NAL PROPOSAL No. 142

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Proposal for a Search for Superheavy Elements by Irradiations at NAL

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Abstract

We propose to irradiate certain heavy metals (tungsten through uranium) or heavy metal alloys in proton beams at NAL in order to investigate by neutron multiplicity counting the possible production of superheavy elements ($Z \geq 108$). Our counter assembly has an efficiency of 30% for a single neutron and efficiencies of about 62%, 35%, and 15% for multiplicities of ≥ 3, ≥ 4 and ≥ 5, assuming 10 neutrons per fission as has been suggested for superheavy elements. It is relatively insensitive to gamma rays and hence it provides a non-destructive test where chemical separations are not required by exceedingly high gamma radiation.

Experimenters

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Physics Justification

Th-232 (1.41×10^{10} y), U-235 (7.1×10^{8} y), and U-238 (4.51×10^{9} y) form an "island of stability" with respect to all modes of radioactive decay among the elements above bismuth. Neither the elements between bismuth ($Z = 83$) and thorium ($Z = 90$) nor protactinium ($Z = 91$) would be found in nature if they were not continually being formed by decay of the above three relatively stable nuclides. None of the elements above uranium have been definitively found in nature, except for plutonium which is formed from naturally occurring neutrons (resulting from cosmic ray showers and (α,n) reactions) reacting with U-238 nuclei. Although Pu-244 (8.3×10^{7} y) has a half-life within an order or magnitude of that of U-235, no report of its existence has been confirmed. Thus the "cut off" in half-lives in order that primordial elements on earth still exist in detectable amounts appears to be $\gtrsim 10^{8}$ y.

Myers and Swiatecki (1) pointed out that shell effects may lead to an

(1) W. D. Myers and W. J. Swiatecki, Nucl. Phys. 81, 1 (1966).

"island of stability" beyond the presently known periodic table. Since then, many authors have made extrapolative calculations (2) which indicate

(2) G. T. Seaborg, Annual Review of Nuclear Science, pp. 83-110, Palo Alto, Calif. (1968).

that somewhere between $Z \simeq 110$ (eka-Pt) and $Z = 164$ (eka-eka-Pb) there may be another "island of stability"; further, there may be additional stability for odd nuclei (3).

(3) J. Grumann, B. Fink, U. Mosel and W. Greiner, Z. Physik 228, 371 (1969).

We feel that it is premature to decide in advance which superheavy element is most likely to be relatively stable--if indeed any is! Further, we feel that relative stability in elements above about Z = 108 may mean half-lives of days to 10^{10} years or so. If the "stability" means half-lives in the lower part of the range, then of course we must rely on accelerators to produce them rather than being able to find them in nature.

In any case, if there is any appreciable enhanced stability, superheavy elements at least in principle should be produced in properly designed accelerator irradiations. Marinov and coworkers (4) claim to have found

(4) A. Marinov, et al., Nature 229, 464 (1971).

a spontaneously fissioning species and a new alpha emitter in a tungsten target irradiated at CERN. Either or both of the new species may be eka-mercury (element 112) since they seem to follow mercury chemically. The range of half-lives appears to be between a few months and about 500 years. Marinov visited ORNL about four months ago and told us that they had another tungsten target in which both the chemical yield of mercury and the fission count were down by about a factor of three from the earlier sample mentioned in the article (which showed 93 fissions in 37 days). Their targets had received about 10^{18} protons of 24 GeV.

Specifically, we have now in operation a neutron multiplicity counting assembly consisting of 20 He-3 counters in a pariffin matrix which we have constructed to look for superheavy elements in nature. Our sample volume is 9 inches in diameter and 20 inches long. It has been estimated by several nuclear scientists (e.g., 5, 6) that superheavy elements may emit some 10

(5) J. R. Nix, Phys. Lett. 30B, 1 (1969).

(6) H. W. Schmitt, private communication (1971).

neutrons in fission (if binary) compared to 2 for U-238 and 4 for Cf-252. The efficiency for counting a single neutron in our multiplicity counter is 30%; for counting $\geq$ 3, $\geq$ 4, and $\geq$ 5 out of a total of 10 in coincidence, it is 62, 35, and 15%, respectively. Our counter assembly is relatively insensitive to gamma rays, compared to a scintillation counter. We can make our assembly even more immune to gamma rays by increasing the lower limit of our energy "window" or by surrounding the sample with increasing thicknesses of lead or both. Both of these measures decrease the efficiency of our counter to some extent, but not excessively so. As far as we know, we have the only neutron coincidence counter assembly of its kind in the United States. There is a scintillation counter for somewhat similar work at Berkeley, but it is our understanding that it is very sensitive to gamma rays.

