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THE STRUCTURE OF HEAVY NUCLEI: A STUDY OF VERY WEAK ALPHA BRANCHING

Berkeley, California

UCRL-11028
UC-4 Chemistry
TID-4500 (24th Ed.)

UNIVERSITY OF CALIFORNIA

Lawrence Radiation Laboratory
Berkeley, California

Contract No. W-7405-eng-48

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Charles Michael Lederer

(Ph. D. Thesis)

September 27, 1963

Printed in USA. Price $2.00. Available from the
Office of Technical Services
U. S. Department of Commerce
Washington 25, D.C.

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ABSTRACT

Very weak alpha branching in heavy elements was studied by a recently developed coincidence technique. This technique makes it possible to measure the energies and intensities of both α-particle groups and de-exciting radiation, even when the transition intensities are as low as 10^{-8} relative to the most intense alpha group. Twenty α-particle emitters from Po^{214} to Fm^{254} have been examined.

00+ states ("beta vibrations") were observed in six even-even nuclei, and analogous states were found in three odd-mass nuclei. They are in general characterized by low alpha-decay hindrance factors and roughly equal de-excitation by electric monopole and quadrupole transitions. However, the de-excitation of these states is in disagreement with vibrational model predictions in certain cases; more important, the de-excitation and other properties of the states exhibit some irregular variations from nucleus to nucleus which are evidence for some particle character in the states.

Information was also obtained about some other types of levels. A number of 1- states ("octupole vibrations") were observed, and a possible 2- state was observed in U^{236}. A state that appears to be analogous to the 1- octupole states of even-even nuclei was observed in U^{235}. In Pu^{239}, a K = 3/2+[631] band was identified with reasonable certainty, and numerous particle states were observed in Bk^{249}, although it was not possible to classify them. No 22+ states ("gamma vibrations") were observed, and it appears that the alpha transitions to these states are rather highly retarded.

I. INTRODUCTION

A recently developed coincidence technique makes it possible to study very weak transitions occurring in alpha decay. The method has been used to study excited levels up to about 1 MeV in heavy nuclei. In the experiments reported in this paper, energy levels of nuclei from Pb^{210} to Cf^{250} have been examined.

Much of what is known about nuclear level structure can be explained in terms of semi-independent nucleons in a central field. In recent years this model has been extended to include spheroidally deformed fields.[1] The level structure of deformed, odd-mass nuclides in the heavy-element region has been successfully interpreted in this way.[2] The band structure which also occurs in the heavy mass region is well characterized as arising from collective surface-wave rotation of the deformed nucleus.

However, a number of states in even-even nuclides cannot be described satisfactorily as particle excitations or as rotations. Like rotations, these have been characterized as collective states because:

1. Their E2 and E3 transition probabilities, as measured in Coulomb excitation, are enhanced beyond single-particle estimates.
2. The states occur below the energy gap defined by the odd-even mass difference.
3. States with the same spin and parity occur somewhat systematically in many nuclei.

More detailed explanations of these states have met with some success. In the two major regions of deformation, the states have been described as surface-wave vibrations of an axially symmetric ellipsoid.[3] The 00+, 22+, and 01- states that occur in even-even nuclides are characterized as beta vibrations, gamma vibrations, and octupole vibrations. (The quantum numbers $KI\pi$ are used throughout the paper to denote states of deformed nuclei. Single-particle levels of odd-mass nuclides are sometimes designated by the additional Nilsson quantum numbers $[Nn_z\Lambda]$. Excited 00+ and 02+ states are

are distinguished from members of the ground-state band by the context in which they appear.) Coulomb excitation studies show that the E3 transition probabilities between the ground-state band and the K = 0- bands are enhanced relative to single-particle estimates. E2 transition probabilities between the ground state and the K = 0+ and K = 2+ bands are likewise enhanced. The K = 0+ bands decay by strong monopole transitions. All these facts are in accord with the vibrational model. On the other hand, there are good reasons for believing that the vibrational description of these states is inadequate. The excitation energies are comparable to the energies of particle excitations, so one would not expect the vibrations to be adiabatic. The E2 transition probabilities are on the order of only a few single-particle units, compared with hundreds for rotational-band E2 transitions.

More detailed experimental information about "collective" states of heavy elements is difficult to obtain, because there is no simple, generally applicable way to excite them. Methods such as Coulomb excitation and nuclear reactions are unsuitable because of the short lifetimes of most heavy nuclides and hence the general absence of suitable target materials. Beta-decay studies are limited to cases in which a suitable beta-decay parent exists, the decay energy is sufficiently high to excite the states, and spin changes are small enough to allow substantial population of them.

Alpha decay possesses many advantages. A large number of the heavy elements are alpha emitters with suitable half lives. Alpha-particle energies fix the energy of an excited state unambiguously if the isotope assignment is certain. The chief disadvantage of alpha decay is the strong energy dependence, which results in very weak population of highly excited states. For example, unhindered transitions to states at about 1 MeV have intensities on the order of 10^{-5} to 10^{-8} relative to the favored transition. (An unhindered transition is defined as one for which the reduced transition probability is the same as for the transition to the ground state of an even-even nucleus. Reduced transition probabilities used to calculate hindrance factors quoted in this paper are calculated from the one-body-model alpha-decay equations of Preston.[4] A simpler formula by Fröman,[5] which yields approximately the same numerical results, was used for even-even nuclei.)

The experiments reported here were designed to measure such weak transitions. The primary experimental tool is a silicon semiconductor charged-particle detector. In order to observe very weak alpha branching, it is necessary to observe a large number of decay events in a reasonable time; the ability to count α particles at rates over 10^5 per second with good energy resolution is an essential property of these detectors.

Even so, very weak alpha groups are obscured in directly determined spectra by the low-energy tail of the intense peaks from the main transitions. However, the decay to higher excited states occurs in coincidence with high-energy γ rays and conversion electrons, and is therefore distinguishable in coincidence experiments. This type of experiment can also yield data on γ-ray branching and conversion coefficients that are very helpful in characterizing states.

Another problem that arises in measurements of very weak transitions is the possibility that they are due to small impurities in the active sample. Simultaneous measurement of alpha and gamma or electron energies in most cases gives unambiguous proof of correct isotopic assignment.

Very weak alpha branching has previously been inferred from the measurement of γ rays and electron-K x-ray coincidences. Such studies are possible for a few alpha emitters which can be obtained in sufficient quantity with high isotopic and chemical purity. They can be done with intense thick sources because the α particles themselves are not analyzed. In one favorable case, Cm^{244} decay, direct measurement of two alpha groups populating 00+ and 02+ states around 870 keV was made in a magnetic spectrograph,[6] although their total intensity was only 2×10^{-6}. The 01- states have been observed in a number of cases by measuring the corresponding alpha groups, because the states lay at relatively low energies.

The decay of Pu^{238} and Cm^{242} populates 00+ states characterized by low alpha-decay hindrance factors[7,8] and strong monopole de-excitation. Such states are designated "beta vibrations" in the remainder of the paper, although this is not meant to imply that their detailed properties correspond to the simple vibrational description. States analogous to the "collective" levels of even-even nuclides are also expected to occur in nuclei with unpaired nucleons among more numerous single-particle states. The association of beta vibrations with low hindrance factors and strong monopole transitions offers a possible means to distinguish them from single-particle levels in odd-mass species. Gamma vibrations have recently been identified in Coulomb-excitation experiments on some odd-mass rare earth isotopes;[9] they are distinguished in this case by their high E2 transition probabilities.

II. EXPERIMENTAL PROCEDURE

A. Apparatus for Measuring Alpha-Gamma and Alpha-Electron Coincidences

The original coincidence counting system, conceived and designed in 1960-1961 by Sven Bjørnholm, was used in a majority of the experiments, and has proved highly versatile and reliable. Figure 1 shows the vacuum chamber that houses the source and detectors. In (a), the chamber is assembled for measuring coincidences between α particles and γ rays. The gamma detector is a 3×3-inch NaI scintillator commercially mounted with a photomultiplier (Harshaw). The efficiency of the gamma detector was calibrated with standards whose absolute intensity was known to ±5%: Na^{22}(511 and 1276 keV), Cs^{137} (662 keV), and Co^{60}(1173 and 1333 keV). A set of calibrated absorbers could be inserted in the slots between the source and the scintillator. These are different thicknesses of lead, cadmium, and copper.

For measuring alpha-electron coincidences, an anthracene scintillator electron detector extended into the chamber through a hole in the wall, Fig. 1(b). Optical contact to a photomultiplier was obtained by a short light guide, and the face of the photomultiplier sealed vacuum-tight against a rubber gasket. The solid angle of the detector was measured with a calibrated Cs^{137} source and found to be 33% of 4π. Calibrated aluminum absorbers could be used to absorb lower-energy electrons.

The alpha detector is a phosphorus-diffused p-n junction silicon semiconductor with a guard ring.[10] The sensitive areas of different detectors ranged from 0.5 to 2.0 cm in diameter. Many such detectors were used; their excellent quality was an important factor in the success of the experiments. The resolution, lifetime, and availability of the detectors were improved considerably during the course of this work. Surface barrier type detectors were also used in a few experiments; their performance characteristics were similar.

The electronic circuit is shown in a block diagram, Fig. 2. The amplifiers for the scintillation counters, fast coincidence unit, and

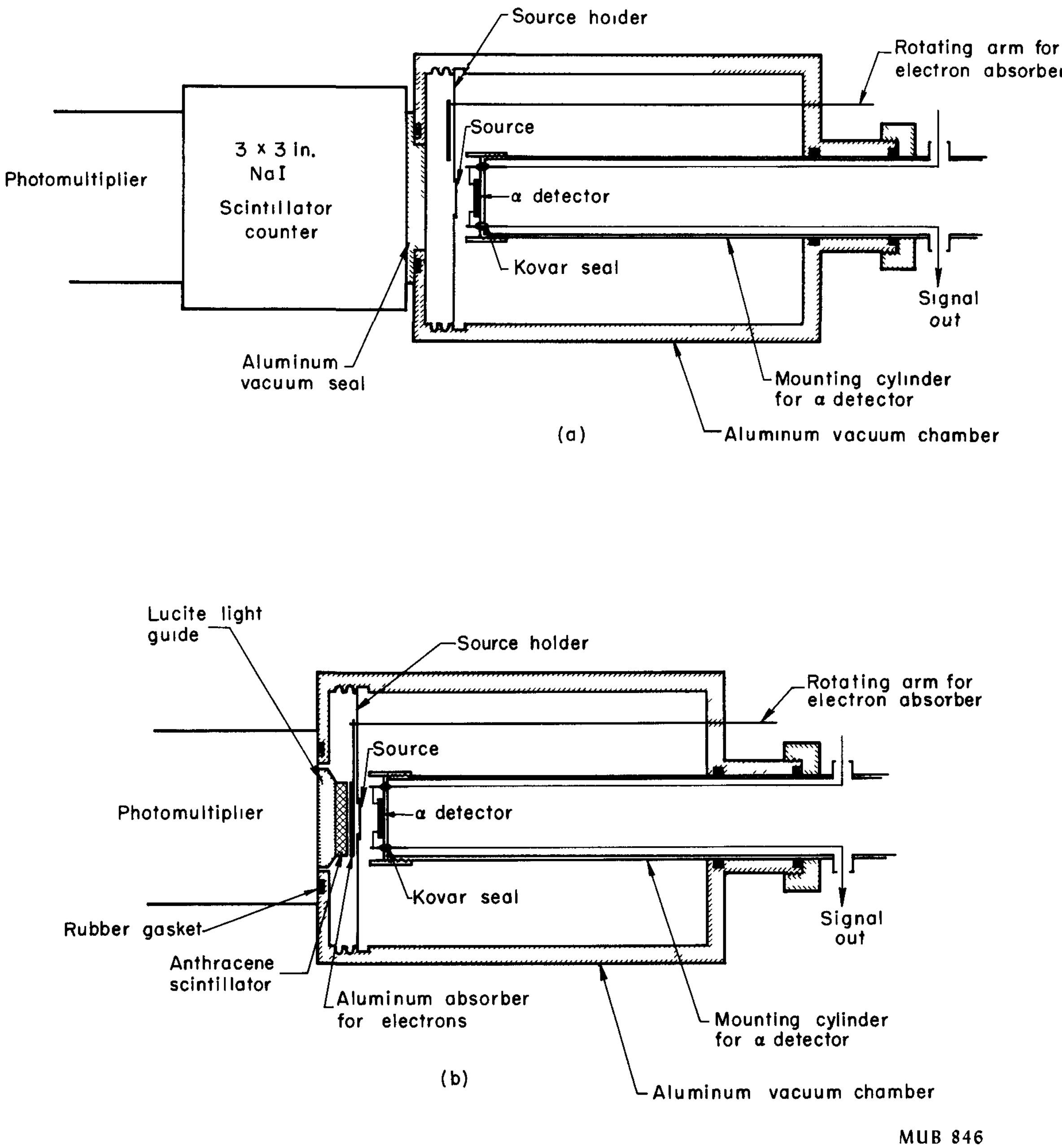

Fig. 1. Source-detector housing (a) for α-γ coincidence measurements, and (b) for measuring α-e⁻ coincidences. The position of the α-particle detector can be varied relative to the source. Absorbers can be inserted between the source and the scintillation detector. The electron absorber, shown in position in (b), was used to correct for response of the anthracene scintillator to γ rays. In a normal α-e⁻ measurement it is moved away.

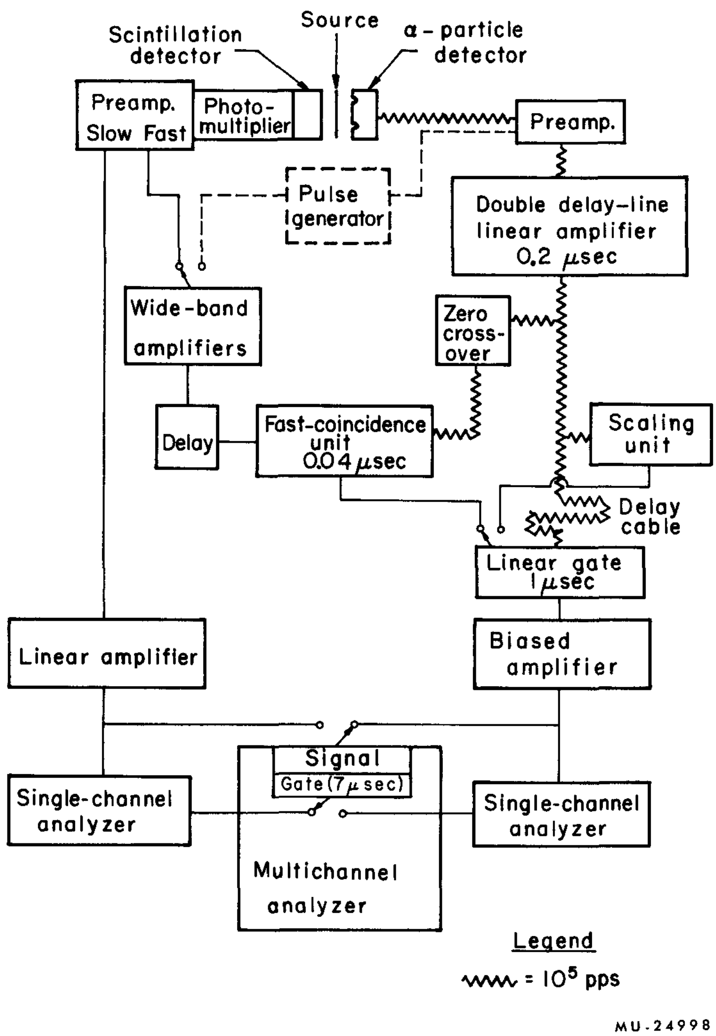

Fig. 2. Simplified block diagram of the electronic circuit. Wavy lines indicate portions of the circuit in which the count rate is high.

single-channel and multichannel analyzers are of conventional design. The preamplifier for the semiconductor detector was designed by Goulding,[10] and the linear amplifier by Landis and Goulding. The zero crossover pickoff[11] extracts a time signal from the alpha pulse which is matched to the fast signal from the wide-band amplifiers by a continuously variable delay. The output of the linear amplifier feeds the linear gate, and three means for opening the gate were available. Normally the gate opens whenever a fast coincidence has occurred. For alpha singles measurements, the gate could be opened by the output of the scale-down unit, permitting every pulse, or every 10th, 100th, 1000th, or 10,000th pulse to pass. This reduced the count rate in the biased amplifier and analyzers. A pulser fed the first stage of the preamplifier, and produced a synchronous pulse to trigger the fast coincidence. It was thus possible to measure singles alpha, coincident alpha, and pulser spectra without changing the counting rate at the detector. This prevented amplitude shifts that would otherwise occur at such high count rates.

