fraction of the residual 30.2 is due to the presence of second-order neutrons. The oscillating field affects the second-order neutrons very little for two reasons: K will be close to one because their polarization is low (-10 percent); when the strength of the oscillating field is such that the probability of flipping first-order neutrons is a maximum, the probability of flipping second-order neutrons is considerably less than maximum. The symmetry of the intensities in cases (3) and (1) with respect to (2) indicates strongly that all first-order neutrons are being flipped. The fact that the intensities in cases (3) and (4) are equal indicates that the depolarizing shim is removing completely the polarization of the beam passing through it. This method could be used, also, if I and II were transmitting blocks of iron. The use of magnetite crystals is advantageous because the high polarization of the first order of reflection from the (220) planes gives large intensity changes, and the polarization is independent of energy.

PHYSICAL REVIEW VOLUME 94, NUMBER 2 APRIL 15, 1954

The Alpha- and Gamma-Ray Spectra of Pu^{238}†

FRANK ASARO AND I. PERLMAN
Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California
(Received January 12, 1954)

The alpha and gamma spectra of Pu^{238} have been studied with an alpha-particle spectrograph and gamma-ray scintillation and proportional counters. Alpha groups of 5.495 (72 percent), 5.452 (28 percent), and 5.352 Mev (0.09 percent) and electromagnetic radiations of 17 (13 percent), 43.8±0.5 (0.038 percent), 99±2 (0.008 percent), and 150±2 kev (0.001 percent) were observed. Spins and parities are assigned to the energy levels, and the results are evaluated with respect to the developing theory and systematics of complex alpha spectra and excited states of even-even nuclei.

I. INTRODUCTION

MARKED regularities have been observed in the alpha spectra of different *even-even* nuclei indicating strongly that corresponding spectroscopic states are involved in the decay processes. Among the heaviest nuclei the low-lying excited states are spaced in such a fashion as to suggest that they constitute a rotational band[1] interpreted according to the Bohr and Mottelson[2] theory which unifies independent particle and collective aspects of nuclear states.

In another publication[3] the details of the alpha spectrum and gamma spectrum of Cm^{242} were reported and discussed in terms of the excited states of Pu^{238}. The present communication deals with Pu^{238} and the energy levels of U^{234}. The close similarity between Cm^{242} and Pu^{238} spectra can be seen from the decay schemes shown in Fig. 1, and it will be seen that the gamma-ray conversion coefficient data also show that comparable spectroscopic states are involved.

II. METHODS

The samples of Pu^{238} used in the present measurements[4] were made by the prolonged neutron irradiation of Am^{241} through the following series of reactions:

$$Am^{241}(n,\gamma)\, Am^{242m} \xrightarrow[16\text{ hr}]{\beta} Cm^{242},$$

$$Cm^{242} \xrightarrow[162\text{ day}]{\alpha} Pu^{238}.$$

The primary objective of the irradiations was to make Pu^{242} through the electron capture branching of 16-hour Am^{242m} so sizable amounts of this isotope were present. Also in a long irradiation the Pu^{238} captures neutrons to give successively Pu^{239}, Pu^{240}, and Pu^{241}, all of which were also present in small quantities. Mass spectrometric analyses[5] were made on two of the preparations

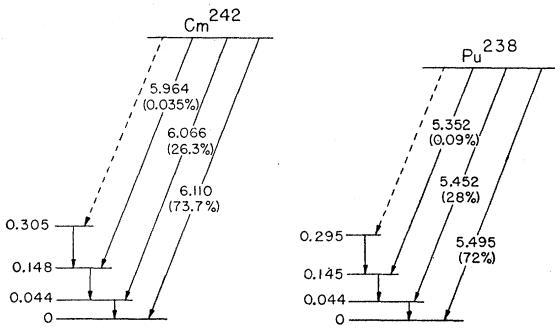

FIG. 1. Comparison of alpha spectra of Cm^{242} and Pu^{238}.

† This work was performed under the auspices of the U. S. Atomic Energy Commission.

[1] F. Asaro and I. Perlman, Phys. Rev. **91**, 763 (1953).

