Alpha-Decay Properties of Some Lutetium and Hafnium Isotopes Near the 82-Neutron Closed Shell

RONALD D. MACFARLANE
Department of Chemistry, McMaster University, Hamilton, Canada
and
Lawrence Radiation Laboratory, University of California, Berkeley, California
(Received 2 November 1963)

The 84- and 85-neutron isotopes of lutetium, Lu^{155} and Lu^{156}, and the 85- and 86-neutron isotopes of hafnium, Hf^{157} and Hf^{158}, were produced by bombarding a Sm^{144} target with high-energy F^{19} and Ne^{20} ions. These nuclides decay primarily by alpha-particle emission. Alpha-decay energies and half-lives were measured. An alpha-emitting isomeric state was observed for Lu^{156}. The alpha reduced widths of these nuclides are close to those observed for other isotones in this region. A search was made for proton-decay branches but none was observed. Some results of a $Sm^{144}+S^{32}$ bombardment are presented, and speculations are made with regard to the future production of the ultra-neutron-deficient 84- to 86-neutron isotopes above hafnium.

I. INTRODUCTION

In previous papers we have reported on the decay properties of alpha emitters from Tb to Yb.[1-4] This paper gives results obtained for some new Lu and Hf alpha-emitting nuclides and brings to a conclusion our study of the alpha-decay properties of nuclides containing between 84 and 86 neutrons. The data presented here will be incorporated in a future paper dealing with the systematics of the alpha decay properties of the rare-earth alpha emitters.

II. EXPERIMENTAL DETAILS

The nuclides, Lu^{155} and Lu^{156}, were produced by the reactions $Sm^{144}(F^{19},8n)Lu^{155}$ and $Sm^{144}(F^{19},7n)Lu^{156}$. The Hf isotopes which were also identified in this study were produced by similar reactions using the same Sm^{144} target and a Ne^{20} beam. The Berkeley heavy-ion linear accelerator was used as the source of high-energy heavy ions. Details of the target assembly, beam-energy degradation, and the method of collection of the activity for alpha-particle analysis are given in a previously published paper.[5] The isotopic analysis of the Sm^{144} target was as follows: Sm^{144}—94.6%, Sm^{147}—1.61%, Sm^{148}—0.87%, Sm^{149}—0.83%, Sm^{150}—0.41%, Sm^{152}—0.98%, Sm^{154}—0.69%.

Alpha-particle energy measurements were made using as standards the following nuclides: Dy^{150}(4.23 MeV),[1] Er^{152}(4.80 MeV),[3] and Po^{210}(5.30 MeV).[6] A calibrated pulse generator provided additional points. Half-life measurements were obtained by analysis of alpha-particle spectra recorded at different times after bombardment.

[1] R. D. Macfarlane and D. W. Seegmiller, Nucl. Phys. **53**, 449 (1964).
[2] R. D. Macfarlane and R. D. Griffioen, Phys. Rev. **130**, 1491 (1963).
[3] R. D. Macfarlane and R. D. Griffioen, Phys. Rev. **131**, 2176 (1963).
[4] R. D. Macfarlane, Phys. Rev. **136**, B941 (1964).
[5] R. D. Macfarlane and R. D. Griffioen, Nucl. Instr. Methods **24**, 461 (1963).
[6] A. H. Wapstra, Nucl. Phys. **57**, 48 (1964).

From the intensities of the various alpha-groups cross-section data were obtained relative to Dy^{150} production, and then converted to absolute cross sections. This was done by measuring the cross section for Dy^{150} production by the reactions $Sm^{144}+F^{19}$ and $Sm^{144}+Ne^{20}$ relative to that by $Pr^{141}+F^{19}$ and $Ce^{140}+Ne^{20}$. The absolute cross sections for Dy^{150} production by the latter two reactions have been measured by Alexander.[7] For $Pr^{141}+F^{19}$ the normalization point was taken at 166 MeV, where the Dy^{150} cross section was taken as 260 mb. For $Ce^{140}+Ne^{20}$ the normalization point was at 191 MeV, lab, where the Dy^{150} cross section has a value of 262 mb. For both reactions, the Dy^{150} cross section is fairly constant over a broad range of bombarding energies.