The chief experimental problem is the compromise between good resolution and high count rates. Silicon detectors, because they generate a higher charge per unit energy in a much shorter time, are greatly superior to the earlier gas ionization chambers in both resolution and ability to handle high count rates. Nevertheless, high repetition rates cause serious problems unless the entire detector-preamplifier-amplifier-gate system is specially designed to prevent them.

The normal 20-μsec decay time of the preamplifier output pulse resulted in severe pile-up overload of later stages, and was accordingly lowered to 0.45 μsec for most experiments. The linear amplifier provided single-delay-line, double-delay-line, or RC pulse shaping. For maximum count rates, up to 2×10^5/sec, double delay line (DDL) shaping was necessary, although the peak width at low count rates was greater than for RC shaping by a factor of 1.5.[12, 13] The DDL pulses (Fig. 3a) decay quickly and cleanly to zero amplitude, which results in minimum spectral tailing. Also, their time integral is nearly zero, which prevents voltage level shifting from the accumulation of pulses

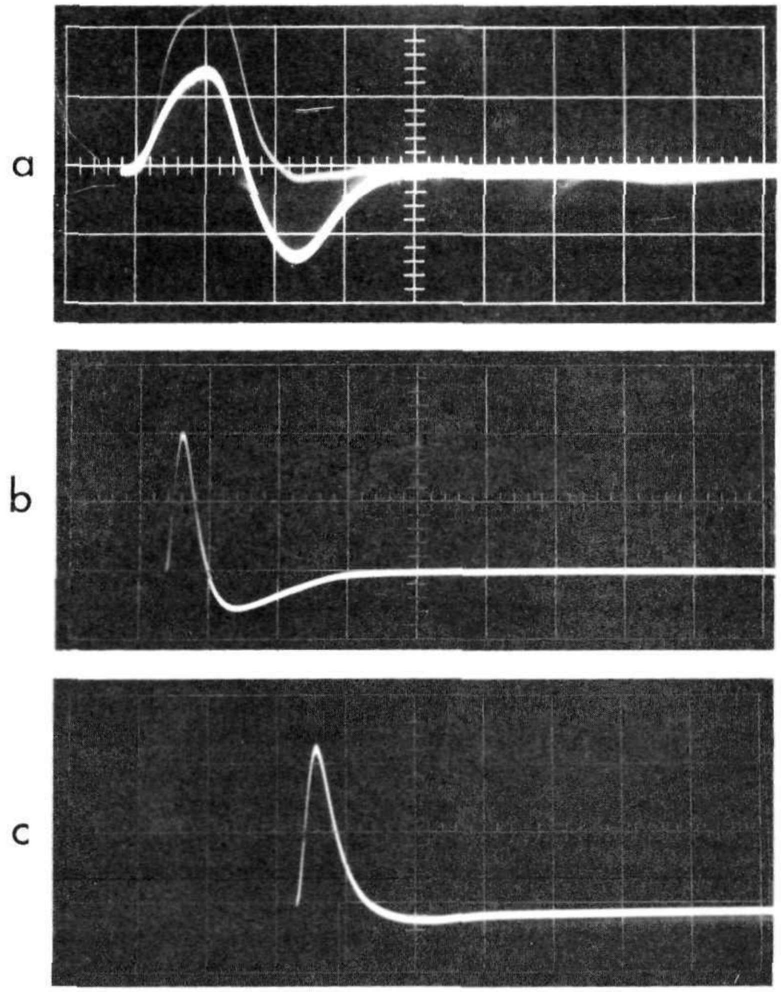

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Fig. 3. Oscilloscope traces of shaped alpha pulses.
(a) Double delay line (DDL) shaping, 0.2 μsec per horizontal division, 2 V per vertical division. The lighter trace is caused by fission pulses from the Cf^{252} source.
(b) RC shaping. Time constants: preamplifier 0.45 μsec, differentiator 0.2 μsec, integrator, 0.1 μsec. Scale: 1 μsec and 2 V per division.
(c) RC shaping. As in (b), but the pre-amplifier time constant has been raised to 2 μsec.

in later stages. Their baseline crossover point (zero voltage) provides a time marker that is independent of the pulse amplitude.

When RC shaping is used with the shortened preamplifier pulse, a double differentiated pulse results (Fig. 3b). This pulse has a nearly zero time integral, and crosses the baseline at a time nearly independent of pulse amplitude. The resolution is considerably better than with DDL shaping at count rates up to 5×10^4/sec.

For better resolution, it is necessary to use a longer preamplifier pulse and RC shaping. The resulting pulse (Fig. 3c) is only slightly double-differentiated, so that the crossover point depends more strongly on pulse amplitude, making it necessary to lengthen the coincidence resolving time by a factor of 5 to 10. Resolution is improved somewhat, but severe pile-ups occur at count rates above 10^4/sec, so that this type of pulse shaping found much smaller use. (See Pu^{236} results.)

The optimum resolution obtained was 0.46%; typically resolution fell in the range 0.5% to 1.3%, depending on the count rate and the detector used. The system has proved reasonably stable, normally drifting less than 0.1% during a 24-hour measurement.

B. Improvements in the Coincidence Counting Technique

Fission Anticoincidence Counter

A few of the heaviest nuclides studied, particularly Cf^{252}, decay to a substantial extent by spontaneous fission, producing high-energy fragments in coincidence with abundant γ rays. Large pulses, produced when a fission fragment strikes the alpha detector, saturate the amplifier, and consequently have a crossover point that occurs too late to give a true coincidence (see Fig. 3b). However, small pulses from the tailing of the fission spectrum will be in true coincidence with fission γ rays, and this will produce a background that can obscure weak alpha groups. Also, ternary fissions, occurring in about 0.2% of the fission events, produce α particles that give rise to additional background.

Since at least two high-energy fragments are produced in every fission, it is possible to eliminate most of the fission background by detecting fission fragments in a separate counter connected to an anti-coincidence circuit. This counter must have nearly 2π geometry in order to detect at least one fragment from a ternary event. This was accomplished by depositing the active source directly on the surface of the anticoincidence detector.

The anticoincidence circuit is shown in Fig. 4. The 10-Mc discriminator rejects the more abundant low-energy alpha pulses. The rest of the circuit is included to produce the proper pulse and dc voltage to operate the blocking input on the multichannel analyzer.

Improved Electron Resolution

In the course of the experiments it became desirable to resolve electron lines in coincidence with α particles. The anthracene detector suffers from inherently poor resolution; semiconductors are capable of better resolution, but it is very difficult to produce a diffused-junction detector with a depletion layer sufficiently thick to stop energetic electrons. With the advent of lithium-drifted silicon detectors, depletion layers thick enough to stop 1-MeV electrons have recently become available. Typical resolution is 30 keV, as compared with $\approx$ 150 keV for anthracene scintillators.

For alpha-electron coincidence measurements that employed these new electron detectors, as well as for measurements employing the fission anticoincidence system, a new vacuum chamber was designed. A new coincidence amplifier unit, designed by Goulding and Landis, was also used. This unit contains a complete coincidence system in a single unit, including two amplifiers and zero crossovers, fast and slow coincidence units, single-channel analyzer, linear gate, and biased amplifier. The entire design represents a considerable improvement over the earlier equipment. Even higher count rates can be accommodated without loss of resolution, and stability and lifetime of the components have been greatly increased.

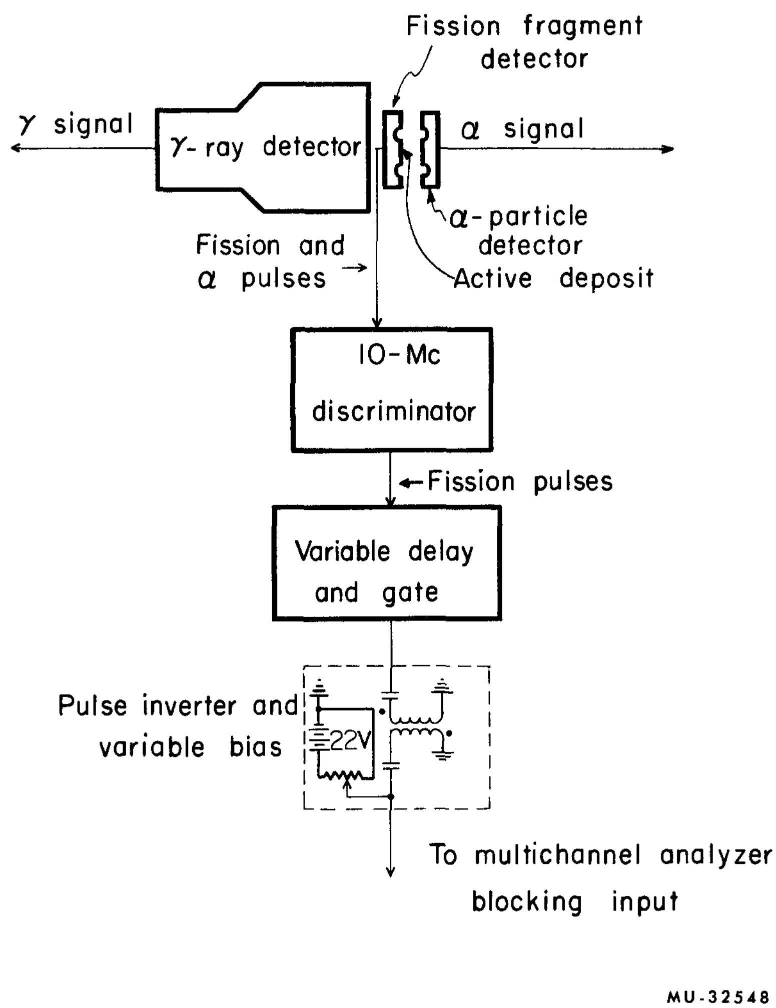


Fig. 4. Fission anticoincidence circuit and source-detector arrangement.

With the new system and a good lithium-drifted detector, electron resolution of 25 keV (almost independent of electron energy) was obtained. Resolution during coincidence experiments was 42 keV, because it was necessary to use a source backing thick enough to stop α particles from reaching the detector. The solid angle of the detector in these experiments was 15% of 4π.

C. Equipment for Gamma-Ray Measurements

Gamma spectra were measured with a 3×3-inch NaI scintillator coupled to standard pulse-height measuring equipment. Gamma-gamma coincidences were measured with two 3×3-inch NaI detectors coupled to a standard fast-slow coincidence circuit. The resolving time of the fast circuit was 0.045 μsec for U^{232} studies, 0.060 μsec for the Cm^{244} studies.

D. Source Preparation

A majority of the alpha emitters employed were available and required little or no purification. Pu^{236} was produced by Frank Asaro in a prolonged bombardment of U^{235} with α particles in the 60-inch cyclotron. Cm^{242} containing negligible Cm^{243} was prepared in a short neutron irradiation of Am^{241}. U^{230} was prepared by V. Subramanyam, as a byproduct of protactinium isotopes he was studying, by bombarding Th^{232} with protons in the 184-inch cyclotron.

The heaviest isotopes were made available from a campaign in which decigram quantities of Cm^{244} were irradiated for several years in the MTR, and the resulting actinides separated. Cf^{252} and E^{253} were available directly from this program. In order to produce Fm^{254} and Cf^{250}, 45 nanograms (2.6×10^9 alpha disintegrations per minute) of E^{253} was carefully purified from californium isotopes and then irradiated for 6 days in the MTR at a flux of 2×10^{14} neutrons/cm^2/sec. Fm^{254} and Cf^{250} are produced by the reactions

$$E^{253} + n \rightarrow E^{254} \xrightarrow[1.5d]{\beta^-} Fm^{254} \xrightarrow[3.3hr]{\alpha} Cf^{250} .$$

Following irradiation, a californium fraction was separated and the einsteinium fraction was milked several times for Fm^{254}.

Radiochemical techniques used for separation and purification of the samples are standard, and are not discussed here. Thin sources were prepared by one of two methods. Substances of low specific activity were vacuum sublimed as oxides or chlorides from a tungsten filament. The source backing was 5- or 10-mil polystyrene, except for a U^{234} source, which was already available on nickel foil backing. Polystyrene was used because it causes a minimum loss of electrons by backscattering. Shorter-lived activities were deposited from solution into a thin cation-exchange layer (10 to 15 $\mu g/cm^2$) prepared by controlled sulfonation of the polystyrene surface with fuming sulfuric acid.[14]

Special care had to be exercised with U^{232} sources, because the growth of daughter activites interferes with spectral measurements. Alpha-gamma and alpha-electron coincidences were studied within 24 hours after purification from daughter activites by anion exchange. Gamma-gamma studies were done within 6 hours after purification. In order to prevent contamination of the vacuum chamber with the 2-year Th^{228} daughter, the vacuum-sublimed alpha sources were covered with a film of VYNS resin[15] thick enough to stop the alpha decay recoils (10 to 20 $\mu g/cm^2$) without seriously affecting α-particle resolution.

III. PROCEDURE

Singles α-particle and γ-ray or electron spectra were measured first to establish the strength and composition of the source. Delays were adjusted, and the coincidence measurement was then run, usually for 24 hours or more. Singles spectra and delay settings were checked at intervals during longer runs to insure adequate stability.

Gamma-ray energy calibrations were made with standard sources. For the anthracene electron detector, energy calibrations were made with a single source (Cs^{137}, $E_{e^-} = 624$ keV); the resolution of this detector did not warrant better calibration.

Alpha-particle energy calibration was done with the source under investigation, because switching to a standard would change the count rate and cause peak shifting to occur. Unfortunately most alpha emitters have only one or two intense alpha groups. This difficulty was overcome by taking pulser spectra with the alpha source in place. By gating the fast coincidence with the synchronous pulse from the pulser, it was possible to measure the pulser spectrum without introducing a substantial increase in the count rate. The pulser had been shown to be precisely linear, so that a constant ratio of alpha energy to pulser voltage could be assumed. This ratio was determined with the main alpha group from the source. A similar procedure was used for electron energy calibration when the lithium-drifted detector was used.

The alpha energy calibration could be checked in a number of cases in which excited-state energies were known more accurately from other measurements. Under circumstances of favorable resolution and counting statistics, the agreement was usually good to about 5 keV.

The intensity of a transition observed in the coincidence measurements is easily calculated:

$$\text{Intensity} = \frac{\text{Number of coincidences observed} \times \text{correction factors}}{\text{Number of alpha singles counts accumulated}} .$$

The number of alpha singles counts accumulated is calculated from the singles alpha spectrum and the length of the coincidence measurement.

Correction factors are for the efficiency and solid angle of the γ ray or electron detector, the efficiency of the fast coincidence unit, and a small correction for pile-up effects.

IV. RESULTS

Coincidence studies of eight alpha emitters have previously been reported.[16] Little or no additional work has been done on U^{234}, Pu^{239}, or Cm^{244}; the results are, however, included here in brief. Results reported for Pu^{238}, Cm^{242}, Cf^{252}, E^{253}, and Fm^{254} include the earlier data plus substantial new information. In addition, this paper includes the results for 12 other alpha emitters that have been examined.

Energies and intensities of all transitions quoted in the text are from these measurements, except where otherwise stated. Excited-state energies given in the decay schemes are sometimes more accurate values when available from other measurements. This is indicated by the absence of error limits. No errors are quoted for the intensities of the transitions observed in Pu^{238} decay, because these were used as secondary standards.

A. $Pu^{238} \xrightarrow{\alpha} U^{234}$

Levels of U^{234} previously reported in the beta decay of Np^{234} (reference 17) and the two isomers of Pa^{234} (references 18, 19) include states at 795 keV (01-), 810 keV (00+), 853 keV (02+), 922 keV (22+), and 1046 keV (00+). As mentioned in the introduction, the alpha decay of Pu^{238} is known from gamma singles[7] and electron-K x-ray coincidence[7,8] spectra to populate the 00+ level at 810 keV.

Figure 5(a) shows the spectrum of alpha particles in coincidence with γ rays of energy greater than 350 keV. The γ-ray discriminator was set at this energy to eliminate the relatively intense coincidences resulting from alpha population of the ground-state rotational band. A peak appears at 4.71 MeV, corresponding to an excitation energy of (800±15) keV. Its intensity is 5×10^{-7}, in good agreement with the intensity of a 765-keV γ ray observed in gamma singles spectra. There are no other alpha groups in true coincidence with an intensity greater than 1×10^{-8}.