[2] A. Bohr and B. R. Mottelson, Phys. Rev. **89**, 316 (1953); **90**, 717 (1953); Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 16 (1953).

[3] Asaro, Thompson, and Perlman, Phys. Rev. **92**, 694 (1953).

[4] We are indebted to Dr. S. G. Thompson of this laboratory and to the personnel of the Atomic Energy of Canada, Ltd., Chalk River, Ontario, Canada, for making some of these preparations available to us.

[5] The analyses were made by Mr. F. L. Reynolds of this laboratory.

TABLE I. Mass composition of plutonium preparations.

	238	239	240	241	242
Prep. I	62.9%	11.7%	1.6%	0.07%	24%
Prep. II	1.5%	80.0%	4.2%	0.5 %	13.7%

and the results are shown in Table I. The large amount of Pu^{239} in Preparation II was the result of Pu^{239} present in the Am^{241} before irradiation. Since Pu^{238} has a half-life which is short relative to the alpha-decay half-lives of the other isotopes, the alpha activity in both preparations was predominantly that of Pu^{238}. In Preparation I the Pu^{238} comprised 99.9 percent of the alpha activity and in Preparation II it was 81 percent. However, the differences in composition were used to assign the rarer alpha groups.

A third sample (Preparation III) of Pu^{238} was not mass analyzed but should have been free of other plutonium isotopes since it came from some previously purified curium (Cm^{242}) which had decayed through approximately one half-life.

All samples were mounted by vacuum sublimation of plutonium chloride onto a platinum plate which was masked to present a band 1 in.$\times\frac{1}{8}$ in. This technique as well as the spectrograph description and operation have appeared in other reports.[6,3] As before, the alpha particles were caught on a photographic plate and the track count plotted according to position on the plate in order to reproduce the spectrum. The record of alpha-particle spectrograph exposures is given in Table II.

Gamma-ray spectra were measured for the most part with a sodium iodide scintillation counter coupled with either a single channel or multichannel pulse-height analyzer. In some experiments a xenon-filled proportional counter was used to produce the pulse for the analyzer.

III. RESULTS

A. Principal Alpha Groups of Pu^{238}

A sample of Cm^{242} which had decayed six months was run on the spectrometer (Prep. III, Exp. 73) and showed two new alpha groups at about the energy previously reported for Pu^{238} from range measurements.[7,8] The intensity of the alpha groups relative to Cm^{242} groups was 0.6 percent in agreement with expectations for the particular growth and decay period. The main alpha group of Cm^{242} was used as an energy standard (6.110 Mev),[3] and the value for the main group of Pu^{238} was found to be 5.492 Mev. The second group was of 42 kev lower energy and therefore 5.450 Mev.

TABLE II. Record of alpha-particle spectrograph exposures.

Exposure number	Plutonium preparation	Exposure duration	Defining slit
Exp. 73	Prep. III	40 hr	1 in.×0.12 in.
Exp. 204	Prep. I	114 hr	1 in.×0.12 in.
Exp. 205	Prep. I	75 min	1 in.×0.12 in.
Exp. 243	Prep. II	37 hr	1 in.×0.12 in.
Exp. 283	Prep. I	54 min	0.75 in.×0.018 in.

The curium of Prep. III could have contained Am^{241} which has a prominent alpha group at 5.476 Mev which could distort the energy determination of the Pu^{238} groups. However, it was found that the sample contained less than 2 percent by weight of Am^{241}, and its alpha group would therefore be at least 300-fold less intense than the Pu^{238} group.

The particle energy of Pu^{238} was also measured in Exp. 243, Prep. II, using the 5.150-Mev group[9] of Pu^{239} as a standard. This gave 5.497 Mev for the main group of Pu^{238} and a 42.5-kev separation for the second group. In Exp. 283 the separation between the groups was again 42.5 kev. We shall take as the best value for the main group 5.495 Mev and 5.453 Mev for the second group.

These groups presumably lead, respectively, to the ground state and first-excited state of U^{234}. Applying the recoil correction to the energy spearation we find that the first-excited state of U^{234} is 43 kev above the ground state. If we use the terminology described in previous publications the alpha group populating the ground state will be designated α_0 and that populating the 43-kev state α_{43}. The intensity of α_{43} relative to total Pu^{238} alpha particles was found to be 24 percent in Exp. 73 and 28 percent in Exp. 283.

