Alpha Decay of Np^{235}†

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The alpha decay of Np^{235} has been found to be complex. Four groups have been identified with energies of 4.864, 4.925, 5.015, and 5.095 Mev. The relative abundances of the respective groups are 0.8, 11.8, 83.6, and 3.8%. Hindrance factors calculated on the basis of a 7×10^4 year alpha half-life for Np^{235} are 35, 6.1, 3.3, and 250, respectively. The electron capture decay energy of Np^{235} has been determined to be 163 ± 16 kev based on a closed cycle calculation and the assumption that the 5.095-Mev alpha transition is to the ground state of Pa^{231}.

I. INTRODUCTION

NEPTUNIUM-235 decays predominantly to U^{235} by electron capture.[1–4] However, a small fraction of the decay is to Pa^{231} by alpha emission.[2–4] James et al.,[2] who first observed the alpha decay of Np^{235}, reported the energy of the alpha particles to be 5.06 ± 0.02 Mev. Later Hoff, Olsen, and Mann[3] found the alpha particles to be in coincidence with gamma rays of 26 and 85 kev and with L x rays. These gamma rays were shown to arise from a metastable state with a half-life of 37 millimicroseconds. In a recent paper[4] the present authors stated that the alpha spectrum of Np^{235} is complex and that they were not able to resolve the various alpha groups. Subsequent improvements in experimental techniques have enabled us to resolve the major alpha groups and to obtain an accurate value for the alpha decay energy.

II. EXPERIMENTAL TECHNIQUES AND RESULTS

Sample Preparation

Two samples containing Np^{235} were produced by deuteron bombardment of U^{235} targets at the Argonne 60-inch cyclotron. A target containing 93.3 atom percent U^{235} was used to prepare the first sample. A large amount of Np^{237} was contained in this sample as a result of: (1) tracer added for a previous experiment, (2) direct formation through the $U^{238}(d,3n)Np^{237}$ reaction, and (3) the β^- decay of U^{237} produced by the $U^{238}(d,p2n)U^{237}$ and $U^{238}(d,t)U^{237}$ reactions. The second sample was obtained from a target of highly enriched (>99.9 atom percent U^{235}) uranium.

Neptunium was separated from the uranium targets, fission products, and spallation products by a series of co-precipitations with lanthanum fluoride and hydroxide, followed by anion exchange using Dowex A-1 resin in a hydrochloric acid medium. The final purification was by solvent extraction using mono-octyl phosphate in toluene, thenoyl-trifluoroacetone in benzene, and diethyl ether as the extracting agents.

At the end of the chemical purification, sample 1 contained a small amount of U^{232} added as tracer for another experiment. Sample 2, as determined by alpha pulse analysis, contained only radiations of Np^{235}. The purified neptunium was volatilized as the oxide onto a 0.001-inch aluminum foil. During the volatilization process, sample 2 was contaminated with a small amount of Po^{210} (approximately 18 disintegrations per minute out of a total of 515 dis/min). No attempt was made to repurify the neptunium in this sample; instead, the Po^{210} alpha peak was used as an internal energy standard.

Alpha Spectra

The alpha chamber and electronics have been described previously.[5] The chamber was operated as a flow counter at atmospheric pressure with a commercial gas mixture of 10% methane and 90% argon.

The 2π geometry alpha spectrum of sample 2 is given in Fig. 1. It is apparent that the Np^{235} spectrum is complex, with evidence of at least two alpha groups at approximately 4.96 and 5.05 Mev. To improve the resolution, both samples were collimated with 0.002-inch Lektromesh screen.[5,6] The resulting spectra are shown in Figs. 2(a) and 2(b) for samples 1 and 2, respectively. Table I lists the alpha energies obtained in six runs: four with sample 1 and two with sample 2. The peak positions of the Np^{235} spectrum were established in the following manner. For sample 1, the U^{232} α_0 distribution was fitted by a normal distribution curve which was then used to resolve the individual alpha groups in the Np^{235} spectrum. The same procedure was followed with sample 2 except that the normal distribution curve was fitted to the Po^{210} distribution. In four of the runs the alpha peak positions were measured in terms of the voltage scale of a precision mercury relay pulse generator in the manner