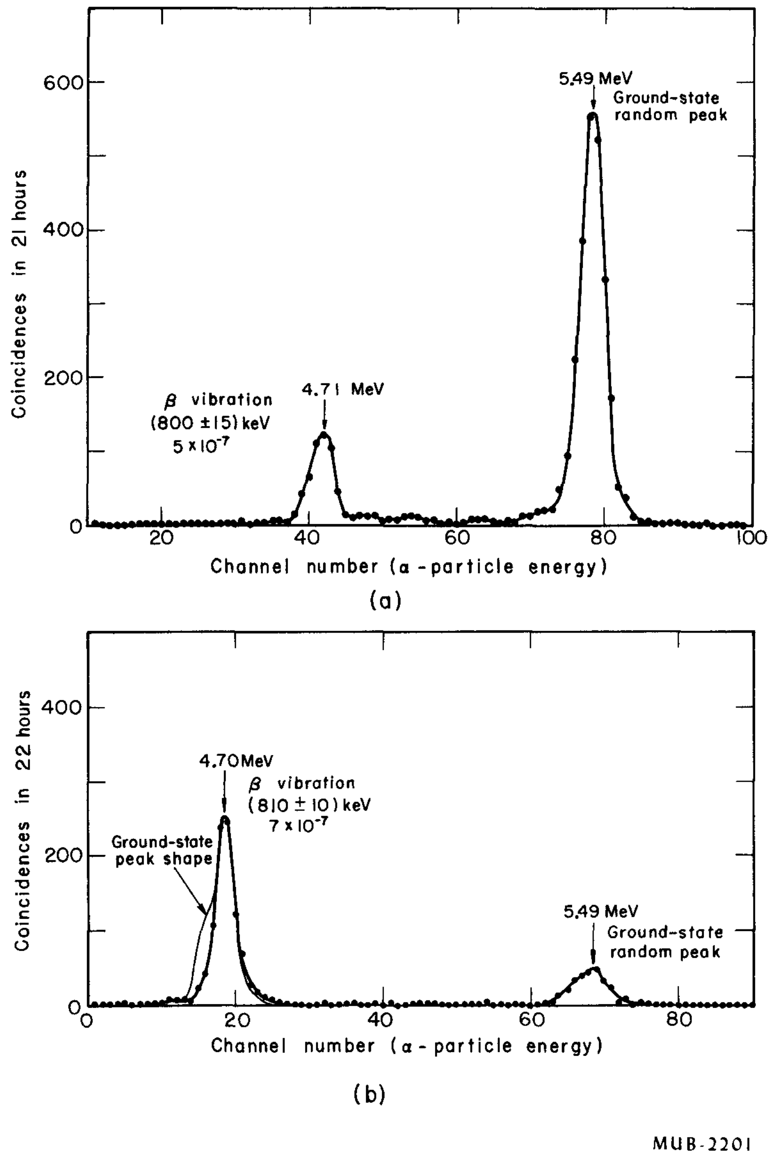


Fig. 5 a, b. Alpha decay of Pu^{238}
(a) Alpha spectrum in coincidence with γ rays > 350 keV. Pulse shaping is DDL; count rate, 1.65×10^5 alpha counts/sec. The resolution is 70 keV.
(b) Alpha spectrum in coincidence with electrons > 450 keV (DDL pulse shaping). Singles count rate is 0.93×10^5 alpha counts/sec; resolution is 45 keV. The shape of the alpha groups to the ground-state band, as recorded in the alpha singles spectrum, is drawn in lightly at the position of the 4.70-MeV peak for comparison.

The spectrum of α particles in coincidence with electrons of energy > 400 keV is shown in Fig. 5(b). A peak appears at 4.70 MeV, intensity 7×10^{-7}, in good agreement with e^--K x-ray measurements. Improved resolution during this run permitted partial resolution of the alpha groups populating the first two members of the ground-state rotational band. In Fig. 5(b), this alpha singles peak has been drawn in lightly at the position of the 4.70-MeV coincidence peak. The coincidence peak lacks the low-energy shoulder, indicating that the 02+ member of the K = 0+ band at 853 keV is either poorly populated, or has a lower E0 transition probability than the 00+ state.

The above spectra were taken with DDL pulse shaping. Figures 5(c) and 5(d) show similar spectra, but the resolution is considerably improved by the use of RC pulse shaping. Figure 5(c) shows the 4.70-MeV alpha peak in coincidence with electrons (semilog scale), again with the alpha singles spectrum to the ground-state band drawn in lightly for comparison; absence of the 853-keV state is seen even more clearly. The alpha-gamma coincidence spectrum, Fig. (5d), appears also to show no population of the 02+ state, but the peak is too broad on the high-energy side. This would be the case if a considerable fraction of the peak represents decay to the 01- state at 795 keV. The data have been interpreted this way, although the evidence is not compelling. This interpretation is not inconsistent with beta-decay data.[17-19]

The decay scheme is shown in Fig. 5(e). Coincident alpha groups in all the above spectra are identified as populating the 00+ state at 810 keV, with the possible exception noted in the last paragraph. Total population of the 00+ state is 1.1×10^{-6}. The hindrance factor (HF) for alpha decay to the state is 6. The 02+ state at 853 keV is not populated with an intensity > 1×10^{-7}; its hindrance factor is > 40. This failure to populate the 02+ member of the beta band is rather surprising since the corresponding transition in the ground-state band is virtually unhindred.

The absence of alpha decay to higher-lying states, limit 1×10^{-8}, permits the assignment of minimum hindrance factors: for the 922-keV 22+ state, HF > 20, and for the 1044-keV 00+ state, HF > 10. Gamma

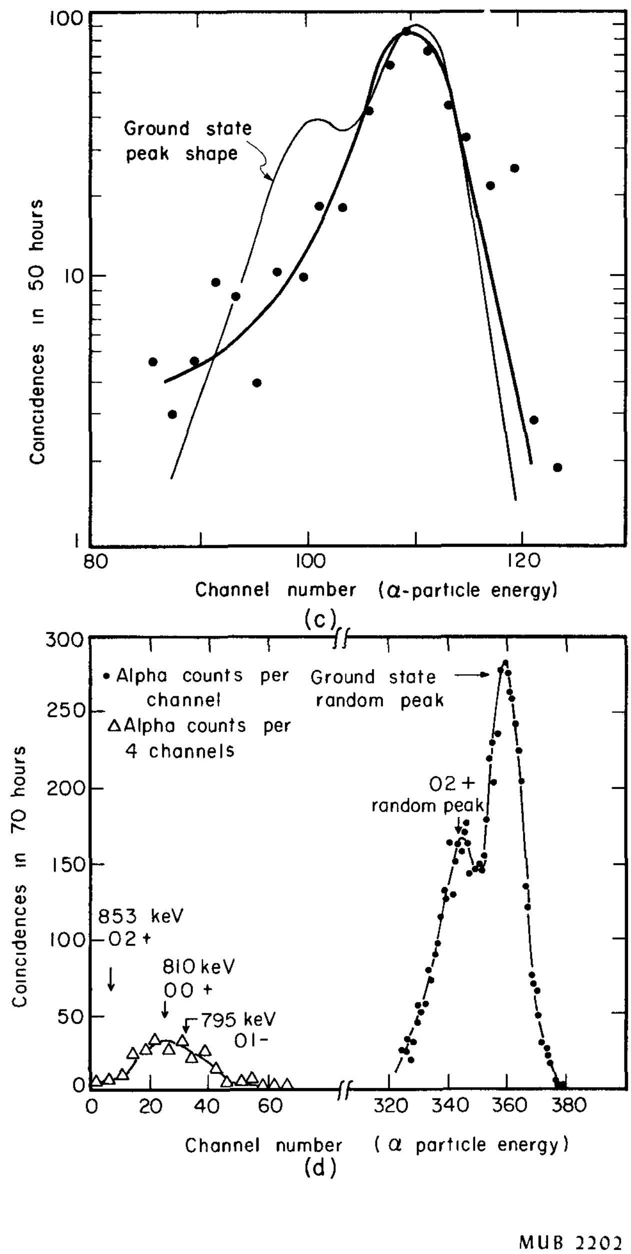


Fig. 5 c, d. Alpha decay of Pu^{238}.
(c) Semilog plot of the 4.70-MeV alpha group of Pu^{238} in coincidence with electrons > 350 keV. RC pulse shaping; count rate, 4×10^4 alpha counts/sec. The alpha singles peak has again been drawn in lightly for comparison.
(d) Alpha spectrum in coincidence with γ rays > 350 keV. RC pulse shaping; 5×10^4 alpha counts/sec. O Alpha counts per channel; Δ alpha counts per four channels.

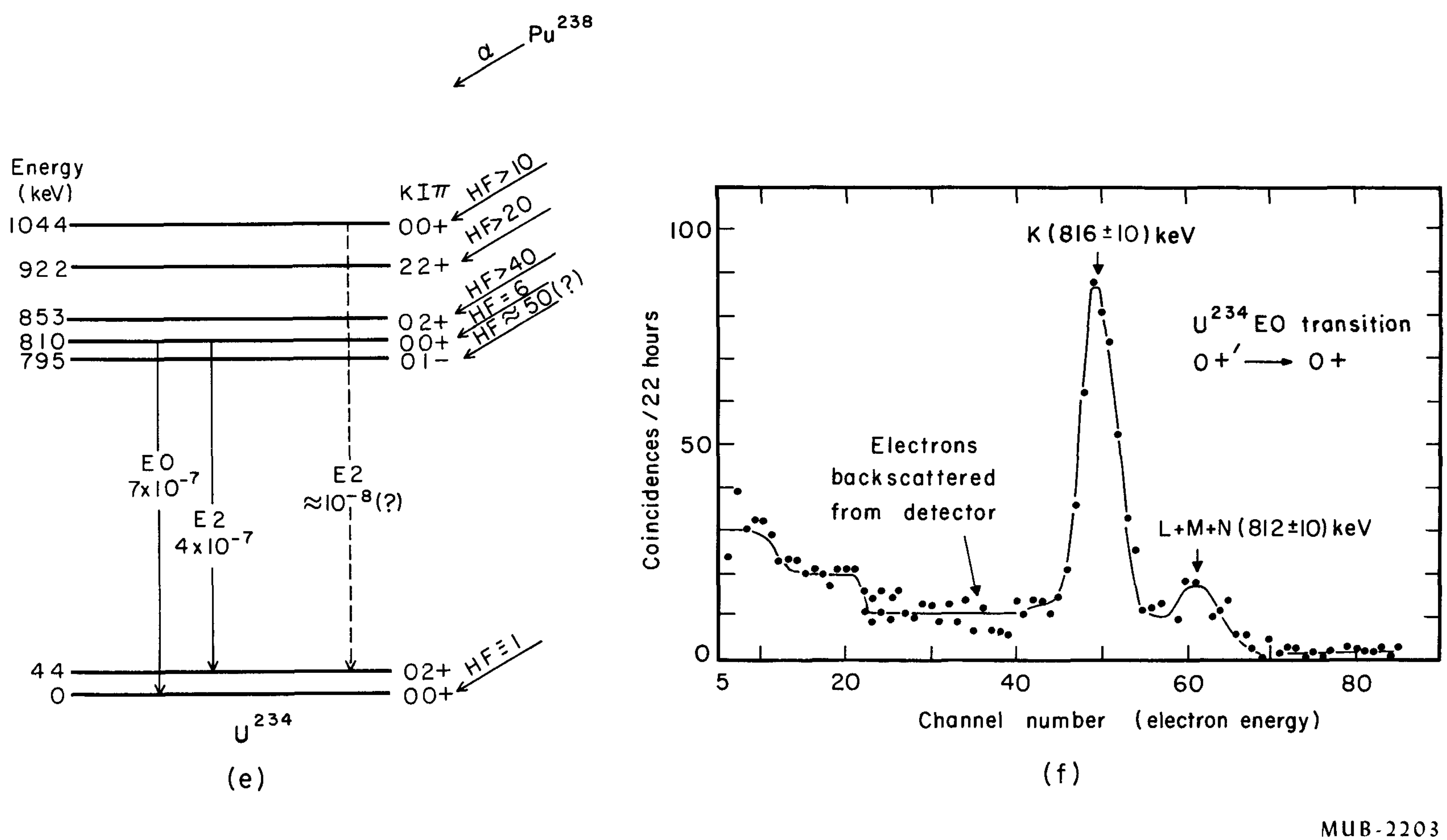


Fig. 5 e, f. Alpha decay of Pu^{238}.
(e) Decay scheme for Pu^{238}. The decay schemes given in this paper contain only the weak transitions studied in these experiments. (f) Electrons in coincidence with the alpha transition to the 810-keV state. The electrons were analyzed with a lithium-drifted silicon semiconductor.

singles spectra give some evidence for population of the 1046-keV level with an intensity of about 1×10^{-8}.

Figure 5(f) shows the spectrum of electrons in coincidence with α_{810}, taken with the lithium-drifted electron detector. The two peaks are identified as K and L+M+N+··· electrons from a single (816±10)-keV transition. There is no corresponding γ-ray transition, confirming again the E0 nature of the transition. The ratio K/K+L+M+··· is 3.5, in good agreement with beta decay measurements and theoretical predictions.[20]

B. The U^{230} Series

U^{230} decays by the chain

$$U^{230} \xrightarrow[20.8\ d]{\alpha} Th^{226} \xrightarrow[30.9\ m]{\alpha} Ra^{222} \xrightarrow[38\ s]{\alpha} Rn^{218} \xrightarrow[0.019\ s]{\alpha} Po^{214} \xrightarrow[1.6\times10^{-4}\ s]{\alpha} Pb^{210} .$$

The time needed for a coincidence measurement is long compared with the 31-minute half life with which the U^{230} daughters grow in. The five alpha emitters were therefore studied simultaneously. There was no difficulty with assignment of the observed transitions to the correct member of the decay chain, so the data are presented separately for each one, although transitions from each of the five alpha emitters were present in the spectra.

U^{230}:

Previous studies of U^{230} alpha decay[21, 22] have shown the existence of a 01- state at about 230 keV, decaying by γ rays of 158 and 232 keV. In my studies, a spectrum of α particles in coincidence with 210- to 370-keV γ rays showed the alpha transition to a state at (229±10) keV, identified with the 01- state. The intensity, $(1.4\pm0.5)\times10^{-3}$, is slightly lower than the intensity reported[22] for the 232-keV γ ray, $(2.4\pm0.5)\times10^{-3}$. (The intensities quoted in this section are all per alpha decay of a single member of the decay chain.)

No other alpha groups of U^{230} were observed. There are no alpha groups in coincidence with γ rays > 390 keV, with a limit of 1×10^{-5}. In coincidence with γ rays > 700 keV, there are no alpha groups more intense than 3×10^{-6}. In coincidence with electrons > 410 keV, there are no alpha groups more intense than 5×10^{-7}.

Th^{226}:

Th^{226} populates a 242-keV 01- state of Ra^{222} with an intensity 1.7×10^{-2}.[21,22] The alpha group to this state was observed in coincidence with 210- to 370-keV γ rays. The intensity, $(1.2\pm0.4)\times10^{-2}$, agrees well with the intensity previously reported for a 242-keV γ ray.[22]

No other alpha groups of Th^{226} were observed. In coincidence with γ rays > 390 keV, there are no alpha groups to states above 600 keV more intense than 1×10^{-5}. Below 600 keV excitation energy, where the spectrum is partially masked by alpha groups of Ra^{222}, a limit of 3×10^{-5} applies. In coincidence with gamma rays > 700 keV, there are no alpha groups of Th^{226} more intense than 3×10^{-6}. In coincidence with electrons > 410 keV, there is no alpha decay to states above 600 keV more intense than 5×10^{-7}.