Our counter will allow non-destructive tests which may have advantages for some samples. We may wish to dissolve such samples for chemical separations only if high multiplicity events are observed beforehand. On the other hand, we may wish to conduct chemical separations or to collaborate with others who are prepared to do so, even if the neutron counter does not show positive results, in order to investigate the possible production of other radioactive species. We are now altering our multiplicity counter so that we can count fission fragments and neutrons in coincidence in chemically separated samples.

In a collaborative effort we have received two tungsten samples which were irradiated at BNL with 28 GeV protons: one of these has been sent to ANL, while the other may be separated into various fractions chemically at ORNL. Unfortunately, they received only about 10^{17} protons and, hence, even if Marinov and coworkers are correct, we did not expect to see any fission events which could be distinguished from background. We did not see any.

We also have some tantalum targets received from SLAC in a collaborative effort; we are now working on these.

We wish to collaborate with scientists at NAL and elsewhere to look for spontaneous fission events (and perhaps other new radioactivities) in targets, beam stops or whatever can be made available which consist of a heavy metal (from about tungsten to uranium) or alloy which has received a high number of energetic protons. One of us (RWS) has discussed this general problem in some detail with Ernest Malamud and Robert Sheldon, and they have made a number of suggestions (see Experimental Arrangements). Also, they already had suggested a somewhat similar proposal in a memo to E. L. Goldwasser of 12 January 1971 even before the paper of Marinov, et al. (4) appeared in print.

Experimental Arrangements

We propose to irradiate a number of heavy metals, metal sandwiches or alloys in a high integrated-intensity proton beam at NAL and look for high neutron multiplicities as an indication of superheavy element production. Any position such as that of a second target or a beam dump would be satisfactory, at least until we know more about what reactions do occur. It would be desirable to get an irradiation of at least about 10^{18} protons.

Location of Experiment

There are many places where targets could be irradiated in a search for superheavy elements. Because of the speculative nature of these searches and the premium on beam time, we propose that all such targets be irradiated parasitically to regularly scheduled experiments.

Because of the uncertain nature of the schedule and which area will likely get the 10^{18} - 10^{19} protons needed for a meaningful search, we feel that the best way to optimize the superheavy element search is to put targets in as many places as is practical. We list below several possibilities which have been discussed with members of the NAL staff: Ernest Malamud, Robert Sheldon, Richard Orr, Timothy Toohig, and Lincoln Read.

1. Enclosure G. A beam dump for the narrow band neutrino beam will be placed here at an early date, probably July 15. It is presently envisioned that this beam dump be a scrap B2 Main Ring magnet which will be placed vertically and the beam will plow into the inner coil at a slight angle. The inner coil would be water cooled. Without affecting the physics done in that beam in any way, the inner coil that the beam hits could be replaced by tungsten or heavy metal bars of the same dimension and cooling hole size as the original copper bars. They would be placed in there without insulation so that after the irradiation is completed they would be slid out from the end of the magnet and cut into pieces for shipment to Oak Ridge and analysis.

2. The high power beam dump in enclosure 100 wide band neutrino beam is presently a water-cooled module with aluminum plates in it. A section of the aluminum plates could be replaced by a high Z piece but the radioactivity handling problems need to be studied carefully.

3. The same kind of approach could be used in area PR of the proton laboratory.

4. The Adair experiment will use a high Z target and probably when his experiment is over that target could be obtained for superheavy element searches. There have been no discussions with Adair yet.

5. As an alternate for the high power beam dump module enclosure 100 of the neutrino lab or in the proton lab a mercury dump could be designed where the mercury is continuously circulated through a cylinder and to a heat exchanger outside the dump. Mercury provides efficient cooling for the entrance window into the dump which makes it easy to subdivide the radioactivity for shipment to other labs, and allows the interesting possibility

of "tapping the barrel" from time to time. The mercury dump also allows other interesting radiochemistry (not included in this proposal) involving very short-lived nuclides because it is being continuously circulated and may be sampled rapidly following a beam pulse.

Time Estimates

It is difficult to estimate the total time needed for testing and data accumulation since we do not know what we will find, how long an irradiation is required, what is the best target material, etc. We expect to continue on this problem for at least a year, but the total time will depend on our findings in the earlier stages.

Apparatus

As indicated above, our neutron multiplicity counter has been in operation for a few months, and it will soon be adapted to count fission fragments (in isolated samples) and neutrons in coincidence.

We would like to collaborate in this program with personnel at NAL, particularly with Ernest Malamud and Robert Sheldon. It would seem most efficient to have the beam dump components fabricated at NAL, although some of these could be fabricated in the ORNL Metals and Ceramics Division if necessary.