B. Low-Intensity Alpha Groups of Pu^{238}

In Exp. 243 (Prep. II) and Exp. 204 (Prep. I) a third group of Pu^{238} was observed. The separation in particle energy between this group and α_{43} of Pu^{238} was found to be 101.5 kev in Exp. 243 and 98.6 kev in Exp. 204. The average value of 100 kev is accepted which means that the energy level of U^{234} is 102 kev above the 43-kev level and 145 kev above the ground state.

Although α_{145} is in very low intensity (0.1 percent of total Pu^{238}), it was proved to belong to Pu^{238} because of its constant relationship with the Pu^{238} alpha particles in the two preparations which differed widely in isotopic composition. This information is shown in Table III.

The exact correspondence of energy levels and alpha transitions to these levels between different even-even nuclei in this region is worthy of note. As shown in Fig. 1, the alpha spectra of Cm^{242} and Pu^{238} point to energy levels of the product nuclei spaced almost identically. Each has a transition to the ground state and to the first-excited state whose intensities can be shown to be in substantial agreement with alpha-decay theory, and each has a transition to the second-excited

[6] Asaro, Reynolds, and Perlman, Phys. Rev. **87**, 277 (1952).
[7] W. Thiel and A. H. Jaffey, reported in Argonne National Laboratory Report ANL-4370, November, 1949 (unpublished).
[8] Chamberlain, Gofman, Segrè, and Wahl, Phys. Rev. **71**, 529 (1947).
[9] F. Asaro and I. Perlman, Phys. Rev. **88**, 828 (1952).

state which is highly hindered (low intensity far beyond expectations according to the energy difference).

C. Gamma Rays of Pu^{238}

The results to be presented include the energies and intensities of the photons accompanying the alpha decay of Pu^{238}. The best picture which can be constructed from these data is that all of the transitions are of the $E2$ type and that they cascade as indicated in Fig. 1. It will be noted that the gamma ray of 150 kev is shown to come from a level which is not populated by an observable alpha group. A similar assignment was made for Cm^{242} and the reasons will be presented below.

A typical gamma-ray spectrum taken with a sodium iodide crystal spectrometer is shown in Fig. 2. The relative intensities of the photons are not directly discernible because absorbers were used to minimize the intense L x-rays (18 kev) which would otherwise obscure the 42-kev peak.

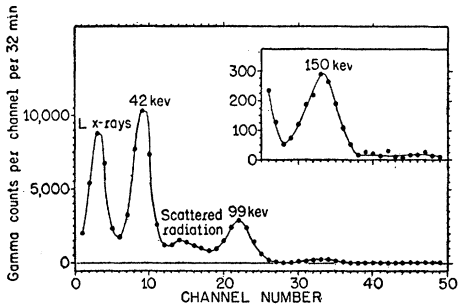

FIG. 2. Pu^{238} gamma spectrum.

In all, ten measurements of the gamma-ray spectrum were made; nine of these were done on Prep. I and for the other a different sample of Pu^{238} was used. The experiments aimed at energy determination are summarized in Table IV. The intensities will be discussed presently.

In experiments 1, 2, and 3 the voltage on the photomultiplier tube was not stable and a single channel analyzer was employed. These energy values are not considered as reliable as the others. Experiments 4, 5, and 6 were run with multichannel analyzers and under more stable conditions.

D. The 43.8-kev Gamma Ray

A xenon-filled proportional counter was used in experiments 7 and 8 and should give the most reliable values for the ~40-kev gamma ray. For these measurements the counter was standardized with the 44.3-kev gamma ray[3] of Cm^{242}. This value (44.3 kev) obtained from the energy difference of alpha groups is in good agreement with the energy obtained by measuring the

TABLE III. Ratios of intensities of $Pu^{238}\alpha_{145}$ and other alpha groups.