Ra^{222}:

Previously reported states of Rn^{218}, observed in the alpha decay of Ra^{222}, include a 2+ state at 325 keV, and states at 650, 800, and 850 keV, tenatively assigned 2+, 1-, and 4+.[22, 23]

The alpha spectrum in coincidence with 210- to 370-keV γ rays shows population of a (326 ± 10)-keV state with an intensity $(4.1\pm1.2)\times10^{-2}$, in good agreement with the intensity previously reported[21] for a 325-keV γ ray, 3.6×10^{-2}. Owing to the presence of interfering transitions, it was not possible to observe decay to the 650-keV state, which decays via the 325-keV state by a cascade of two 325-keV γ rays.[23]

The situation with the two higher-lying levels is somewhat different than reported earlier. Figure 6(a) shows the alpha spectrum in coincidence with 370- to 550-keV γ rays, and 6(b) shows the alpha spectrum in coincidence with γ rays > 700 keV. Alpha groups populating

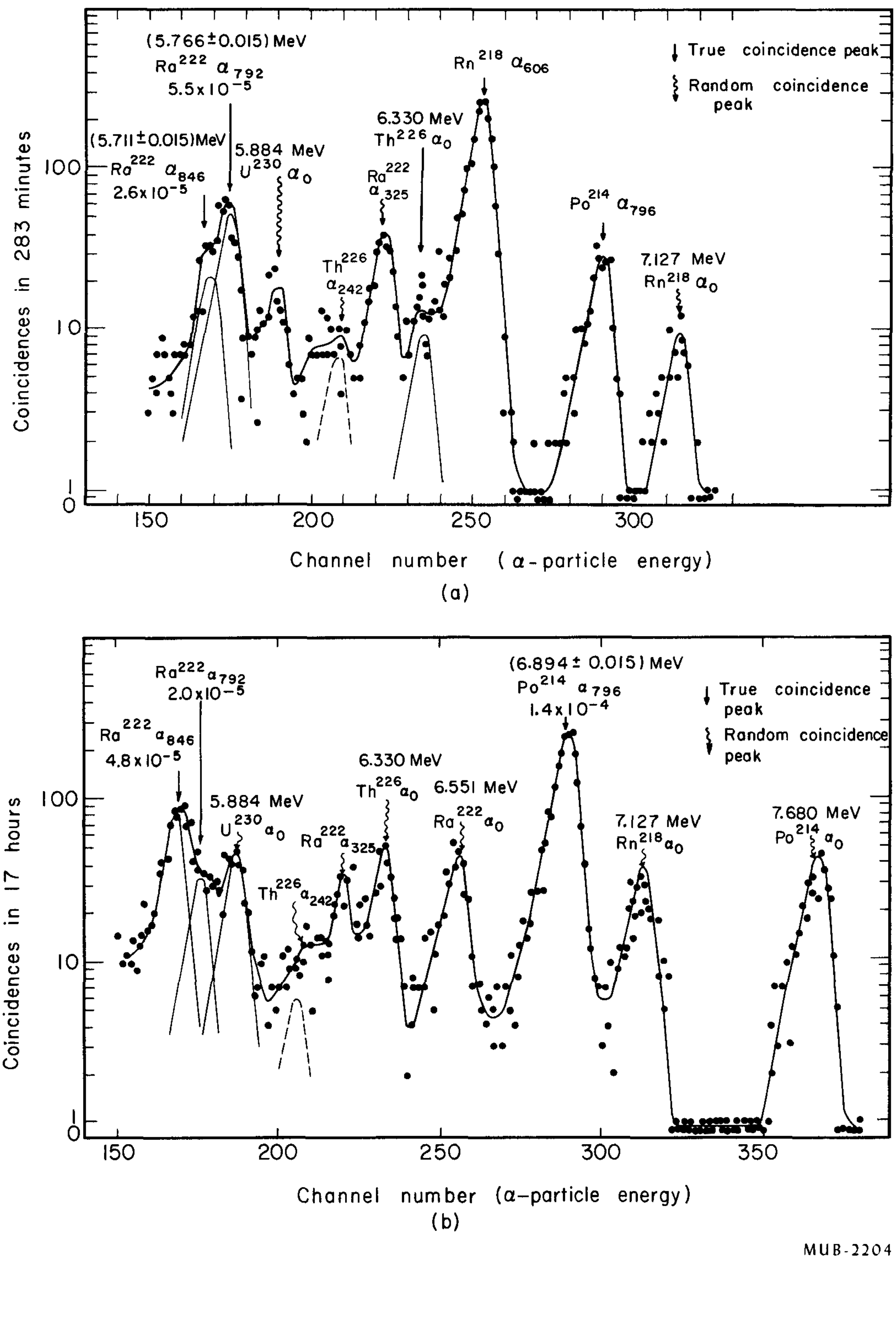
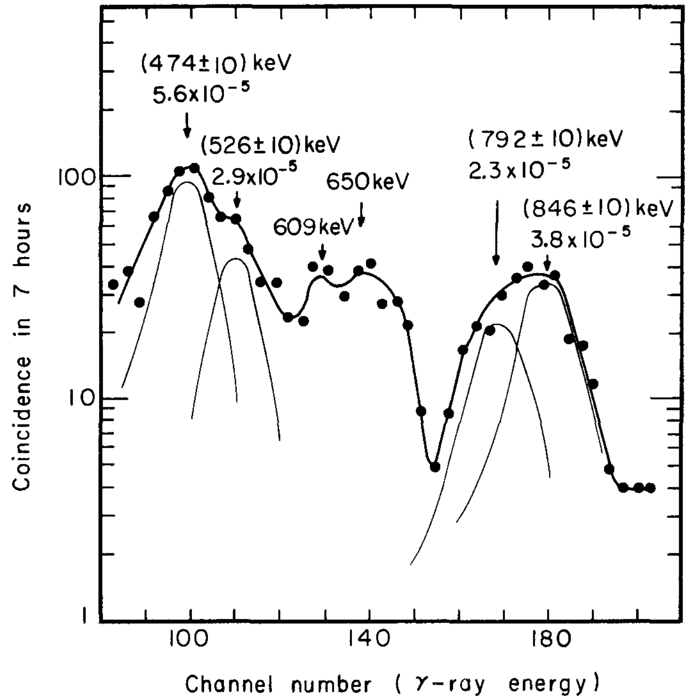


Fig. 6 a, b. The U^{230} decay series.
(a) Alpha particles in coincidence with 370- to 550-keV γ rays. The alpha groups α_{606} of Rn^{218} and α_{796} of Po^{214} appear because of coincidences with compton scattered γ rays.
(b) Alpha particles in coincidence with gamma rays > 700 keV.

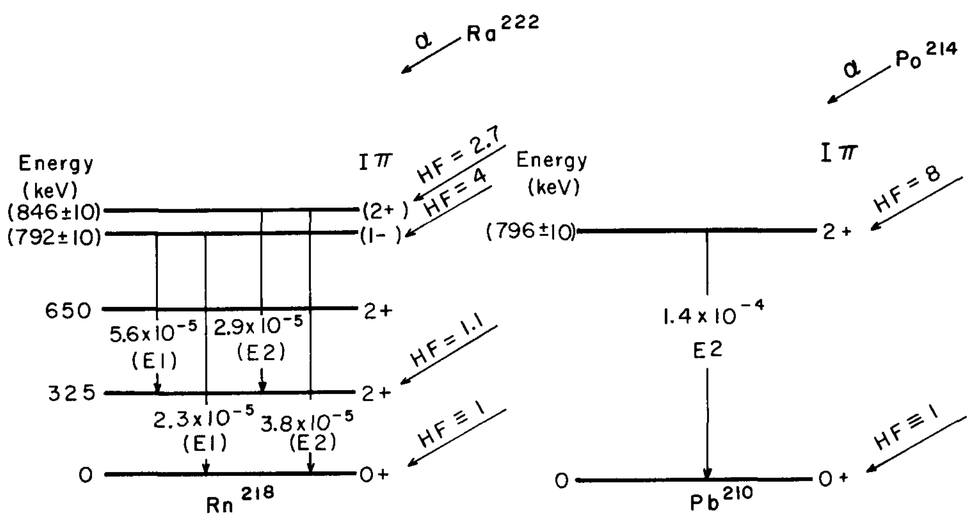
states of Rn^{218} around 800 and 850 keV excitation energy appear in both spectra. A portion of the gamma spectrum in coincidence with these two alpha groups is shown in Fig. 6(c). There are γ rays at 474, 526, 792, and 846 keV. The intensities of these γ rays are listed in Table I. The 474- and 526-keV γ rays have previously been observed in coincidence with the 325-keV γ ray[23]; their intensities from gamma-gamma coincidence measurements agree reasonably with the values listed in the table. The intensity reported[23] for a single 798-keV γ ray, previously observed in a gamma singles spectrum, is higher than the sum of the 792- and 846-keV gamma intensities listed in Table I. The overestimate of this intensity and the failure to resolve the two γ rays was undoubtedly due to the presence in the gamma singles spectrum of a 796-keV γ ray from Po^{214} decay; this will be seen shortly.

Figure 6(c) also shows some γ-ray intensity in the region of 600 to 700 keV. This must be caused partially by coincidences with the 609-keV γ ray from Rn^{218} decay, stack-up of the 325-keV cascade γ rays de-exciting the 650-keV state of Rn^{218}, and Compton tails from the higher-energy peaks. The possibility that the states around 800 keV de-excite partly by γ rays in the 600- to 700-keV region cannot be ruled out entirely. An alpha spectrum in coincidence with γ rays > 390 keV showed population of the two states around 800 keV with a total intensity 1.3×10^{-4}. This agrees well with the sum of the intensities of the four γ rays listed in Table I, indicating that these γ rays probably account for the entire de-excitation of the two states. A limit of 1×10^{-5} can be placed on all other γ rays above 390 keV de-exciting the two states.

An attempt to resolve the conversion electrons in coincidence with $\alpha_{792} + \alpha_{846}$ was only partially successful. The K electrons from the 474- and 526-keV transitions appear to have roughly equal intensity. The K-conversion coefficients are, very roughly, 0.01 for the 474-keV γ ray, and 0.025 for the 526-keV γ ray. This leads to a probable E1 assignment for the former and a probable E2 assignment for the latter. The fact that both the 792- and 846-keV states are populated



(c)



(d)

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Fig. 6 c, d. The U^{230} decay series.
(c) A portion of the γ-ray spectrum in coincidence with 5.65- to 5.85-MeV α particles.
(d) Decay schemes for Ra^{222} and Po^{214}. The decay schemes for U^{230}, Th^{226}, and Rn^{218} are as reported in the Table of Isotopes, 1958 (Reference 35).

Table I. States of Rn^{218} around 800 keV.

Excited-state energy (keV)	De-exciting radiation: Energy and intensity of γ rays	K-conversion coefficient	Transition assignment	Assignment of state Iπ
(792±10)	(474±10) keV $(5.6\pm1.7)\times10^{-5}$	~0.01	(E1)	(1-)
	(792±10) keV $(2.3\pm0.7)\times10^{-5}$	——	(E1)	
(846±10)	(526±10) keV $(2.9\pm0.9)\times10^{-5}$	~0.025	(E2)	(2+)
	(846±10) keV $(3.8\pm1.1)\times10^{-5}$	——	(E2)	

by alpha decay and de-excite to 0+ and 2+ states limits their possible spins and parities to 1- or 2+. The preferred choices are shown in the table and the decay scheme, Fig. 6(d).

Rn^{218}:

The only known excited state of Po^{214} below 1.3 MeV is a 2+ state at 609 keV.[21] The alpha spectrum in coincidence with γ rays > 390 keV shows population of a state of Po^{214} at (606±15) keV with an intensity $(1.6\pm0.5)\times10^{-3}$. This is in reasonable agreement with the value $(2.0\pm0.5)\times10^{-3}$ previously reported for the intensity of a 609 keV gamma ray.[21]

The same alpha group was also observed in coincidence with electrons > 410 keV, with an intensity $(3.6\pm1.1)\times10^{-5}$. The conversion coefficient for the 609-keV γ ray is thus 0.022 ±0.006, in excellent agreement with the theoretical value for an E2 transition, 0.021. The theoretical conversion coefficient for an E1 is 0.006; for all other multipolarities it is greater than 0.07. This confirms the E2 transition and the 2+ assignment for the state.

Po^{214}:

The alpha spectrum in coincidence with γ rays > 700 keV, Fig. 6(a), shows an alpha group at 6.894 MeV, (801±15) keV excitation in Pb^{210}. A (792±10)-keV γ ray was observed in coincidence with this alpha group. The intensity is $(1.4\pm0.4)\times10^{-4}$ in both cases, and the best energy for the state is (796±10) keV. As noted in the above section on Ra^{222} decay, the 792-keV γ ray masked weaker transitions from the decay of Ra^{222} in earlier gamma singles measurements.

The alpha transition to the 796-keV state has been observed in a magnetic spectrograph.[25] The energy of the excited state quoted is 796 keV, and the intensity of the alpha transition is 1×10^{-4}, in good agreement with my determination. A γ ray around 780 keV has been reported in the β^- decay of Tl^{210}, and a 2+ state of that energy was tentatively assigned in Pb^{210}.[24]

The 6.89-MeV alpha group also occurs in coincidence with electrons > 410 keV, with an intensity $(2.5\pm0.8)\times10^{-6}$. The conversion coefficient for the 792-keV γ ray is 0.017±0.005, compared with the theoretical values 0.0114 for an E2, 0.003 for an E1, and > 0.04 for all other multipolarities. The agreement is good enough to warrant an E2 assignment, and the (796±10)-keV state is assigned a spin and parity of 2+.

C. $U^{232} \xrightarrow{\alpha} Th^{228}$

Levels of Th^{228} have been studied extensively in the electron-capture decay of Pa^{228},[26] and in the β^- decay of Ac^{228}.[27,28] (Other references are not quoted because they add no information, or contain substantial errors in the decay scheme.) Levels reported include 328 keV (01-), 396 keV(03-), and 969 keV (22+).

Gamma spectra of U^{232} (references 29, 30) display gamma rays of 266- and 324 keV, interpreted as depopulating the 328-keV 01- state of Th^{228}. The alpha spectrum in coincidence with γ rays between 240 and 360 keV is shown in Fig. 7(a). The intensity of the peak at 5.000 MeV, or (323±10) keV excitation, is $(6\pm2)\times10^{-5}$, in good agreement with the value from gamma singles measurements. No decay to the 03- state at 396 keV is evident.

A much weaker alpha group appears at about 820 keV excitation. When one gates on this group (see the pulse-height window, PHW, in Fig. 7(a)) the gamma spectrum of Fig. 7(b) is observed. In addition to the γ rays at (272±10) and (334±10) keV, a considerable portion of which must have resulted from tailing of the 5.00-MeV alpha group into the PHW, there is a single peak at (500±10) keV, and very weak peaks at higher energies. The 500-keV γ ray is interpreted as a transition from the state at about 820 keV to the 328-keV 01- level. In a subsequent measurement, an alpha group at (823±10) keV was observed in coincidence with the 500-keV γ ray (Fig. 7(c)).

The existence of the cascade transition was also verified in a γ-γ coincidence experiment (Fig. 7(d)). The intensity of the cascade, $(2.1\pm0.6)\times10^{-7}$, agrees excellently with the value obtained in α-γ coincidence spectra.

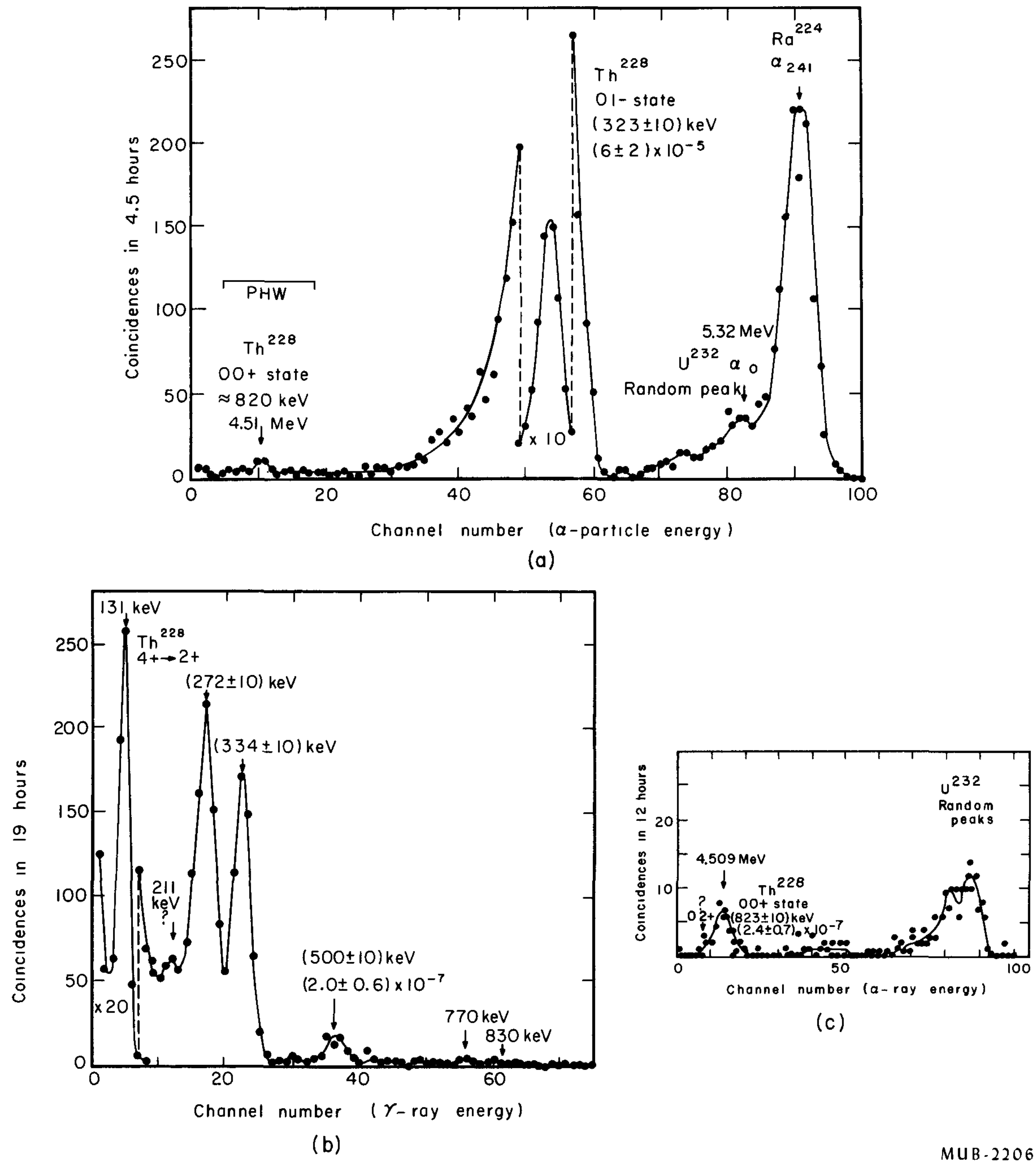

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Fig. 7 a, b, c. Alpha decay of U^{232}.
(a) Alpha particles in coincidence with 240- to 360-keV γ rays. PHW refers to the pulse-height window (alpha energy discriminator) used to gate the γ-ray spectrum, Fig. (b).
(b) Gamma spectrum in coincidence with the alpha group to the state around 820 keV.
(c) Alpha particles in coincidence with 440- to 650-keV γ rays.