III. RESULTS

The general method used for assigning the new alpha activities observed in the $Sm^{144}+F^{19}$ and $Sm^{144}+Ne^{20}$ bombardments was the same as used in our work on Tm and Yb alpha emitters.[4] Because of the short half-lives and low yields it was not possible to use chemical techniques to identify the elements involved or to establish parent-daughter relationships. For the activities assigned to Lu, element assignments were made on the basis of their production as products of the $Sm^{144}+F^{19}$ reaction and the absence of these activities as products of $Sm^{144}+O^{16}$ and $Nd^{142}+Ne^{20}$ reactions. The latter two reactions can produce all of the nuclides observed in the $Sm^{144}+F^{19}$ bombardments with the exception of isotopes of Lu.

Similarly, activities detected in the $Sm^{144}+Ne^{20}$ bombardments were assigned to isotopes of hafnium if they were not observed in the $Sm^{144}+F^{19}$ experiments.

Mass assignments were made on the basis of the results of excitation function measurements. Information obtained previously on the energetics of (H.I.,xn) reactions (where H.I. means heavy ion) in the rare-earth region were used to characterize the reaction

[7] J. Alexander (private communication).

leading to the production of a particular Lu or Hf alpha activity.[2–4]

A. $Sm^{144}+F^{19}$

The Sm^{144} target was bombarded with 130- to 185-MeV F^{19} ions and several alpha-particle spectra of the products were obtained. The spectra contained a large number of groups in the energy region of 3.9 to 5.7 MeV. The 84- and 85-neutron isotopes of Dy, Ho, Er, Tm, and Yb were readily identified. In addition, a very weak group was observed at an alpha-particle energy of 5.63 MeV, a second more intense group at 5.54 MeV, and a third weak group at 5.43 MeV. These activities were not seen in the $Sm^{144}+O^{16}$ or $Nd^{142}+Ne^{20}$ bombardments and were thus assigned to isotopes of lutetium.

Lu^{155} (E_α = 5.63 MeV)

Alpha-particle spectra containing the weak 5.63-MeV alpha group are shown in Fig. 1. This activity was found to decay with a half-life of 0.07 ± 0.02 sec. The excitation function for the production of this activity [Fig. 2(a)] peaks at a bombarding energy of 160 MeV which corresponds to an excitation energy of 111 MeV. (The excitation energy referred to here is the compound nucleus excitation energy and is obtained by subtracting the Q value for compound nucleus formation from the center-of-mass bombarding energy.[8] The peak cross section for the production of this activity (based on a 100% alpha branch) was found to be 0.07 mb.

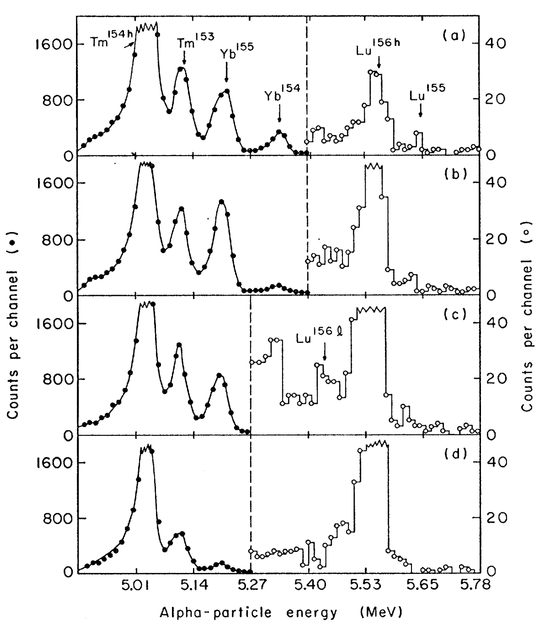

FIG. 1. Alpha-particle spectra of some of the products of the $Sm^{144}+F^{19}$ reaction showing the new Lu alpha emitters. Spectra were taken at bombarding energies of (a) 180 MeV, (b) 166 MeV, (c) 155 MeV, (d) 144 MeV. The closed circles refer to the scale on the left and the histogram to the scale on the right.