* Based on work performed under the auspices of the U. S. Atomic Energy Commission.
[1] R. A. James, A. E. Florin, H. H. Hopkins, Jr., and A. Ghiorso, *The Transuranium Elements: Research Papers* (McGraw-Hill Book Company, Inc., New York, 1949), Paper No. 22.8, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, Div. IV.
[2] R. A. James, A. Ghiorso, and D. Orth, Phys. Rev. **85**, 369 (1952).
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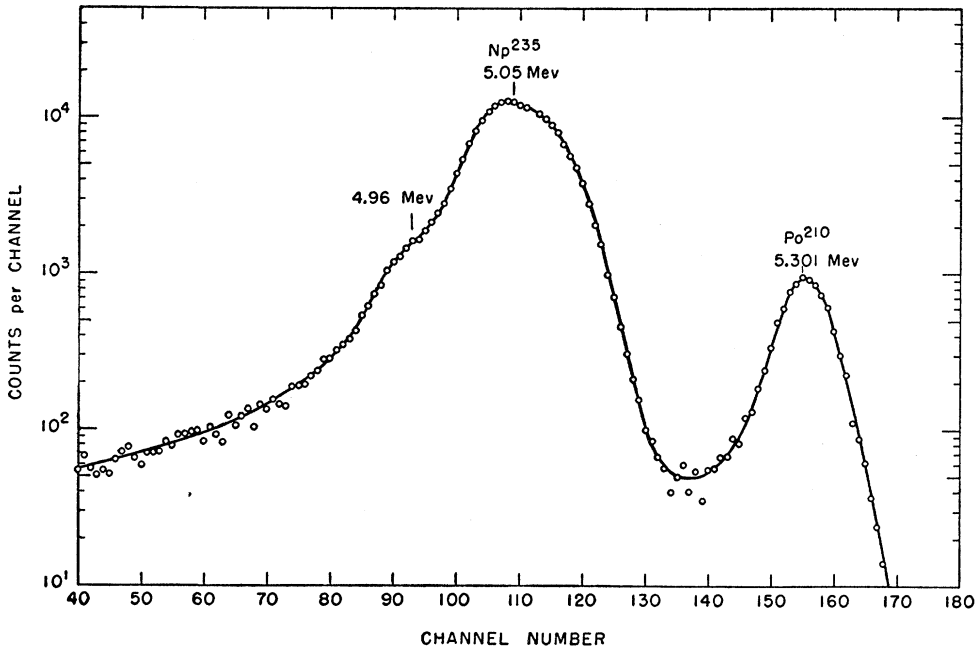

FIG. 1. 2π geometry alpha spectrum of Np^{235}, sample 2.

described by Harvey et al.[7] The voltage scale of the pulse generator was calibrated in terms of the alpha energies of the three standards, Np^{237} α_{85}, U^{232} α_0, and Po^{210}. Corrections were applied for grid shielding inefficiency (~5 kev), ionization losses within the collimator (~2 kev) , and for a zero-correction of +90 kev. This zero-correction is the intercept on the energy axis of an extrapolation of the pulse height versus energy calibration curve. In the other two runs a linear interpolation was made between the U^{232} and Np^{237} peaks to obtain the Np^{235} alpha energies.

The four alpha groups listed in Table I were assigned to Np^{235} on the basis of their decay. Runs made four and ten months after the original on sample 1 and six months after the original on sample 2 showed the various groups to decay with approximately the Np^{235} half-life.[4]

TABLE I. Energy and relative abundance of Np^{235} alpha particles; levels of Pa^{231} populated.