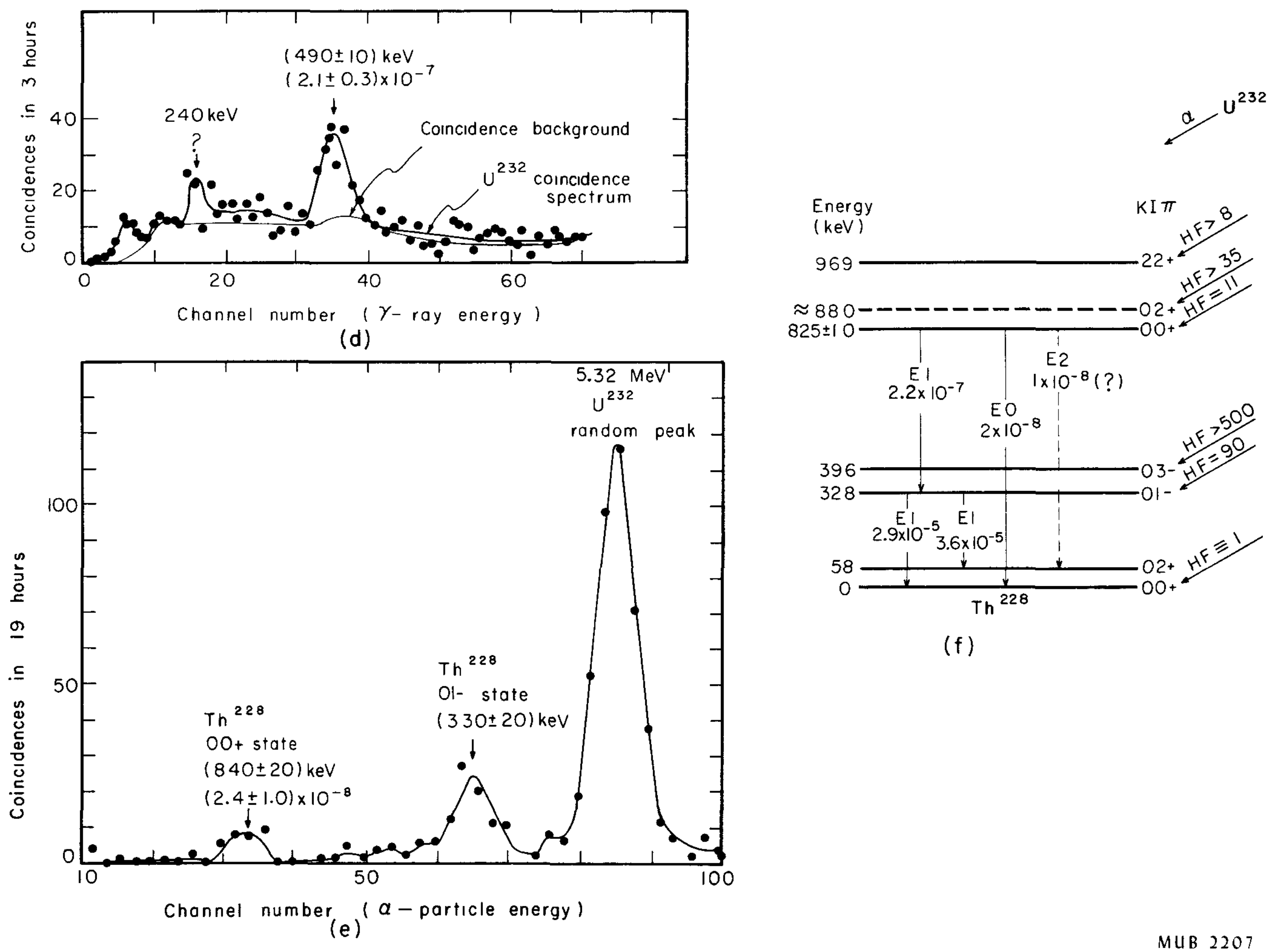

Fig. 7. d, e, f. Alpha decay of U^{232}.
(d) Gamma-gamma coincidence spectrum: γ rays in coincidence with 235- to 365-keV γ rays.
(e) Alpha particles in coincidence with electrons > 283 keV.
(f) Decay scheme for U^{232}.

Arbman et al.[26] failed to observe such cascades in electron-gamma coincidence measurements on Pa^{228}. The β^- decay of Pa^{228} thus does not populate the 825-keV state in appreciable intensity. There is also no evidence for population of this level in Ac^{228} decay, although the search was not so thorough.

Weak γ rays found in coincidence with α_{823} at about 770 and 830 keV result largely from stack-up of the cascading γ rays, owing to the close geometry of the NaI detector. Correcting for this, one finds the intensities of each of these transitions to be $(1\pm1)\times10^{-8}$.

The best energy value deduced for the new state is (825±10) keV. Because it decays only to the 01- member of the octupole band, it is probably a 00+ state. The low hindrance factor, 11±3, is excellent evidence that in fact is a beta vibrational state, and it has been so assigned.

Figure 7(e) shows the alpha spectrum in coincidence with electrons > 285 keV. The peak at (840±20) keV, intensity 2.4×10^{-8}, is identified with the same 00+ state. On the assumption that the 500-keV γ ray is an E1 transition, most of the electrons in coincidence with this alpha group must represent a monopole transition to the ground state, intensity $(2\pm1)\times10^{-8}$. This information and the rest of the decay scheme are summarized in Fig. 7(f).

As can be seen from Fig. 7(c), no population of the expected 02+ member of the beta band is observed, limit 3×10^{-8}. The same limit applies to the 22+ state at 969 keV.

On the basis of vibration-rotation distortion of the ground-state band, it has been predicted that the beta vibration of Th^{228} should lie at about 520 keV.[31] From these measurements, no state in the region 430 to 780 keV is populated with an intensity > 5×10^{-8}. This limit applies whether the state decays to the ground-state band or to the odd parity band. More is said about this in Section V.

D. $U^{234} \xrightarrow{\alpha} Th^{230}$

Previously observed levels of Th^{230} include 508 keV (01-),[32,33] 634 keV (00+),[7,8,33] 677 keV (02+),[34] and 783 (22+).[33]

The alpha spectrum in coincidence with electrons shows population of a state of Th^{230} at (650±20) keV, intensity $(2.1\pm0.5)\times10^{-7}$. This state is identified with the 634-keV 00+ level. The intensity is higher than previously reported,[7,8] 6×10^{-8} for the K line. The spectrum of γ rays in coincidence with 3.98- to 4.38-MeV α particles shows population of the 00+ state and a state at 510 keV, identified with the 508-keV 01- state; the intensities are in good agreement with the results of gamma singles measurements.[32]

The decay scheme is shown in Fig. 8. It is impossible to tell from these data whether or not the 00+ state decays partly by E1 cascades via the 01- state, as in Th^{228}. This is so because the low specific activity of U^{234} makes it very difficult to resolve the alpha groups to the 508- and 634-keV states, and the possible 126-keV γ ray connecting these states would be obscured by radiations from U^{235} present in the source. In the absence of experimental information, it has just been assumed that no such E1 cascades occur.

The hindrance factor for the 01- state is 300±80, for the 00+ state it is 40±10, and for the 22+ state, HF > 20.

E. $Pu^{236} \xrightarrow{\alpha} U^{232}$

The levels of U^{232}, as populated by the β^- decay of Pa^{232}, have been carefully studied by examination of the continuum spectrum and de-exciting radiations with magnetic spectrometers and a bent-crystal γ-ray spectrometer.[35] The states include 565 keV (01-), 630 keV (03-), 693 keV (00+), 735 keV (02+), and 868 keV (22+).

Alpha spectra in coincidence with γ rays and with electrons are shown in Figs. 9(a) and 9(b). The relatively intense peaks are well suited to studies at higher resolution, and the spectra shown were obtained by using RC pulse shaping and a 1-μsec preamplifier time constant. The high intensity of electrons de-exciting the (700±10) keV

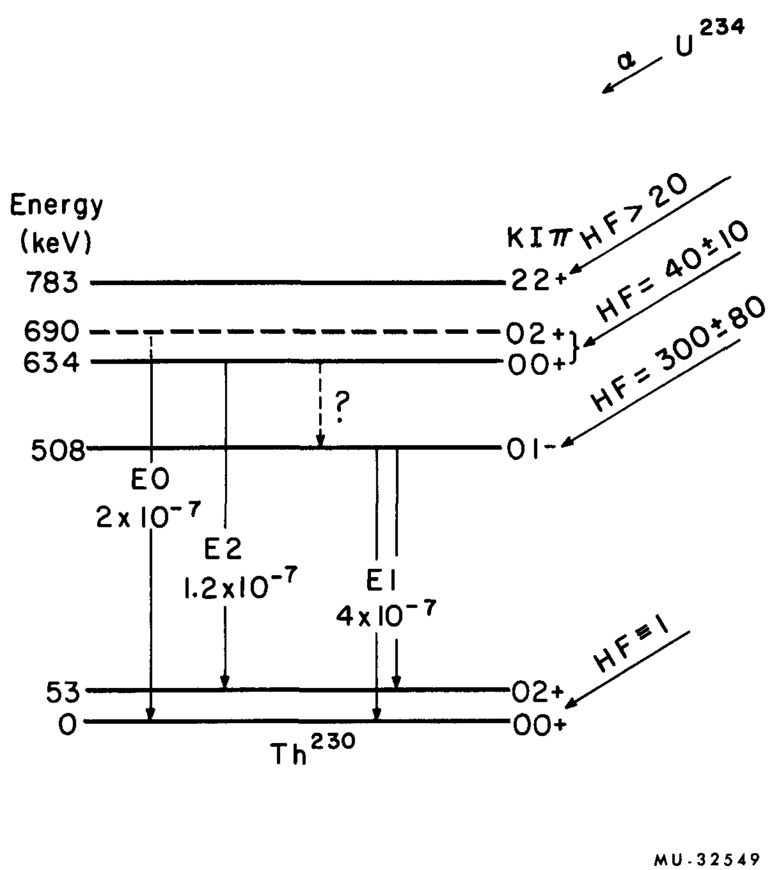

Fig. 8. Decay scheme for U^{234}.

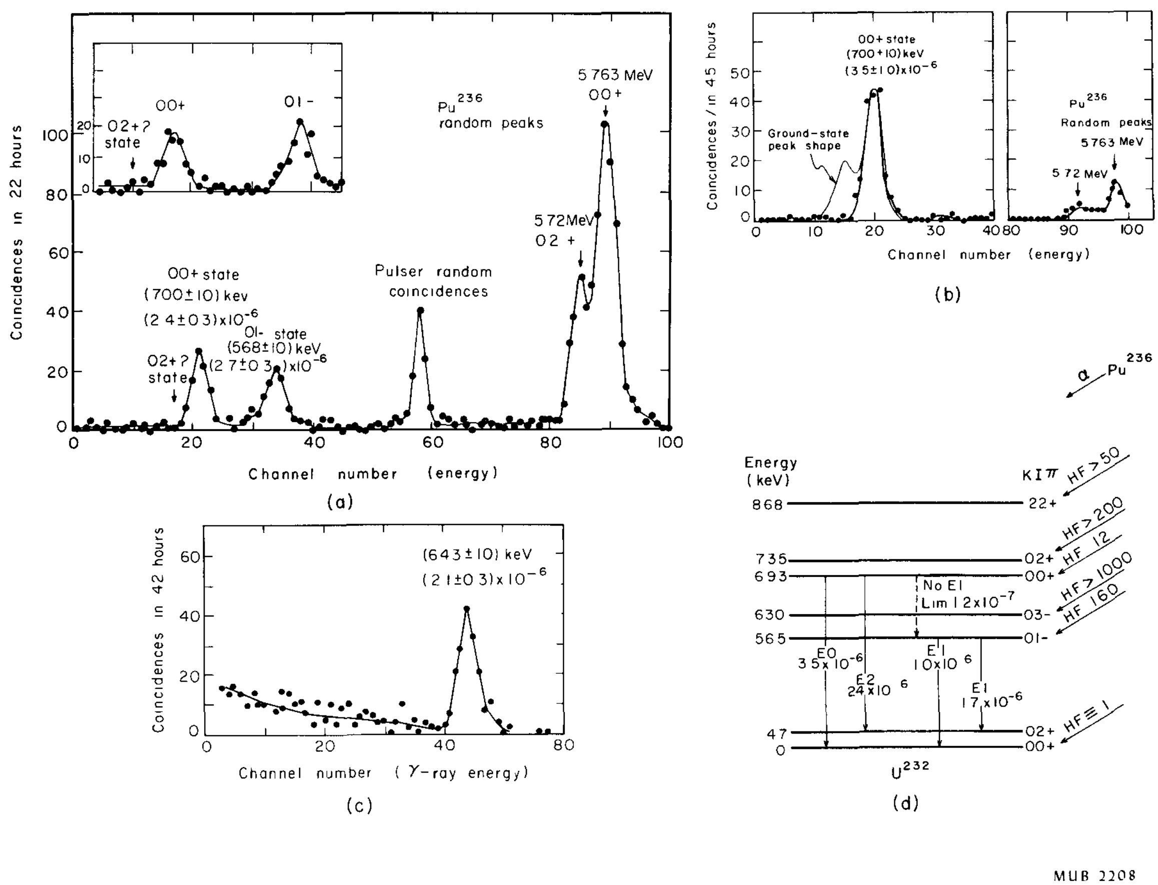

Fig. 9 a, b, c, d. Alpha decay of Pu^{236}.

(a) Alpha particles in coincidence with γ rays > 420 keV. The pulser peak was added during the entire run to check stability. The inset shows a portion of a better-resolved spectrum, run for 90 hours at a lower count rate.

(b) Alpha particles in coincidence with electrons > 300 keV. The alpha singles peak is drawn in lightly at the position of the coincident group for comparison.

(c) Gamma rays in coincidence with the alpha group to the 693-keV 00+ state.

(d) Decay scheme for Pu^{236}.

level helps to confirm its identification as the 693-keV 00+ state. Failure to observe the 565-keV state in the α-e^- spectrum sets an upper limit of 0.015 on the conversion coefficient of γ rays de-exciting that state, confirming their E1 multipolarity and the 1- assignment for the state. In another spectrum (not shown), the expected two γ rays de-exciting the state were observed. Their relative intensities are in agreement with theoretical predictions and the value found in Pa^{232} decay.[35]

The gamma spectrum in coincidence with α_{693} (Fig. 9(c)) shows clearly the expected E2 γ ray at (643±10) keV, but gives no evidence of γ rays at 128, 518, or 565 keV that would indicate the presence of E1 cascade transitions, such as occur in Th^{228}. A limit of 1×10^{-7}, 2% of the total decay of the 00+ state, can be placed on such transitions.