Intensity relative to:	Exp. 204	Exp. 243
Pu^{238} alpha activity	0.09_2%	0.09_9%
Pu^{239} alpha activity	130%	0.5%
Pu^{240} alpha activity	265%	2.6%
Pu^{241} beta activity	13%	0.05%
Pu^{242} alpha activity	1300%	61%

photon in a proportional counter (44.6 kev),[10] from the conversion electron spectrum (44.1 kev),[11] and from the conversion electron spectrum in the decay of Np^{238} (44.1 kev).[12] The gamma-ray energy for Pu^{238} so obtained was 43.8±0.5 kev.

The intensity of this gamma ray relative to total alpha particles of Pu^{238} was determined by Reed[13] using a xenon-filled Geiger tube and absorbers. He found the intensity to be 4.2×10^{-3}. In the present study (experiment 2) the intensity was measured with the scintillation spectrometer using the 60-kev gamma ray of Am^{241} as a standard. The intensity of the Am^{241} gamma ray is 0.40 per disintegration as measured by Beling, Newton, and Rose.[14] The intensity of the gamma ray so obtained was 3.8×10^{-4} which is considerably lower than Reed's measurement and should be more accurate because of the inherent better precision in the method used.

There can be no question that this gamma ray results from the de-excitation of the state reached by Pu^{238} α_{43} (see Fig. 1.) Since α_{43} populates this state to the extent of 28 percent, the low photon intensity would imply a large conversion coefficient. O'Kelley[15] has observed in good intensity a series of conversion lines attributable to a transition of ~45 kev and Dunlavey and Seaborg[16] found by the photographic plate technique that 23 percent of Pu^{238} alpha particles are in coincidence with electrons corresponding to an ~40-kev gamma ray. Within experimental uncertainties this number (23 percent) is the same as the intensity of α_{43} (28 percent)

TABLE IV. Energies of Pu^{238} electromagnetic radiations.

Experiment No.										Best
1	2	3	4	5	6	7	8	9	10	value
157[a]	...	145	146	148	150±2	...	...	...	151	150
107	102	99	101	96	99±2	...	...	...	97	99
45	42	...	...	...	42±2	43±1	43.8±0.5	...	41	43.8
17	18	...	...	...	18	...	...	17±0.5	...	17

[a] Energies in kev.

[10] B. Rose and J. O. Newton, reported at Birmingham Conference on Nuclear Physics, July, 1953 (unpublished).
[11] T. O. Passell (unpublished).
[12] H. Slätis and J. O. Rasmussen, Jr. (to be published).
[13] G. W. Reed, Jr., U. S. Atomic Energy Commission Declassified Report AECD-3185, January, 1951 (unpublished).
[14] Beling, Newton, and Rose, Phys. Rev. **86**, 797 (1952); erratum: Phys. Rev. **87**, 1144 (1952).
[15] G. D. O'Kelley, Ph.D. thesis, University of California Radiation Laboratory Report UCRL-1243, March 15, 1951 (unpublished).
[16] D. C. Dunlavey and G. T. Seaborg, Phys. Rev. **87**, 165 (1952).

indicating a large conversion coefficient. With 28 percent as the population of the 44-kev state and 0.038 percent for the photon intensity, the total conversion coefficient is 740. In order to determine the L-conversion coefficient one must know the relative amounts of conversion in the L and higher-order shells. For a similar transition in the decay of Cm^{242} it was found that 80 percent of the conversion electrons were from the L shell and 20 percent from M and higher shells. Applying these values to the Pu^{238} case, the L-shell conversion coefficient becomes 600. This can be compared with theoretical calculations of Gellman, Griffith, and Stanley[17] from which one would expect values of 1, 60, and 700 for $E1$, $M1$, and $E2$ transitions, respectively. The transition is obviously of $E2$ type on this basis and this is added evidence that the first-excited state of an even-even nucleus has spin 2 and even parity.

E. L X-Rays

Photons of 17–18 kev were the most prominent electromagnetic radiation in the decay of Pu^{238}. The best energy value was 17 ± 0.5 kev determined in experiment 9 (Table IV) with the scintillation spectrometer and a motor-driven single channel analyzer. These are undoubtedly uranium L x-rays which arise almost entirely from the internal conversion of the 43.8-kev gamma ray.