Run No.	α_0	α_1	α_2	α_3
495[a,b]	5.096	5.015	4.927	
498[a,b]	5.092	5.017	4.927	
587[a,c]	5.092	5.015	4.926	
594[d,b]	5.093	5.015	4.923	4.864
598[d,b]	5.096	5.016	4.925	4.864
907[a,c]	5.099	5.014	4.925	
Average	5.095	5.015	4.925	4.864
% abundance	3.8	83.6	11.8	0.8
$\alpha_i-\alpha_0$	0	0.080	0.170	0.231
Pa^{231} level energies	0	0.081	0.173	0.235

[a] Sample contained both Np^{235} and Np^{237}.
[b] Pulse generator voltage used to determine energy linearity of analyzer.
[c] Energy linearity assumed between Np^{237} α_{85} and U^{232} α_0.
[d] Sample contained primarily Np^{235}.

[7] B. G. Harvey, H. G. Jackson, T. A. Eastwood, and G. C. Hanna, Can. J. Phys. **35**, 258 (1957).

From Fig. 2(b) it is apparent that the resolution of the four alpha groups by this method leaves residual counts between the peaks. This may be attributed to either or both of the following:

(1) The alpha spectrum probably is more complex than the four groups shown.

(2) Summing of alpha and conversion electron pulses may produce a high-energy satellite to the peak of an alpha group which is in coincidence with converted gamma transitions. Although Lektromesh collimation greatly reduces this summing it does not eliminate it entirely. With 2 mil Lektromesh the sum peaks may be 10% as high as the main peak.

Broadening of the peaks by sample thickness or by the intense background of electrons and x rays associated with the electron capture branch of Np^{235} (the α branching is only 1.59×10^{-5}) should not be a factor because both the Np^{235} and the Po^{210} peaks would be affected equally. It should be noted that the peaks are broader than expected: the observed width at half-height is 36 kev whereas widths from 27 to 29 kev are usually obtained for volatilized, collimated samples.

The absolute accuracy of the alpha energies is estimated to be ±5 kev.

Coincidence Measurements

In a repetition of the coincidence experiments of Hoff et al.,[3] the gamma spectrum in coincidence with all alphas was measured with a one-eighth inch sodium iodide crystal covered with a 60-mg/cm² copper ab-