Total population of the 01- and 00+ states is, respectively, $(6\pm2)\times10^{-6}$ and $(2.7\pm0.6)\times10^{-6}$. For the 01- state, HF = 160±40, and for the 00+ state, HF = 12±3. The 00+ state decays 60% by E0 electrons and 40% by E2 gammas, in fair agreement with the value obtained in Pa^{232} decay.[35]

No decay to the 630-keV 03- state was observed, limit 1.5×10^{-7}, or HF > 1000. More surprising, no population of the 735-keV 02+ state was found, limit 2×10^{-7}, or HF > 200. For the 22+ state at 835 keV, HF > 50.

Figure 9(d) summarizes the results.

F. $Pu^{239} \xrightarrow{\alpha} U^{235}$

The alpha decay of Pu^{239} has been studied by many investigators.[36] Recent studies of the alpha spectrum have revealed more than 20 levels below 550 keV, a number of which have been assigned to three rotational bands of U^{235}.[37,38] The ground state of Pu^{239} is 1/2+[631],[39,40] and favored alpha decay populates the well-known 1/2+ isomeric level of U^{235} 75 eV above the 7/2- ground state.[41]

The alpha spectrum in coincidence with electrons > 400 keV establishes a state or band at (780 ± 20) keV, decaying by electron emission with an intensity of $(7\pm2)\times10^{-8}$. This is in reasonable agreement with the value $(1.5\pm0.8)\times10^{-7}$ deduced from electron—K-x-ray coincidence measurements.[42]

The same alpha group appears in coincidence with γ rays > 680 keV, intensity $(1.8\pm0.7)\times10^{-7}$. The total intensity of the alpha group, $(2.5\pm0.8)\times10^{-7}$, corresponds to a hindrance factor of 25±8. In comparing this value with hindrance factors for states of even-even nuclei, it is more reasonable to divide it by the hindrance factor for the least hindered factor for the least hindered ("favored") alpha transition, 2.5 in this case.[43] The "reduced" value of the hindrance factor is then 10±3.

The high electron intensity de-exciting the state shows the existence of an E0 transition. Together with the low hindrance factor, this indicates a beta vibration analogous to the 810-keV 00+ state of U^{234}.

Another state was observed at (650±20) keV. It decays by γ rays with an intensity of $(8\pm3)\times10^{-7}$, and by electrons with an intensity of less than 1% of this value. Gamma radiation of this intensity has previously been reported around 640 keV.[44-46] The de-exciting radiation is unambiguously E1, and the state has been assigned K = 1/2-, by analogy with the 01- states of even-even nuclei.

Figure 10 illustrates the decay scheme.

G. $Pu^{240} \xrightarrow{\alpha} U^{236}$

There is very little published information on the excited levels of U^{236} above the ground-state rotational band. In the Coulomb excitation of U^{236} with protons, Durham et al. observed a 909-keV transition, which they assigned as a monopole from a 02+ state at 953 keV to the 45-keV 02+ state.[34] This would place the 00+ member of the beta band around 909 keV. Gallagher and Thomas, in their study of Np^{234} decay, observed some weak electron lines which decayed with the 22-hr Np^{236} half life.[17] These were assigned to 687.0- and 641.7-keV transitions de-exciting a 687-keV state of U^{236} to the ground and first excited states. Pu^{240} γ-ray singles spectra by Murri and Cline,[45, 46] show the existence of a (640±10)-keV transition, intensity 1.7×10^{-7}.

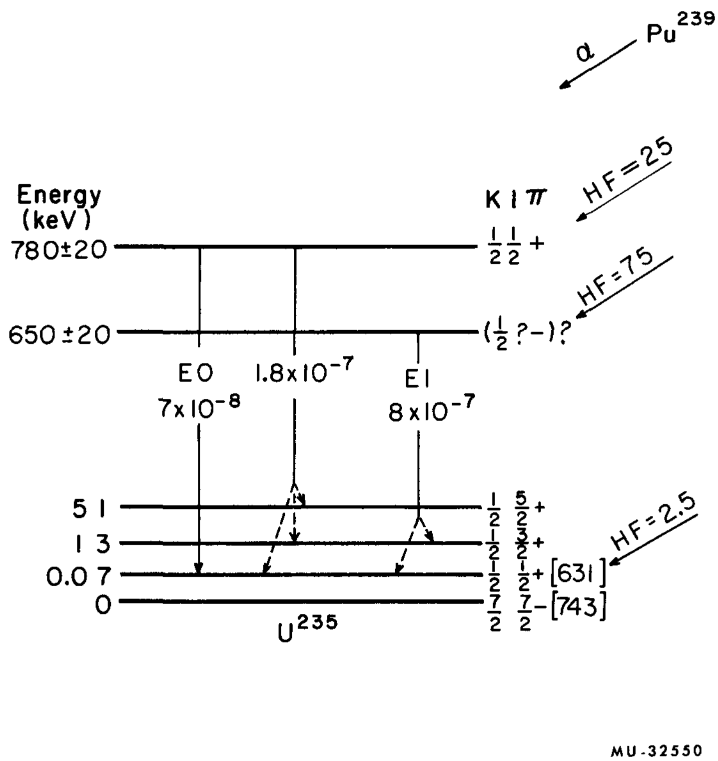

Fig. 10. Decay scheme for Pu^{239}.

My studies give scant evidence for population of the proposed 910-keV 00+ state. A questionable (860±30)-keV γ ray was seen in the Pu^{240} gamma singles spectrum, and in coincidence with 4.0- to 4.4-MeV α particles (5 counts observed). The alpha-electron coincidence spectrum gives questionable evidence for an alpha group to a state around 880 keV, intensity $\approx 3\times 10^{-9}$. (See Fig. 11(a)).

Alpha decay to a state around 690 keV was observed in coincidence both with electrons (Fig. 11(a)) and with γ rays. Total population of the state is $(2.1\pm 0.4)\times 10^{-7}$, or HF = 70±15.

Figure 11(b) shows the gamma spectrum in coincidence with the 4.48-MeV alpha group. The peak at (650±10) keV has an intensity 1.7×10^{-7}. A gamma singles spectrum showed this peak resolved into two γ rays. The best energies and intensities for the transitions are (644±5) keV, $(1.44\pm 0.24)\times 10^{-7}$, and (692±10) keV, $(0.36\pm 0.09)\times 10^{-7}$.

The electron spectrum in coincidence with the 4.48-MeV alpha group, Fig. 11(c), shows clearly the existence of two different K-electron lines. The electron intensities and conversion coefficients are listed in Table II, together with the γ-ray data. Nielsen and others have recently observed 642- and 687-keV transitions in the electron-capture decay of Np^{236}.[47] Their conversion coefficients and relative intensities are included in the table for comparison with the results from alpha decay. The close agreement between the values is the basis for assignment of the transitions observed in alpha decay to Nielsen's accurate energy values, and to the more precise energies of Gallagher and Thomas, 641.7 and 687.0 keV.[17]

The results are puzzling, and no straightforward interpretation fits the data. The 687-keV transition terminates at the 0+ ground state of U^{236}, and should therefore be of a single multipole order. Its conversion coefficient is consistent only with an M2 assignment, which requires that there be a 2- state at 687 keV. The measured alpha energy, however, places the excited state populated by alpha decay at (693±20) keV. Although this is very close to 687 keV, parity conservation forbids alpha decay of an even-even parent to a 2- state. There appear to be

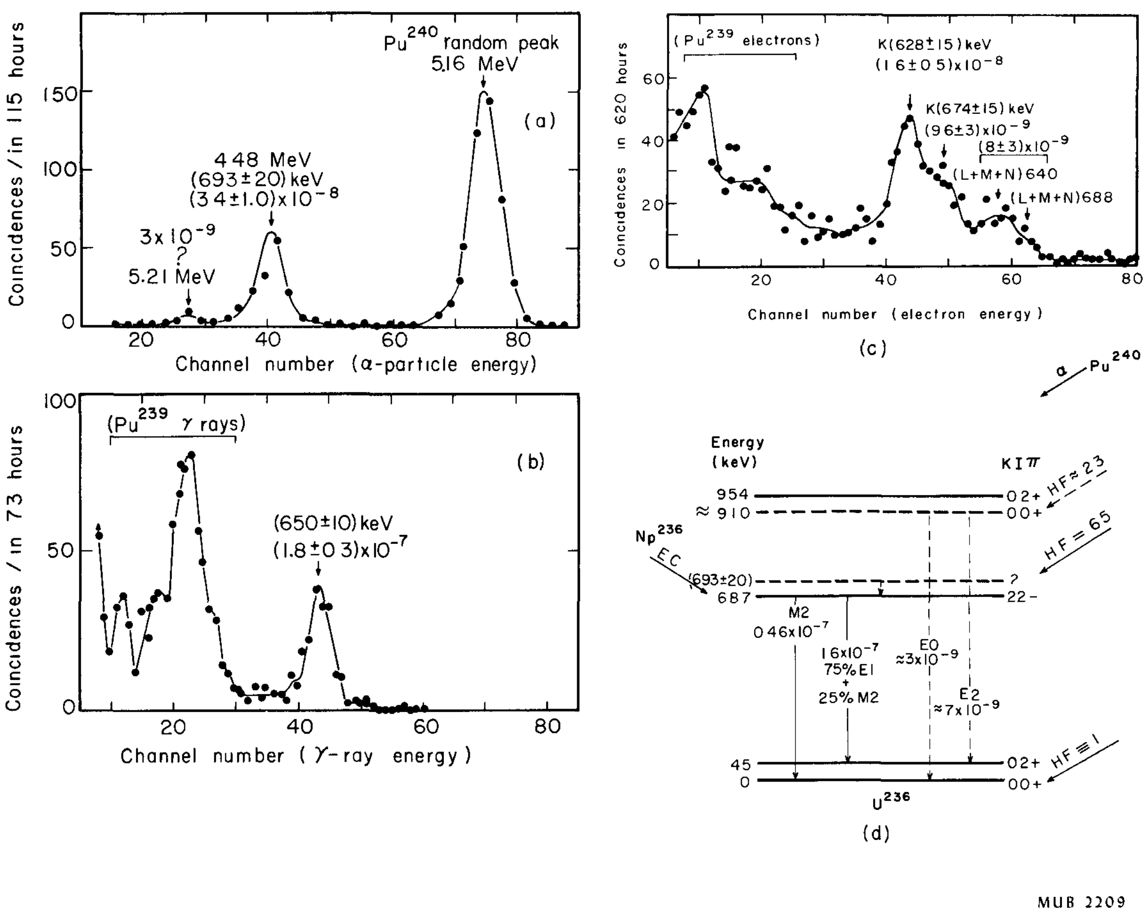

Fig. 11 a, b, c, d. Alpha decay of Pu^{240}.

(a) Alpha particles in coincidence with electrons > 450 keV.

(b) Gamma rays in coincidence with α_{693}. The Pu^{240} source contained 2% Pu^{239} by alpha activity, and some Am^{241}. The source used for the other measurements contained < 0.7% Pu^{239}, and negligible quantities of other active contaminants.

(c) Electrons in coincidence with α_{693}. The electrons were analyzed with a lithium-drifted silicon semiconductor.

(d) A possible decay scheme for Pu^{240}.

Table II. Gamma transition in coincidence with Pu^{240} $\alpha_{693 \pm 20}$

		642 keV	687 keV
Transition assignment[a]		642 keV	687 keV
Measured γ-ray energy		(644±5) keV	(692±10) keV
Measured electron transition energy		(628±15) keV	(674 ±15) keV
Gamma ray intensity		$(1.44 \pm 0.24) \times 10^{-7}$	$(0.36 \pm 0.09) \times 10^{-7}$
K-electron intensity		$(1.6 \pm 0.5) \times 10^{-8}$	$(1.0 \pm 0.3) \times 10^{-8}$
α_K	From this work	0.115±0.04	0.27±0.10
	From Np^{236} E. C. decay[a]	0.11±0.03	0.20±0.05
K/L+M+N+···	From this work	3.5±1.0	3.0±1.2
	From Np^{236} decay[a]	2.6±0.5	3.4±0.5
$\frac{W_\gamma(642)}{W_\gamma(687)}$	From this work	4±1	
	From Np^{236} decay[a]	3.3±0.5	
$\frac{W_K(642)}{W_K(687)}$	From this work	1.70±0.25	
	From Np^{236} decay[a]	1.70±0.15	

[a]from reference 47

several possible explanations:

(a) The 687-keV transition is in fact not an M2, and its conversion coefficient is anomalous, or

(b) the 687-keV transition is not of pure multipole order; this can be the case only if the "transition" consists of two different transitions of the same energy, such as an E0 and an E2, originating from different states, or

(c) alpha decay does not populate the 687-keV state directly.

There is precedent for the first explanation, in that anomalously high E1 and M1 conversion coefficients have been observed. They occur in special cases in which selection rules inhibit contributions to internal conversion from the radiation field outside the nuclear volume.[48] There is, for example, an 84-keV E1 in Pa^{231} decay, whose K-conversion coefficient is 10 times the theoretical value, and whose L_I and L_{II} conversion coefficients are 20 times too large.

There are, however, substantial reasons for ruling out anomalous conversion in this case. M1 transitions are not expected, although there could be E1's de-exciting an octupole vibrational state. The E1 transitions from such a state to the ground-state band should not be subject to the special selection rules that apply to anomalously converted transitions. The E1's de-exciting octupole states, including those of Th^{228}, Th^{230}, U^{232}, U^{235}, Pu^{238}, and Pu^{240} studied in my experiments, do not have anomalously high conversion coefficients. Stephens and Diamond have determined from Coulomb excitation that the E1's de-exciting octupole states of Th^{232} and U^{238} are hindered by factors of 10^2 to 10^3, whereas anomalously converted E1's are hindered by much larger factors.

It should be possible to rule out anomalous E1's by observing the L-subshell ratios of the 687-keV transition in Np^{236} decay, since the L_{III} conversion coefficient should be normal, while the L_I and L_{II} would be high in the case of an anomalous transition.

The possibility that the transitions are actually double lines from two nearly degenerate states at around 687 keV must be considered more seriously, because admixtures of E0 transitions, which de-excite beta vibrational states, occur commonly in deformed nuclei. Nevertheless, there is very strong, though not conclusive evidence, that the gamma transitions are not unresolved double lines:

(a) Attempts were made to reproduce the conversion and relative-intensity data in Pu^{240} decay, using combinations of hypothetical 00+, 02+, 04+, 22+, 23+, 24+, 01-, and 03- states around 687 keV, but all such schemes failed to give the observed intensities. All combinations tried required a 00+ state in the vicinity; this is also objectionable because the hindrance factor for alpha decay to this state would be higher than observed for other 00+ states, and because a "beta vibrational" band of U^{236} probably exists around 910 keV.

(b) The excellent agreement (Table II) between the transition intensities observed in Pu^{240} alpha decay and those observed in Np^{236} electron-capture decay is not possible for transitions originating from more than one state, except in the unlikely event that alpha and beta decay populate the several states with the same relative intensities.

(c) Nielsen's high-resolution electron spectra would show complex lines or energy discrepancies unless the transitions originated from a single state, or several states within 1 keV of each other, whereas in fact no such complex lines or energy discrepancies were seen. The energy difference between Gallagher and Thomas's[17] two lines agrees with the spacing between the ground state and first excited state to a few tenths of a kilovolt.

The third possibility, that the 687-keV level is populated indirectly in alpha decay, requires a state above 687 keV and a fast transition between this state and the 687-keV level, although there is no evidence that either of these really exists. If the 687-keV level were a 22- octupole state, there should be a 23- state around 730 keV which should decay to the 22- state by a fast M1-E2 rotational transition. However, the state populated directly by alpha decay is at (693±20) keV, which appears to be too low for the hypothetical 23- state, and is disturbingly close to 687 keV.

In spite of these objections, the third possibility seems to be the most plausible one. A possible decay scheme along this line is proposed in Fig. 11(d).

The proposed decay scheme is reasonable and self-consistent. One can expect 22- octupole vibrations to occur, although none have been definitely identified to date. Theoretical and experimental conversion coefficients agree very nicely in this scheme. The predicted ratio of K electrons $W_K(642)/W_K(687)$, based on a 3:1 E1/M2 mixing ratio for the 642-keV transition, is 1.50, in good agreement with the observed value, 1.70±0.25. The 3:1 E1/M2 mixing ratio for the 642-keV transition was calculated on the assumptions that the 687-keV γ ray is pure M2, and that the ratio $W_{M2}(642)/W_{M2}(687)$ is 1.02, as predicted by theory.