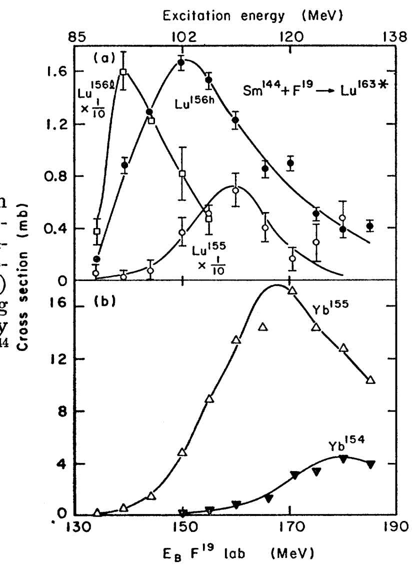

FIG. 2. Excitation functions for the production of (a) the 84- and 85-neutron isotopes of Lu and (b) the corresponding isotopes of Yb by the reaction $Sm^{144}+F^{19}$.

From our previous work on the alpha emitters below Lu, we have been able to accumulate a considerable amount of information regarding the energetics of (H.I.,*xn*) and (H.I.,*pxn*) reactions in the rare-earth region.[2–4] To summarize those results pertinent to this work, we have established that the excitation functions for (H.I.,8*n*) reactions and (H.I.,*p*7*n*) reactions peak at excitation energies between 114 and 118 MeV. For (H.I.,7*n*) and (H.I.,*p*6*n*) reactions these excitation functions peak at excitation energies between 103 and 106 MeV. (The excitation functions for low-spin isomer production peak at significantly lower energies because of angular-momentum effects.)

From our $Sm^{144}+F^{19}$ measurements, we found that the excitation function for Yb^{155} production by a (F^{19},*p*7*n*) reaction peaks at an excitation energy of 117 MeV, a value in good agreement with previous results for the same reaction [Fig. 2(b)]. The Lu^{155} excitation function should also peak at the same excitation energy. However, the excitation function for the 5.63-MeV alpha group which we suspected to be due to Lu^{155} peaks $\approx$6 MeV lower than this, at a value half-way between the (H.I.,8*n*) and (H.I.,7*n*) energies. We did observe an activity, however, which was assigned to the (F^{19},7*n*) reaction so that the only plausible mass assignment for this activity is Lu^{155}.

Because of the very low intensity of this alpha group, there is considerable uncertainty in the cross-section measurements but not enough to account for the observed shift in the excitation functions. We feel that the shift is indeed real and is caused by the increasing tendency of compound nuclei of high angular momentum to fission as these compound nuclei and their neutron evaporation products become more neutron-

[8] P. A. Seeger, Nucl. Phys. 25, 1 (1961).

deficient. A more detailed account of these effects will be presented in a future paper.

$Lu^{156h}(E_\alpha = 5.54\ MeV)$

A second group was observed at an alpha-particle energy of 5.54 MeV. This alpha group is present in the spectra shown in Fig. 1. The half-life for this activity was found to be 0.23±0.03 sec. The excitation function for the production of this activity by the reaction $Sm^{144}+F^{19}$ is shown in Fig. 2(a) and is labeled Lu^{156h} which denotes that it is a high-spin isomer. The peak of the excitation function falls at an excitation energy of 103 MeV which is in good agreement with the values previously observed for (H.I.,$7n$) reactions. (See the above discussion of the Lu^{155} results.) The mass assignment of this activity must, therefore, be Lu^{156}. Another Lu activity, the results of which are described below, has been identified as a low-spin isomer of Lu^{156}. The more intense 0.23-sec Lu^{156} activity has been assigned to a high-spin isomeric state on the basis of the excitation function results.

$Lu^{156l}(E_\alpha = 5.43\ MeV)$

A third Lu-alpha activity with a half-life of ≈0.5 sec was found at an alpha-particle energy of 5.43 MeV. This alpha group can be seen in Fig. 1(c). The excitation function for this activity is shown in Fig. 2(a). The peak cross section which occurs at an excitation energy of 92 MeV, was found to have a value of 0.16 mb (assuming 100% alpha branch). We have assigned this activity to a low-spin isomer of Lu^{156} on the basis of the following arguments. The excitation function has a pronounced asymmetric shape with a sharp cutoff on the low-energy side and a tail on the high-energy side. Excitation functions of the same shape were found in the results obtained for the low-spin isomers of Tb^{149}, Ho^{151}, and Ho^{152} produced by similar heavy-ion reactions.[2,9] The peak of the excitation function is ≈10 MeV lower than for the activity we have assigned to Lu^{156h}. This is consistent with the results obtained for the Tb^{149}, Ho^{151}, and Ho^{152} isomer pairs. The peak cross section for the production of this activity is much lower than one would expect for a (H.I.,$6n$) reaction in this region. (See discussion of Hf^{158} results below.) This too, is consistent with previous results on the production of low-spin isomers by heavy-ion reactions. All of these observations can best be explained by assigning this activity to a low-spin isomer of Lu^{156}.