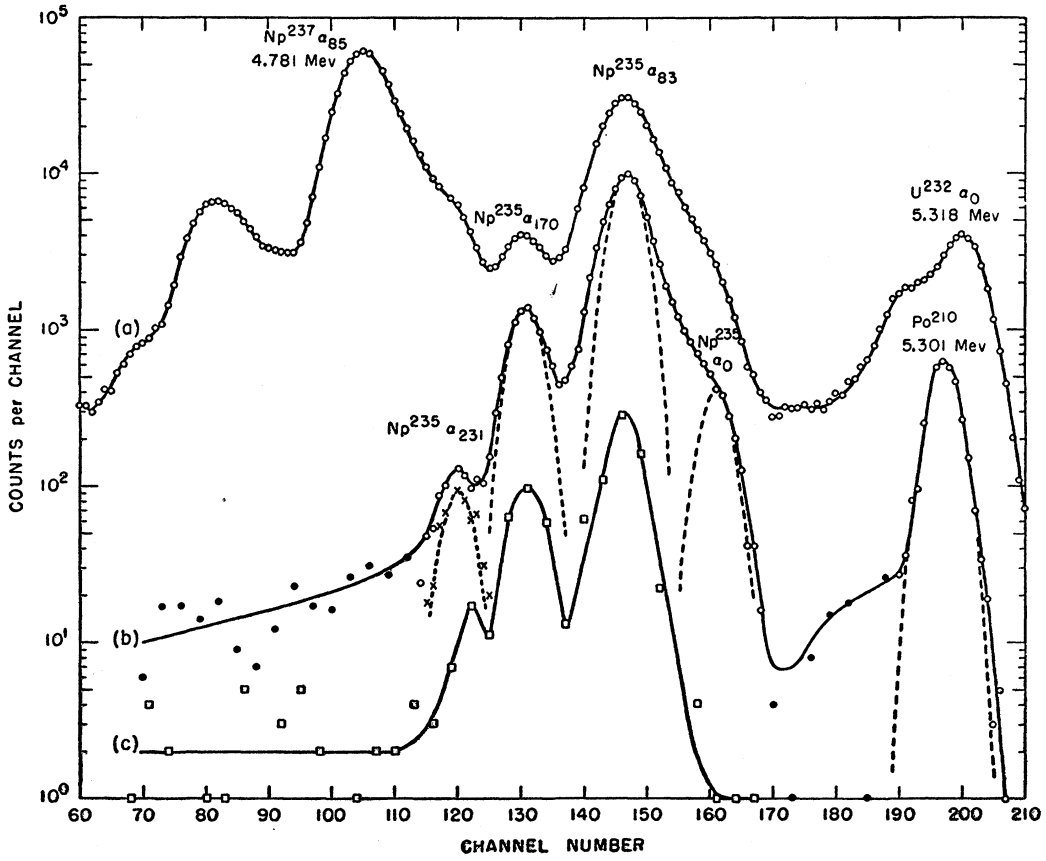

FIG. 2. Alpha spectra of Np^{235} with Lektromesh collimation. (a) Sample 1, singles. (b) Sample 2, singles, solid points are 3-channel averages. (c) Sample 2, coincidences with (70–97)-kev gamma rays, 3-channel sums plotted on mid-channel.

sorber to attenuate most of the L x rays from the intense electron capture branch. Alphas were detected with a zinc sulfide scintillation counter. The coincidence spectrum, Fig. 3, has gamma peaks at 26.5 and 84.1 kev plus what appears to be a K x ray peak. Three channel sums, plotted at the midchannel, are shown in

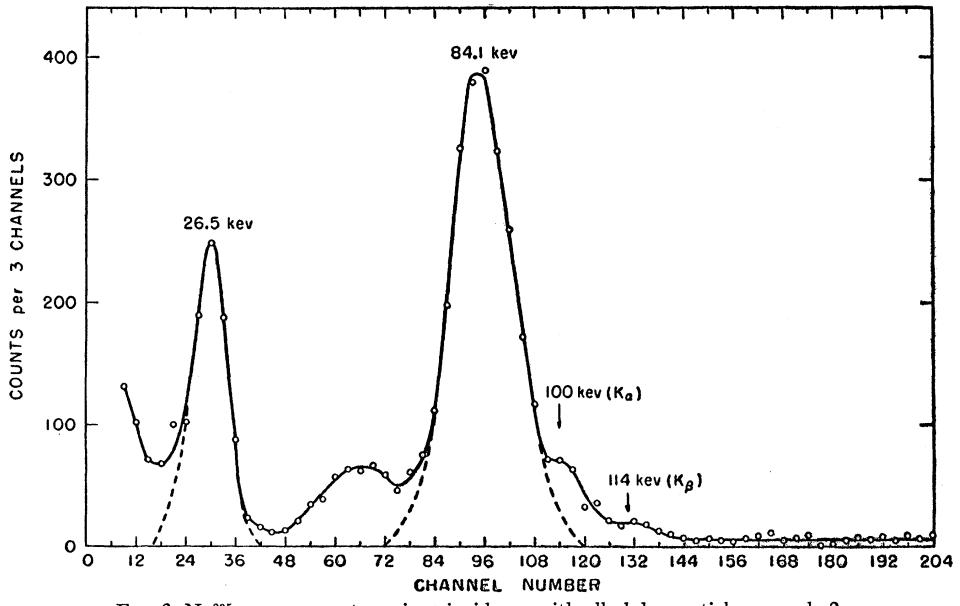

FIG. 3. Np^{235} gamma spectrum in coincidence with all alpha particles, sample 2.

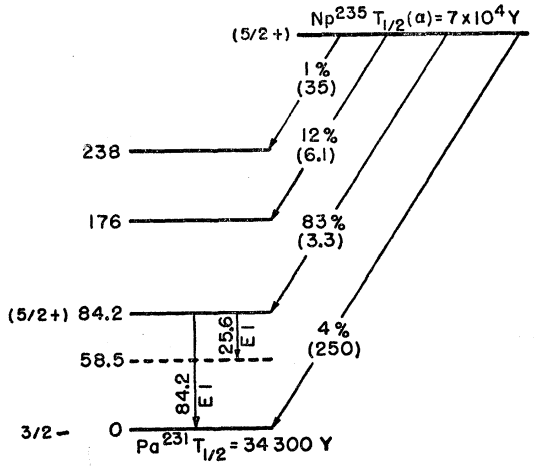

FIG. 4. Alpha decay scheme of Np^{235}.