The intensity of the γ rays measured in the gamma singles spectra is the same as the value found in alpha-gamma coincidence measurements, showing that the lifetime of the 687-keV state is shorter than the resolving time of the coincidence circuit, $\approx 2\times10^{-8}$ sec. This implies that the M2 transitions have partial half lives shorter than $\approx 5\times10^{-8}$ sec, corresponding to a minimum reduced transition probability of 0.02 s. p. (single particle) units, a reasonable value. Assuming that the M2's are between 0.02 and 1 s. p. units, the E1 transition has a partial half life between 8×10^{-10} and 3×10^{-8} sec, corresponding to a reduced transition probability between 2×10^{-6} and 4×10^{-8} s. p. units. As a very rough estimate, the E1 should be hindered by about 3 orders of magnitude for an octupole state, and 2 or 3 orders of magnitude for K forbiddenness, in reasonable proximity to this range. It should be noted that a small proportion of collective E3 transitions (about 20 s. p. units for a 03- state) could also compete with the M2 and E1 transitions.

Several further tests of this decay scheme should be possible. As mentioned above, resolution of the L-subshell conversion lines in Np^{236} decay could check the possibility that the transitions are anomalously converted, and may be able to establish the multipolarity of the transitions unambiguously. In Pu^{240} alpha decay, it may be possible to measure the lifetimes of the transitions, a measurable delay ($\gtrsim 10^{-9}$ sec) being possible evidence that they are M2's. It might also be possible to Coulomb-excite the state in U^{236}; if it is in fact an octupole vibration, the excitation function should show an E3 dependence.

H. $Am^{241} \xrightarrow{\alpha} Np^{237}$

The alpha spectrum of Am^{241} has been well studied with magnetic alpha spectrographs, most recently by Baranov et al.[49] Levels of Np^{237} have also been studied in the β^- decay of U^{237},[50-52] and in the electron-capture decay of Pu^{237}.[53] Most of the observed states fit well into rotational bands based on four Nilsson levels.[49,51] The ground state of Np^{237} is 5/2+[642].[54,55,51] The ground state of Am^{241} is 5/2-[523],[56,57,40] and favored alpha decay populates a 59.6-keV state in Np^{237}.

The alpha spectrum in coincidence with γ rays > 500 keV is shown in Fig. 12(a). A surprisingly intense peak, $(1.0 \pm 0.3) \times 10^{-5}$, appears at 4.829 MeV, or 719 keV excitation in Np^{237}. The peak is intense enough for Baranov et al.[49] to have seen in their magnetic spectrograph, had they scanned this energy region. A small shoulder on the low-energy side indicates a second state about 55 keV higher, intensity about 9×10^{-7}. A limit of 3×10^{-7} applies to the population of any higher-lying state.

In coincidence with the 4.83-MeV alpha group (PHW in Fig. 12(a)) is a complex γ-ray peak, consisting of at least two γ rays, Fig. 12(b). In order to better resolve the γ rays, a singles spectrum of an intense sample of pure Am^{241} was studied. Figure 12(c) shows a portion of this spectrum. The peaks at (617±10), (663±5), and (727±10) keV are well established. The peak at 698 keV is probable, as well as necessary to the decay scheme, Fig. 12(d).

The four γ rays are interpreted as depopulating a single state, at (721±5) keV, to the first four levels of Np^{237} at 0, 33, 60, and 103 keV. The large alpha population of the 721-keV level indicates that it

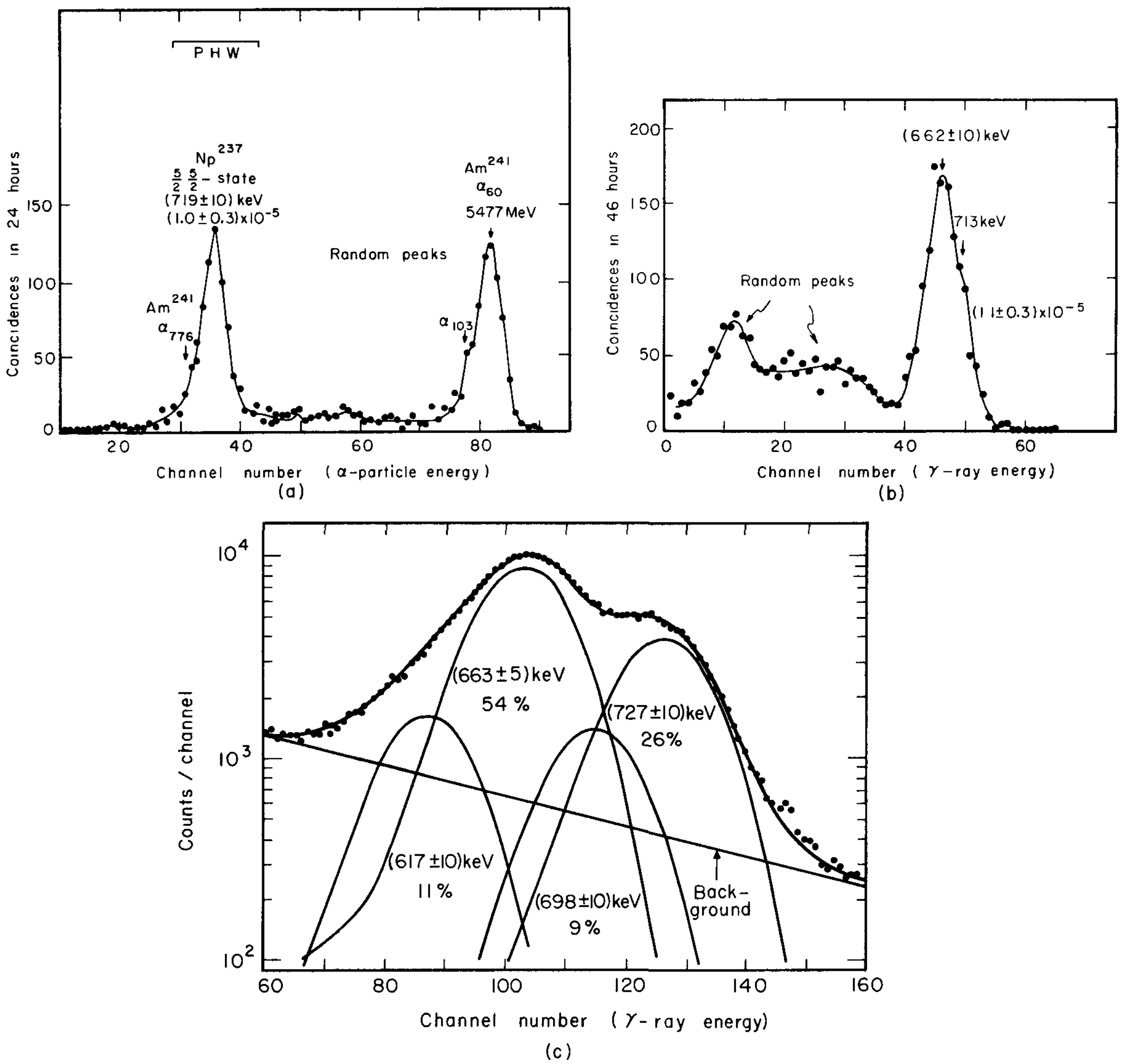

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Fig. 12 a, b, c. The alpha decay of Am^{241}.
(a) Alpha particles in coincidence with γ rays > 500 keV.
(b) Gamma rays in coincidence with the 4.83-MeV alpha group.
(c) A portion of the gamma singles spectrum.

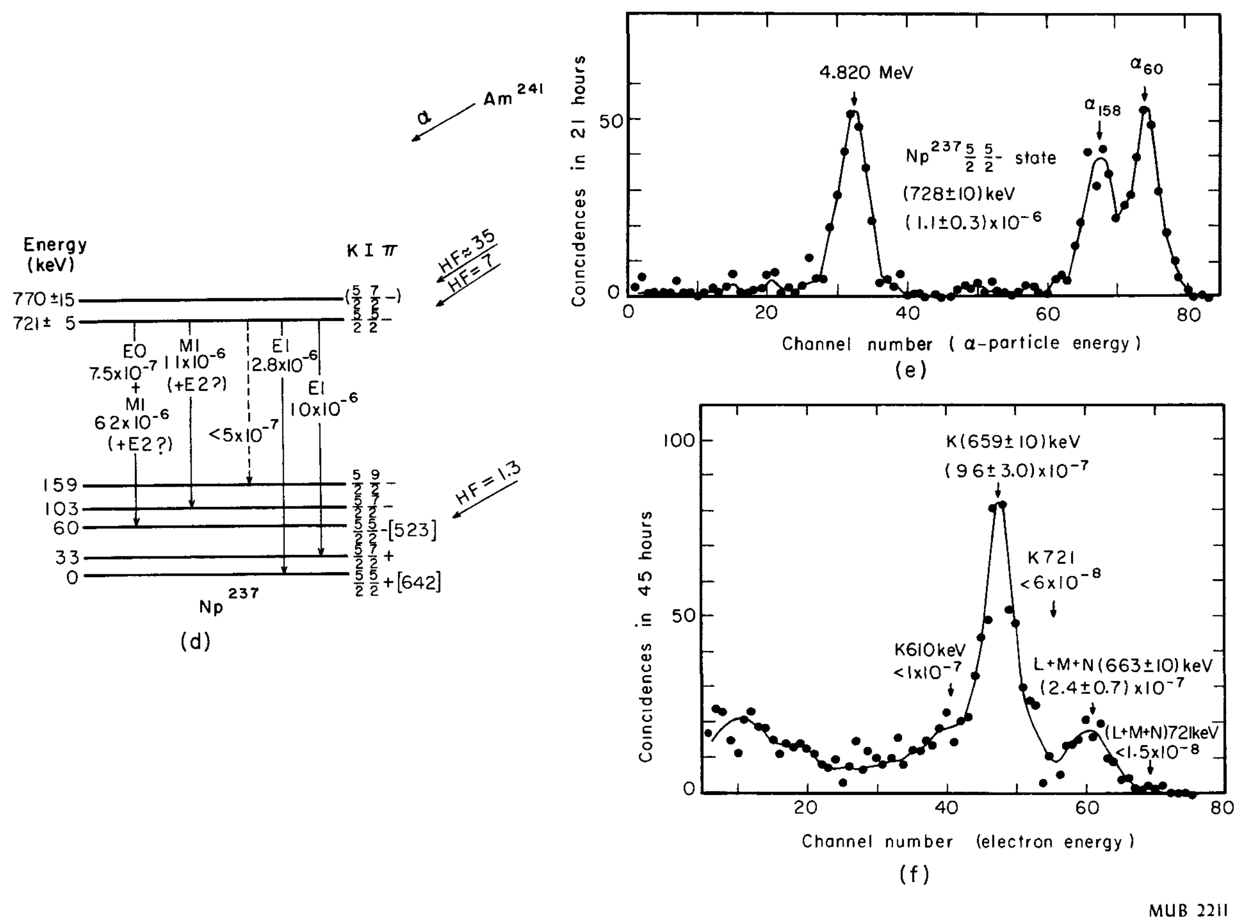

Fig. 12 d, e, f. Alpha decay of Am^{241}.
(d) Decay scheme for Am^{241}. (e) Alpha particles in coincidence with electrons > 380 keV. (f) Electrons in coincidence with 4.83 MeV alpha particles: lithium drifted electron detector.

is probably a 5/2 5/2- beta vibrational state based on the 5/2 5/2-[523] state at 60 keV. The 617- and 663- keV transitions to the lower K=5/2- band are interpreted as M1 transitions. The 698- and 727-keV transitions are assigned E1 multipolarities. The assignment of the lower-energy transitions as M1 instead of E2 contradicts definite predictions of the vibrational model. Presence of the E1 transitions is also unexpected. Some discussion and the justification for the M1 assignment is given in Section V.

Measurements on conversion electrons complete the decay scheme and confirm most of the above hypotheses. Figure 12(e) is the alpha spectrum in coincidence with conversion electrons > 380 keV. The peak at 4.820 MeV, intensity $(1.1\pm0.3)\times10^{-6}$, is identified with the (721 ± 5) keV state. Total population of the state is $(1.2\pm0.3)\times10^{-5}$. The hindrance factor is 7±2, or 5.5±2 relative to the favored alpha transition, and the state decays 88% by γ rays, 12% by electrons.

Because four γ rays de-excite the state, it is essential to examine the conversion lines to confirm the decay scheme. The electron spectrum in coincidence with α_{721} is shown in Fig. 12(f). The predominant features are the K and L+M+N+··· lines of a single transition at (659±10) keV. This is identified unambiguously with the 663-keV γ ray. Apparent conversion coefficients are: $\alpha_K = 0.18\pm0.04$, $\alpha_{L+M+N+\cdots} = 0.045\pm0.015$, K/L+M+N+··· = 4±1. If the γ ray is a pure M1, the transition is therefore (11±4)% E0, but the E0 component may be slightly higher if the transition contains an E2 component also. This E0 transition to the 60-keV state definitely establishes the 5/2- assignment for the 721-keV state.

It follows from Fig. 12(f) that the K-conversion coefficient of $\gamma_{617\pm10}$ is < 10%. This limit is consistent with either an M1 or an E2 assignment. The preference for M1, on other grounds, is discussed in Section V. For the 727-keV γ ray, $\alpha_K < 0.02$, and $\alpha_{L+M+N+\cdots} < 0.005$. This is consistent with the E1 or an E2 assignment, but an E2 multipolarity can be ruled out, because the parity of the 721-keV level is different from the parity of the ground state.

The weak alpha group at about 4.77 MeV is interpreted as the 5/2 7/2- rotational member of the beta band. Its intensity is (9±4)% of the 5/2- member, compared to a ratio of 15% for the corresponding members of the lower-lying K = 5/2- band.

I. $Am^{243} \xrightarrow{\alpha} Np^{239}$

The low-lying levels of Np^{239} and the alpha groups of Am^{243} populating them are quite similar to the corresponding structure observed in Am^{241} decay.[58-61] The ground states of Np^{239} and Am^{243} are 5/2+[642][61,62,40] and 5/2-[523],[63,40] respectively, and favored alpha decay of Am^{243} populates the 5/2-[523] state of Np^{239} at 75 keV.

The alpha singles spectrum of the source employed showed the following composition by activity: Am^{243} 71%, Cm^{244} 12%, Am^{241} + Pu^{238} 12% (mostely Pu^{238}). Coincidence studies have been performed on each of these nuclides, so it was possible to determine which transitions belonged to Am^{243}. Fortunately, the other alpha emitters present did not interfere with the determination of the Am^{243} decay scheme.

The alpha spectrum in coincidence with γ rays > 420 keV, Fig. 13(a), shows a prominent coincidence peak at 4.675 MeV, 676 keV excitation in Np^{239}, intensity $(1.6\pm0.5)\times10^{-5}$. In coincidence with this alpha group is the gamma spectrum in Fig. 13(b). It displays mainly a peak at 654 keV. This peak is interpreted as a γ ray from the 676-keV state to the ground state of Np^{239}, although there is a rather large discrepancy (22 keV) in the energy. The discrepancy is probably due to a shift in the gamma energy calibration which occurred after substitution of a different pulse-height analyzer into the system. The energy of the alpha group was accurately reproduced in three measurements, and this value has been used to determine the excited-state energy.