B. $Sm^{144}+Ne^{20}$

Hafnium isotopes near the 82-neutron closed shell were produced by bombarding a Sm^{144} target with high-energy Ne^{20} ions. The 84-neutron isotope, Hf^{156}, can be produced by a (Ne^{20},$8n$) reaction. A careful search was made for Hf^{156} alpha decay. This nuclide was expected to have an alpha-particle energy between 5.75 and 5.85 MeV. However, no groups were observed in this energy region when bombardments were made over a range of bombarding energies considered to give the optimum yield for this activity. Most likely, the reason the activity was not seen in the $Sm^{144}+Ne^{20}$ bombardments was because of its short half-life. The shortest half-life activity which can be detected in our experiments is 0.03 sec. On the basis of the predicted alpha decay energy, the half-life of Hf^{156} may be as short as 0.01 sec.

At the lower bombarding energies (130 to 170 MeV) two alpha groups were observed which had not been seen in the $Sm^{144}+F^{19}$ study. These activities were assigned to isotopes of hafnium. Excitation function data showed that they were due to Hf^{157} and Hf^{158}.

$Hf^{157}(E_\alpha = 5.68\ MeV)$

One of the Hf alpha groups was observed at an alpha particle energy of 5.68 MeV and was found to decay with a half-life of 0.12±0.03 sec. This group can be seen in the spectra shown in Figs. 3(c) and 3(d). The excitation function for the production of this activity by the reaction $Sm^{144}+Ne^{20}$ [Fig. 4(a)], has a peak cross section of 0.2 mb at an excitation energy of 106 MeV. This energy agrees closely with values observed for (H.I.,$7n$) reactions, which means that this activity is due to Hf^{157}.

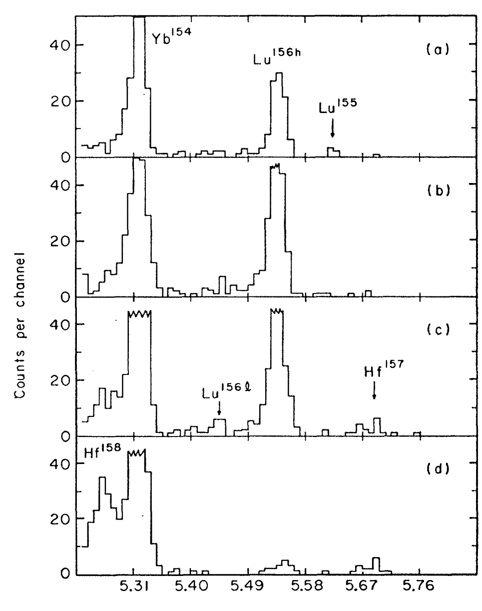

FIG. 3. Alpha-particle spectra of some of the products of the $Sm^{144}+Ne^{20}$ reaction showing the new Hf alpha emitters. Spectra were taken at bombarding energies of (a) 195 MeV, (b) 183 MeV, (c) 171 MeV, (d) 159 MeV.

[9] R. D. Macfarlane, Phys. Rev. **126**, 274 (1962).

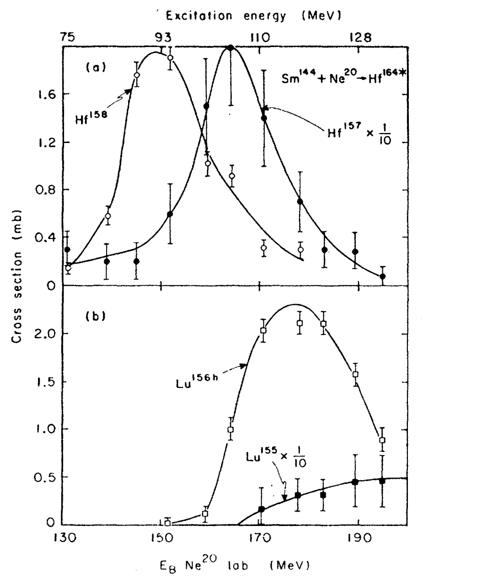

FIG. 4. Excitation functions for the production of (a) the 85- and 86-neutron isotopes of Hf and (b) the 84- and 85-neutron isotopes of Lu, by the reaction $Sm^{144}+Ne^{20}$.