the figure. With a resolving time 2τ equal to 84 millimicroseconds, the chance rate, obtained by adding 200 millimicroseconds delay in the gamma channel, amounted to less than one percent of the true rate and was not subtracted. Intensities of 0.15 ± 0.02, 0.088 ± 0.008, and 0.006 ± 0.002 photon per alpha were found for the 26-kev, 84-kev, and K x radiations, respectively. The intensities of the 26- and 84-kev gamma rays have been corrected for the fraction of the decays ($\sim$46%) of the 37-millimicrosecond level which did not occur within the resolving time of the circuit; the K x rays were assumed to be prompt. The probable errors include statistical errors, counting efficiency errors, and errors in the estimates of the fraction of the metastable state decays counted. These intensities are appreciably higher than those reported by Hoff et al., who found approximately 0.054 photon per alpha for both the 84- and 26-kev gamma rays.

A measurement of the alpha spectrum in coincidence with the 84-kev gamma ray was made in order to establish the position of the 84-kev gamma ray in the level scheme. Gamma pulses in the energy band from 70 to 97 kev were selected with a single channel analyzer connected to the one-eighth inch sodium iodide scintillation counter. Coincident pulses from the alpha ionization chamber were displayed on the 256-channel analyzer. A 57-mg/cm^2 copper absorber was placed over the gamma detector to attenuate L x rays. A relatively high chance rate was present (22% of the true rate) because of the long resolving time (4.6 microseconds) and the large number of K x rays from the electron capture branch which were counted by the gamma spectrometer. The coincidence spectrum, after subtraction of the chance spectrum, is shown in Fig. 2(c). Only the highest energy alpha group is missing which demonstrates that the 84-kev transition originates at or below the level populated by the most abundant alpha group of 5.015 Mev but does not follow the highest energy alpha group. The simplest interpretation is that the highest energy alpha group observed leads to the ground state and that the 84-kev transition is between the level populated by the 5.015-Mev alphas and the ground state of Pa^{231}.

TABLE II. Calculation of the electron capture energy of Np^{235}.

Nuclide	Decay mode	Q_0 (kev)	Reference
Np^{235}	α	5187 ± 5	Present work
Th^{231}	β^-	-386 ± 2	a
U^{235}	α	-4638 ± 15	b
Np^{235}	e^- capt.	$+163\pm16$	

[a] The energy was determined by adding the Th^{231} β^- decay energy of 302 kev [M. S. Freedman, A. H. Jaffey, F. Wagner, Jr., and J. May, Phys. Rev. **89**, 302 (1953)] to the 84-kev γ transition.
[b] R. C. Pilger, Jr., University of California Radiation Laboratory Report, UCRL-3877, 1958 (unpublished).

III. DISCUSSION

The alpha decay scheme deduced from all available data is given in Fig. 4. Hindrance factors in the alpha decay are given in parentheses beneath the alpha abundances. Population of the dotted level at 58.5 kev by alpha decay was not observed; however, if it did occur, it might easily have been missed. Radiations from this level to the ground state are highly converted[8] and therefore would be present only very weakly in alpha-gamma coincidence measurements. The relationship between the 58.5-kev level and the 84.2-kev metastable level has been demonstrated in the β^- decay[8] of Th^{231} and the electron capture decay[9] of U^{231}; the energy difference between the observed 84.17- and 25.65-kev gamma rays from the metastable level is 58.52 kev which corresponds closely to the energy of the 58.53-kev level. Since Newton[10] has shown that the 58.5-kev level may be reached by Coulomb excitation of Pa^{231}, the metastable level must be placed at 84.2 kev.