The intensity of this L x-ray complex was obtained by comparison with the L x-rays of Am^{241} decay, the intensity of which was determined by Beling, Newton, and Rose.[14] The value so obtained was 13 percent (Table V) and this may be compared with the expectations from the internal conversion of the 43.8-kev gamma ray.

Since the 43.8-kev gamma ray is about 80 percent converted in the L shell (see previous discussion) there should be 0.22 L vacancies per alpha disintegration. The calculations of Gellman, Griffith, and Stanley[17] show that an $E2$ transition such as this is converted almost entirely in the L_{II} and L_{III} shells and Kinsey[18] has estimated that the fluorescent yield for these L vacancies is ~0.5. The expected number of L x-rays is then 11 percent (0.5×22) which is in satisfactory agreement with the observed intensity, 13 percent.

TABLE V. Intensities of Pu^{238} electromagnetic radiations.

Energy (kev)	Absolute intensity per alpha disintegration	Intensity relative to 43.8-kev radiation	Intensity relative to 99-kev radiation	Experiment number	Best value for absolute intensity per alpha disintegration
17	13%			9	13%
43.8	0.038%			2	0.038%
99		22%		2	
					0.0080%
		20%		10	
150			11.8%	1	
			12.8%	4	
					0.0010%
			12.6%	5	
			13%	10	

[17] Gellman, Griffith and Stanley, Phys. Rev. **85**, 944 (1952).
[18] B. B. Kinsey, Can. J. Research. **A26**, 404 (1948).

F. The 99-kev Gamma Ray

As indicated in Fig. 1, this gamma ray has been placed between the states populated by α_{145} and α_{44}. It will be noted that a gamma ray of 150 kev was also observed and it might be assumed that this is the crossover transition from the state reached by α_{145} to the ground state and that the 99-kev photon is the K x-ray resulting from internal conversion of the 150-kev gamma ray. From the relative intensities of the 99-kev and 150-kev photons the K-conversion coefficient would have to be 8. Furthermore, all transitions to the ground state of an even-even nucleus from levels populated by alpha decay must be electric. From the data on K-shell conversion coefficients by Rose and co-workers[19] and by Spinrad and Keller,[20] an electric transition of 150 kev with conversion coefficient 8 would have to have a multipolarity of at least 5 and would therefore have an extremely long lifetime. It may be concluded therefore that all or nearly all of the 99-kev photons are gamma rays and the transition is placed best between the levels reached by α_{145} and α_{44}.

The total conversion coefficient of the 99-kev transition can be calculated from the measured intensity of the gamma ray and the population of the 145-kev state. With 0.09_6 percent as the population of the 145-kev state and 8×10^{-3} percent (Table V) as the gamma-ray intensity, the conversion coefficient is 11. The amount of internal conversion other than in the L shell is probably small so that this figure may be taken as approximately the L-conversion coefficient. From the tables of Gellman, Griffith, and Stanley[17] the theoretical values for $E1$, $E2$, and $M1$ transitions are respectively 0.1, 8, and 4. The $E1$ transition is ruled out, and, although $E2$ gives a better fit than $M1$, a choice probably should not be made on this ground alone. Since the 44-kev state has even parity (2+), the 145-kev state must therefore also have even parity. Only even spin numbers are allowed since the level is reached by alpha decay of an even-even nucleus. The possible states are therefore 0+, 2+, and 4+, and of these the selection of 4+ is favored for reasons to be given below.

G. The 150-kev Gamma Ray

The low intensity photon of 150 kev (see Fig. 1 and Tables IV and V) has already been mentioned in the preceding section where it was shown that it cannot account for much of the de-excitation of the 145-kev state. Gamma-gamma coincidence measurements showed this gamma ray to be in coincidence with the

[19] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951); Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report ORNL-1023, June, 1951 (unpublished).
[20] B. I. Spinrad and L. B. Keller, Phys. Rev. **84**, 1056 (1951).

99-kev photon. Furthermore, the coincidence rate was in agreement with the assignment of the 99-kev photon to a gamma ray with conversion coefficient ~10.