$Hf^{158}(E_\alpha=5.27\ MeV)$

The second Hf alpha group has an alpha-particle energy of 5.27 MeV and decays with a half-life of 3±0.5 sec. This group is clearly seen in the spectra shown in Figs. 3(c) and 3(d). The excitation function for this activity is shown in Fig. 4(a). The cross section reaches a maximum value of 2 mb at an excitation energy of 92 MeV. Previous results on the production of Ho, Er, and Yb isotopes showed that the (H.I.,$6n$) reaction peaks at an excitation energy between 86 and 88 MeV for these nuclides.[2–4] One would expect that the (Ne^{20},$6n$) excitation function to produce Hf^{158} would peak at a little higher energy because of the higher neutron binding energies in the region around Hf^{158}. The results obtained above, therefore, are consistent with the mass assignment of Hf^{158} to this activity.

An alpha-particle spectrum taken with a large detector for a long bombarding time is shown in Fig. 5. The Hf^{157}, Lu^{155}, and Lu^{156} activities which were not very prominent in the spectra shown in Figs. 1 and 3 are more clearly seen in this spectrum.

IV. DISCUSSION

A. Estimation of Alpha-Branching Ratios

In order to relate the data obtained in this work to alpha-decay theory the alpha-branching ratios of these nuclides are needed. An estimate of alpha-branching ratios was obtained by adopting gross logft values for these nuclides based on decay data for neighboring nuclides in this region and using calculated estimates of the beta-decay energies of these nuclides. Analysis of the data for Dy, Ho, and Er alpha emitters near the

TABLE I. Summary of results.

Nuclide	Q_α (MeV)	$t_{1/2}$ (sec)	Estimated alpha branch	δ^2 (MeV)
Lu^{155}	5.78±0.03	0.07±0.02	100%	0.08
Lu^{156h}	5.69±0.02	0.23±0.03	85%	0.049
Lu^{156}	5.57±0.03	≈0.5	70%	0.05
Hf^{157}	5.83±0.02	0.12±0.03	100%	0.085
Hf^{158}	5.41±0.02	3±0.5	80%	0.1

82-neutron shell showed that gross logft values grouped around a value of 5. Taking this as the gross logft for the nuclides reported in this study and using Q_{β^+} values derived from the Seeger mass formula,[8] we estimated beta-decay half-lives and from the observed half-lives obtained approximate alpha-branching ratios. These values are given in Table I which also summarizes the experimental results.

B. Alpha Reduced Widths

Alpha reduced widths (δ^2) are proportional to the probability of alpha decay after the alpha-decay energy and angular momentum dependence has been removed. The exact definition of δ^2 and the method of calculation is that given by Rasmussen.[10] The reduced widths which were calculated using the experimental alpha-decay energies and estimated alpha half-lives are given in Table I.

These reduced widths have very much the same values as those for the lighter 84- to 86-neutron isotopes. In a recent paper we have calculated relative reduced widths between $Z=54$ and $Z=80$ for a constant $N=82$ neutron number using product wave functions derived from Bardeen-Cooper-Schrieffer calcu-

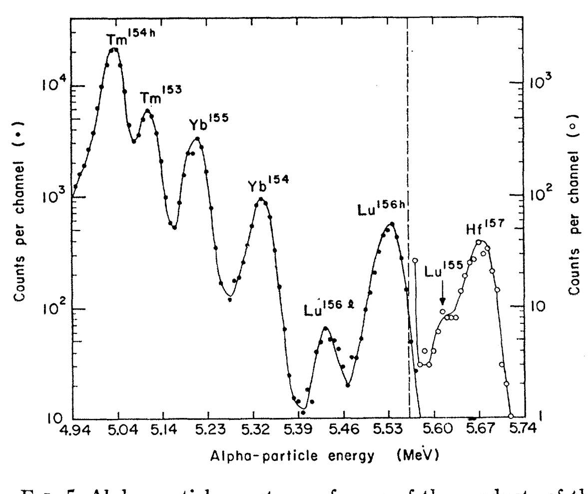

FIG. 5. Alpha-particle spectrum of some of the products of the $Sm^{144}+Ne^{20}$ reaction taken at a bombarding energy of 171 MeV. This spectrum shows the very weak alpha groups of Lu^{156}, Lu^{155}, and Hf^{157}.