The ground-state spin of Pa^{231} has a measured value[11] of $\frac{3}{2}$ which has been interpreted by Stephens et al.[12] as the $I=\frac{3}{2}$ member of an odd parity $K=\frac{1}{2}$ rotational band. Their assignments of $\frac{5}{2}+$ to the 84.2-kev level of Pa^{231} and to the ground state of Np^{235} are consistent with the existence of a favored alpha transition (hindrance factor 3.3) to the 84.2-kev level.

Since the ground-state alpha energy is poorly defined by the alpha spectrum of Fig. 2, a more accurate value may be deduced from the energy of the alpha populating the 84.2-kev level and the accurately measured gamma energy. This method gives an energy of 5.099 Mev as

[8] F. S. Stephens, F. Asaro, and J. M. Hollander, 1958 (unpublished); quoted by D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 811 (1958).
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compared with the measured energy of 5.095 Mev. With a ground-state alpha energy of 5.099 Mev, the disintegration energy, Q_α, equals 5.187 Mev.

The disintegration energy of the electron capture branch of Np^{235} was calculated from the closed energy cycle shown in Table II. The result, 163±16 kev, is in approximate agreement with the value of 123 kev calculated from the K/L capture ratio.[4]

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Interaction of 6- to 14-Mev Deuterons with Helium Three and Tritium*

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The angular distributions of the cross sections for the $T(d,n)He^4$ and $He^3(d,p)He^4$ reactions have been measured at five incident deuteron energies between 6 and 14 Mev. The data have been fitted with Legendre expansions. A search for excited states in He^4 near 22 Mev and for evidence of the formation of H^4 gave negative results.

INTRODUCTION

THE present paper concludes that portion of a program, on the interactions between the hydrogen and helium isotopes, which has been concerned with the emission probabilities for charged products from reactions induced by 6- to 14-Mev deuterons on He^3 and T.

The elastic scattering experiments[1,2] have already been described, as have also the results[3] on the identification and systematics of the reaction $d+T \rightarrow He^3+n+n$.

The present measurements evaluate the differential cross sections, at a number of incident energies, for the reactions $He^3(d,p)He^4$ ($Q=18.4$ Mev) and $T(d,n)He^4$ ($Q=17.6$ Mev). A search has also been made for excited states in He^4, at ~22 Mev, and for evidence of the formation of H^4 by investigating the range distributions of the protons emitted from the breakup of the intermediate Li^5 and He^5 nuclei, respectively.

Both the $He^3(d,p)He^4$ and $T(d,n)He^4$ reactions have been previously studied at ~10 Mev.[4,5] In addition, the $T(d,n)$ reaction has been extensively surveyed at lower energies.[6,7] Prior to the present work, however, all experiments on the $T(d,n)He^4$ reaction have been performed by detecting the monoenergetic group of high-energy neutrons in a counter telescope.

Theoretical work on the subject reactions is limited to a "stripping" analysis of the 10-Mev data.[8]

EXPERIMENTAL DETAILS

The source of deuterons is the Los Alamos variable-energy cyclotron. A portion of the deflected beam is brought to a focus at the center of a scattering chamber. Brass diaphragms limit the maximum beam divergence to 0.5°, the greatest part of which is due to multiple small-angle scattering in the beam entrance window and target gas. The target gas is confined to the center of the scattering chamber by a hollow cylinder whose axis of symmetry is perpendicular to the plane of scattering and whose vertical wall incorporates a window of 0.0005-in. duraluminum. After traversing the target, the beam enters a well-evacuated Faraday cup which is magnetically and electrically biased to avert the capture of externally produced electrons as well as the loss of those generated inside the cup.

Nuclear emulsions mounted around the periphery of the scattering chamber record the charged particles and each detector views a precise portion of the reaction volume through its own slit system at a well-defined angle with respect to the incident beam.

Most of the experimental techniques utilized in the present investigation have been carried over from previous experiments. Thus, the ion-optical system for conducting the beam to the reaction volume, as well as the Faraday-cup construction and method of current integration, has been described in a previous paper by

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† One of the authors wishes to acknowledge the hospitality of the medium-energy nuclear physics division of le Centre d'Etudes Nucleaires de Saclay during the preparation of this manuscript.

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