As will be borne out in the discussion, it is reasonable to assign the 150-kev transition to another of the series of cascading $E2$ transitions (see Fig. 1). On this basis, its conversion coefficient would be 2 and the population of the 295-kev level would be 3×10^{-3} percent. This intensity was below the limits of our detection of an alpha group which for this case was $\sim10^{-2}$ percent, so the failure to observe the corresponding alpha group is not inconsistent with the decay scheme.

IV. DISCUSSION

The energy levels of U^{234} observed from the alpha decay of Pu^{238} conform in remarkable fashion with the picture for even-even nuclei well removed from a closed shell developed by Bohr and Mottelson.[2] In this model collective aspects of nuclear structure and individual particle aspects are coupled and in a region well removed from a closed shell there should be a series of energy levels corresponding to a rotational band in which only even spin states (0+, 2+, 4+, ···) appear. On this basis electric quadrupole transitions should predominate. It was seen that the 44-kev transition definitely falls into this category and the 99-kev transition probably does so. A unique assignment could not be made for the 150-kev gamma ray because the conversion coefficient could not be measured, but the failure to observe an alpha particle leading to the 295-kev state means that the conversion coefficient is not an order of magnitude greater than unity and is therefore in the proper range for an $E2$ transition.

Another aspect of the Bohr and Mottelson model concerns the energy level spacings of the states 0, 2, 4, ···. The spectrum is that of a rigid rotator although it should be pointed out that in the treatment the moment of inertia is not that of a rigid body but is an effective moment associated with the angular momentum of the surface waves. The rotational energy levels go as $I(I+1)$, where I is the spin number so the energy levels go as 1, 3.3, 7.0 for the first three excited states of spin 2, 4, 6. In this case, the ratio between the second- and first-excited states (145/44) is 3.3 and between the third and first (295/44) is 6.7. This striking agreement with the theory is the principal reason for assigning the second- and third-excited states 4+ and 6+, respectively, in the absence of independent means of making assignments.

We wish to acknowledge the assistance of Mrs. J. A. Simmons in counting the alpha tracks.

PHYSICAL REVIEW VOLUME 94, NUMBER 2 APRIL 15, 1954

Measurement of the Tritium Concentration in Natural Waters by a Diffusion Cloud Chamber*

E. L. FIREMAN AND D. SCHWARZER
Brookhaven National Laboratory, Upton, New York

(Received January 8, 1954)

The radioactivity of tritium is easily recognized and visually counted in a hydrogen-filled diffusion chamber. A 6-in. diameter diffusion chamber, filled with 100 psi of hydrogen obtained by completely converting 14 cc of water in a Mg furnace, is used to measure the tritium concentration in surface water of the Long Island Sound, in New Jersey well water, and in a number of rain and atmospheric moisture samples. These samples had tritium concentration (T/H ratios) that ranged from 10^{-15} to 3×10^{-18}. Some samples were measured directly, others required some electrolysis to give measurable tritium concentrations. No tritium could be detected in glacial water.

INTRODUCTION

THE existence of natural tritium in Norwegian lake water has been demonstrated.[1] This was accomplished by enriching the natural tritium content by electrolysis between 1 and 10 million fold. A series of measurements on natural tritium in rain and ground water is being carried out at the University of Chicago[2] with a low-level Geiger counter and an electrolytic plant. We have applied a diffusion chamber technique to the same problem. This technique is more sensitive than a low-level Geiger counter[3] that we have been using here. Therefore, considerably less enrichment is required to measure the tritium in natural waters. In fact, in two rain samples and one atmospheric moisture sample the tritium could be measured directly without any enrichment.

DESCRIPTION OF APPARATUS

Figure 1 is a schematic diagram of the diffusion chamber and its associated hydrogen-converting ap-

* Research performed under the auspices of the U. S. Atomic Energy Commission.

[1] Grosse, Johnston, Wolfgang, and Libby, Science **113**, 1 (1951).
[2] W. F. Libby, Phys. Rev. **93**, 1337 (1954); Proc. Natl. Acad. Sci. **39**, 245 (1953).
[3] E. L. Fireman, Phys. Rev. **91**, 922 (1953).