22. Ghiorso, Thompson, Harvey, and Choppin, unpublished work.

FIGURE CAPTION

Fig. 1. Plot of alpha energy vs. mass number.


7.60
7.20
6.80
6.40
6.00
5.60
5.20
ALPHA PARTICLE ENERGY
100
99
98
97
96
95
94
144
148
152
156
160
NEUTRON NUMBER

22263 - 2