[10] J. O. Rasmussen, Phys. Rev. 113, 1593 (1959).

lations.[11] These results show that because of the effect of extensive configuration mixing in this region, the alpha-decay transition matrix elements are little affected by the addition of a few protons and a nearly constant alpha reduced width has been calculated between $Z=68$ and $Z=78$.

C. Search for Proton Radioactivity

The Lu and Hf isotopes reported here are probably the most neutron-deficient species presently known. They are very close to the region of proton instability and, in fact, Lu^{155} may be proton unstable as well as alpha-particle unstable. The Seeger mass formula gives Lu^{155} a proton binding energy of -0.24 MeV which may be a valid number since it is within 50 keV of giving the correct alpha-decay energy.[8]

Some particle spectra were obtained over the energy range of 0.5 to 3 MeV but no proton groups were observed. The low-energy tails of the intense alpha groups of the Ho, Er, and Tm activities made it difficult to observe very weak low-energy proton groups.

D. Alpha Decay in the 82-Neutron Region above Hafnium. Some Results of $Sm^{144}+S^{32}$ Bombardments

The problem in studying the 84- and 85-neutron alpha emitters above hafnium presently lies in the production of these nuclides in sufficient quantities to measure their decay properties. We were able to extend our study of the decay of these nuclides to as high as hafnium because of the special stability of the neutron-deficient target nucleus Sm^{144}. However, we have found no available reaction which could be expected to give any reasonable cross section for the production of the corresponding isotopes of the elements above hafnium. These nuclides may exhibit unusual decay properties. For example, the 84- and 85-neutron isotopes of Ta, Ta^{157}, and Ta^{158}, are probably proton unstable as well as alpha-particle unstable. The isotopes, W^{158} and W^{159}, may exhibit the 2-proton decay which has been predicted by Goldansky.[12]

As a final experiment to determine whether any new information could be obtained near the 82-neutron closed shell for the elements above Hf, we bombarded our enriched Sm^{144} target with high-energy (200 to 300 MeV) S^{32} ions in order to determine whether some of the 84- and 85-neutron isotopes between Ta and Os could be produced. The alpha-particle spectra of the products showed that the nuclides Dy^{150}, Ho^{151}, $Er^{152,3}$, $Tm^{153,4}$, and Yb^{155} were produced in good yield at 300-MeV bombarding energy. The yields of Yb^{154}, $Lu^{155,6}$, and Hf^{157} were considerably reduced, but still readily detectible. A new alpha group was observed at an alpha-particle energy of 5.75 MeV. The yield of this activity and all the others detected at 300 MeV dropped sharply with decreasing bombarding energy. We can only speculate on the origin of the 5.75-MeV group because of the lack of more information about it. A possible explanation is that it arises from the decay of W^{160} which should have an alpha-particle energy around 5.75 MeV if the systematics of alpha-decay energies continue in a regular manner above Hf. For this energy the half-life of this isotope may be as high as 0.2 sec which is long enough to be detected by our method of recoil collection. Interestingly, the alpha decay daughter of W^{160} is the 84-neutron isotope of Hf, Hf^{156}. This isotope will have close to the same alpha energy as W^{160} so that the group observed at 5.75 MeV may be due to a composite of the alpha decay of these two isotopes.

We feel that for the future, the most attractive modes of production of the higher members of the 84-neutron isotopes will involve the use of high-energy "super-heavy" ions. For example, it should be possible to produce W^{158} in good yield by the reaction $Sr^{84}(Kr^{80},6n)W^{158}$. A large number of "ultra-neutron-deficient" nuclides in this region should be accessible when high-energy Kr^{78} and Kr^{80} beams become available.

ACKNOWLEDGMENTS

The author would like to acknowledge the kind hospitality extended to him by Professor I. Perlman and Professor J. O. Rasmussen during his stay at Berkeley. He is also indebted to Albert Ghiorso for his contributions relating to the experimental measurements. The assistance of the Hilac personnel is gratefully acknowledged. This work was performed under the auspices of the U. S. Atomic Energy Commission.

[11] R. D. Macfarlane, J. O. Rasmussen, and M. Rho, Phys. Rev. **134**, B1196 (1964).
[12] V. I. Goldansky, Nucl. Phys. **19**, 482 (1960).