The inset in Fig. 13(b), top half shows an attempt to resolve the gamma spectrum. There are probably four γ rays leading to the first four levels of Np^{239}. Transitions to the 75- and 117-keV levels, particularly the latter, are so weak that they can not be considered definitely established. The level assignments and transition multipolarities, Fig. 13(c),

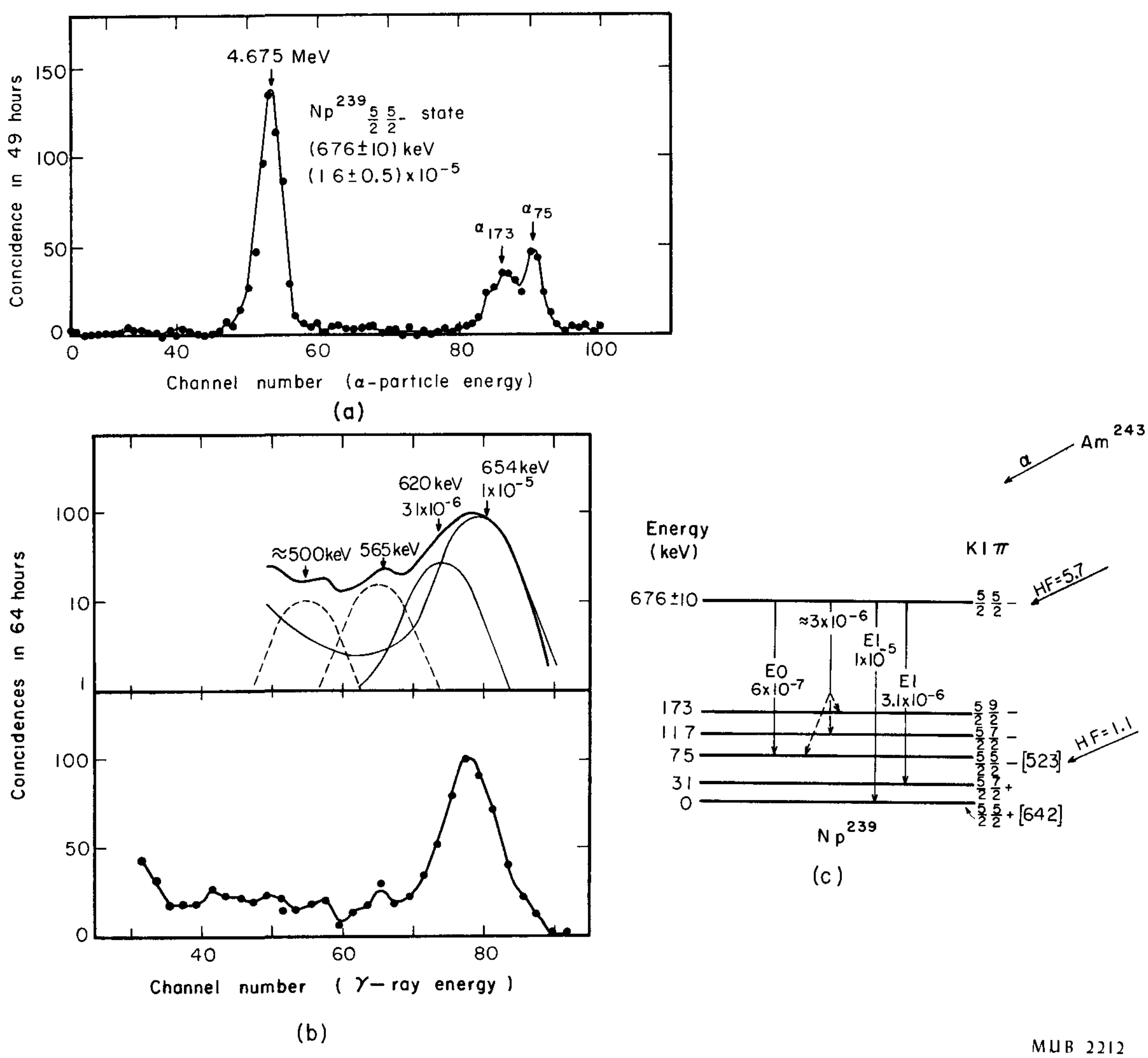

Fig. 13 a, b, c. The alpha decay of Am^{243}.
(a) Alpha particles in coincidence with γ rays > 420 keV.
(b) Gamma rays in coincidence with the 4.67-MeV alpha group. The inset shows an attempt to resolve this spectrum (semilog scale).
(c) Decay scheme for Am^{243}.

are hypothesized here largely by analogy with the decay scheme for Am^{241}. There is no basis in this case for a choice between an M1 and an E2 assignment for the weaker lines.

The alpha spectrum in coincidence with electrons > 350 keV also shows population of the 676-keV state with an intensity of 7×10^{-7}. Total population of the state is $(1.7\pm0.5)\times10^{-5}$; HF=5.7±1.7, or 5.0±1.5 relative to the favored alpha transition. This is good evidence for identifying the 676-keV level as the 5/2- beta vibrational state. Most of the electrons de-exciting the state should undoubtedly be assigned to an E0 transition to the 75-keV level, intensity $(6\pm2)\times10^{-7}$.

J. $Cm^{242} \xrightarrow{\alpha} Pu^{238}$

The previous study of this decay scheme was made with a source containing about 2% Cm^{243}.[16] In the experiment reported herein a much purer Cm^{242} sample was used; this simplified the spectra considerably. The earlier assignment of transitions to Cm^{242} and Cm^{243} is seen to be correct, and some new information can be added.

Levels of Pu^{238} have previously been observed in the electron-capture decay of Am^{238},[69] and in the β^- decay of Np^{238}, most recently by Albridge and Hollander[65] and by Borggreen et al.[66] The levels include 943.1 keV (00+), 984.5 keV(02+), 1030 keV(22+), and 1071 keV(23+). High-energy gamma spectra[7,8,67] of Cm^{242} show transitions of 562, 605, and 890 keV; conversion-electron measurements[7,8] show a transition at (941.4±2) keV. The 890-keV γ ray and the 941-keV electron transition are interpreted as an E2 and an E0 transition de-exciting the 943-keV 00+ state to the ground and first excited states. The 562- and 605-keV γ rays are characterized as E1 transitions depopulating a 01- state at 605 keV.

Figure 14(a) shows the alpha spectrum in coincidence with γ rays > 350 keV. Two alpha groups in true coincidence populate states at (950±20) keV with an intensity $(2.5\pm0.8)\times10^{-7}$, and (613±20) keV with intensity $(2.6\pm0.7)\times10^{-6}$. These are interpreted as the 943-keV 00+ state and the 605-keV 01- state. Only the transition to the 00+ state

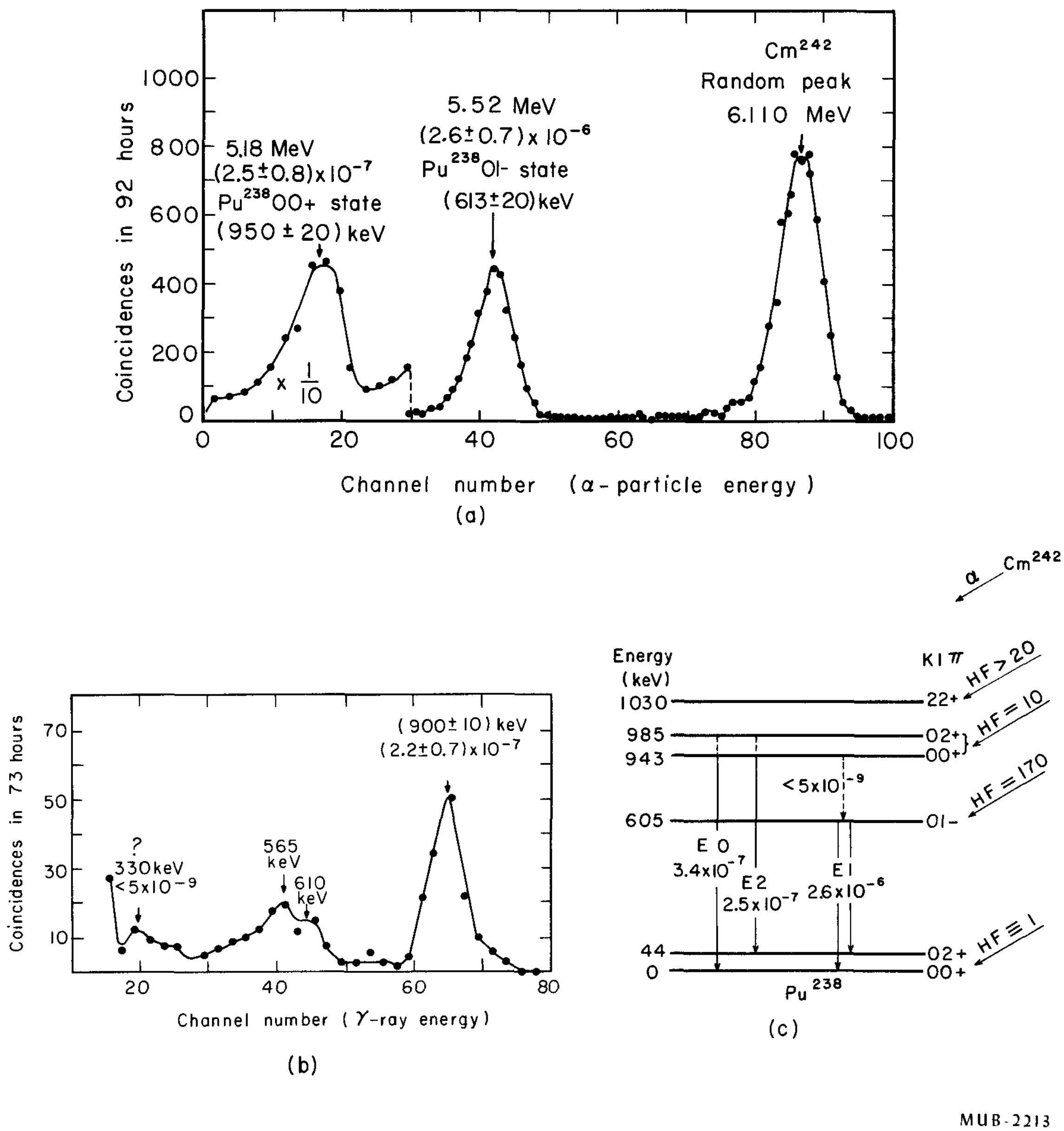

Fig. 14 a, b, c. Alpha decay of Cm^{242}.
(a) Alpha particles in coincidence with γ rays > 350 keV.
(b) Gamma rays in coincidence with 5.17-MeV α particles.
(c) Decay scheme for Cm^{242}.

was observed in coincidence with electrons, intensity $(3.4\pm1.0)\times10^{-7}$. Absence of the 605-keV state sets an upper limit of 0.01 on the conversion coefficient for γ rays de-exciting this state, confirming their E1 assignment and the 1- assignment for the state.

Total population of the state at 950 keV is $(6\pm2)\times10^{-6}$, HF=10±3 (assuming that the 02+ state at 985 keV is not appreciably populated). For the 01- state, HF = 170±50.

The γ-ray spectrum in coincidence with α_{950}, Fig. 14(b) shows predominantly the expected (900±10)-keV E2 γ ray. Weak γ rays in the region of 580 keV are probably due to overlap of the stronger α_{613} group in the pulse-height window. The possible 335-keV E1 transition connecting the 00+ and 01- states is absent or very weak; a limit of 1% of the total population of the 00+ state applies to this transition.

No alpha decay to the 22+ state at 1030 keV was observed, and a lower limit of 20 can be set on the hindrance factor for this transition. The decay scheme is shown in Fig. 14(c).

K. $Cm^{243} \xrightarrow{\alpha} Pu^{239}$

Levels of Pu^{239} have been observed in the electron-capture decay of Am^{239},[68,64] and in the β^- decay of Np^{239}.[61,69-72] Most of these levels have also been observed in the alpha decay of Cm^{243}.[73] As previously mentioned, the ground state of Pu^{239} is 1/2+[631]. Favored alpha decay of Cm^{243} populates a 286 keV level of Pu^{239}. This level and the ground state of Cm^{243} have been assigned 5/2+[622].[61,40]

Samples of curium prepared by prolonged neutron irradiation of Am^{241} contain Cm^{243} and Cm^{244}, in roughly equal abundance by activity, after the Cm^{242} has decayed away. Analysis of the alpha and gamma spectra of the source used in this investigation gave the following composition by activity: Cm^{243} 50%, Cm^{244} 45%, Cm^{242} 4%, and Pu^{238} 1%.

The primary feature of the alpha spectra in coincidence with gamma rays (Fig. 15(a)) and electrons (not shown), is a complex peak at 5.30 MeV. Its intensity is $(4.0\pm1.2)\times10^{-5}$ in coincidence with γ rays, and $(5\pm2)\times10^{-6}$ in coincidence with conversion electrons. It was possible

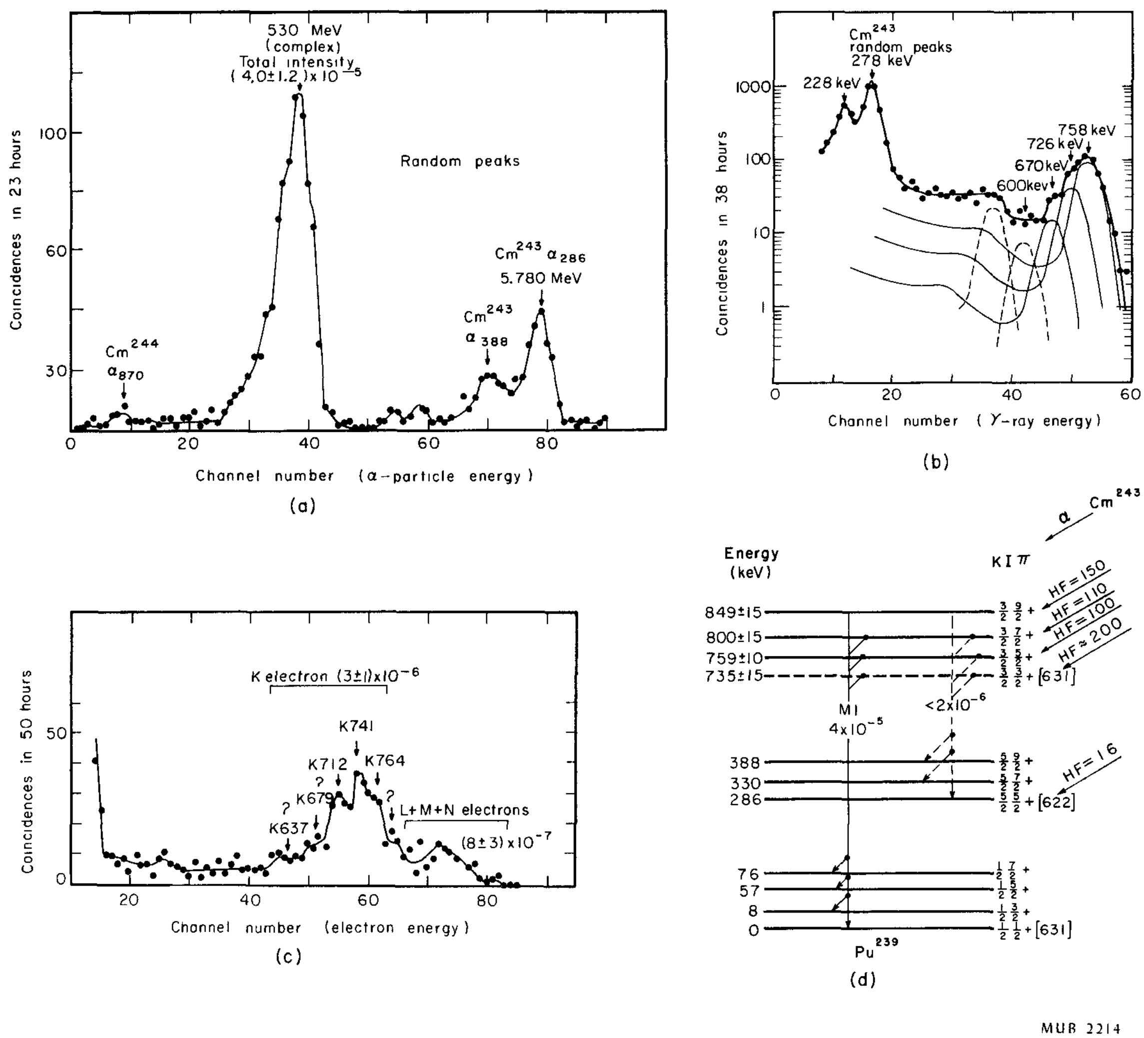

Fig. 15 a, b, c, d. Alpha decay of Cm^{243}.

(a) Alpha particles in coincidence with γ rays > 380 keV.

(b) Gamma rays in coincidence with the complex 5.30-MeV alpha peak (semilog scale). The single γ-ray peaks drawn in probably represent only a partial resolution of the spectrum.

(c) Electrons in coincidence with the complex 5.30-MeV alpha peak (lithium-drifted electron detector).

(d) Decay scheme for Cm^{243}.

to partially resolve the complex peak into four components, each in the α-γ spectrum and in the α-e^- spectrum. The information obtained is recorded in Table III.

The gamma spectrum in coincidence with the complex 5.30-MeV alpha peak, Fig. 15(b), is very complex. The four states de-excite to members of the ground-state band of Pu^{239} by numerous γ rays in the energy region 600 to 760 keV. Undoubtedly the peaks drawn into the γ-ray spectrum do not represent all the transitions, since many of the possible ones are too closely spaced to be resolved. The intensity of the gamma spectrum in the 400- to 500-keV region is somewhat too high to be accounted for by Compton tails of the higher-lying peaks. Possibly there are weak transitions to members of the K = 5/2+ band.

The electron spectrum in coincidence with the complex 5.30-MeV alpha peak is shown in Fig. 15(c). The K peaks resemble the ≈ 750-keV gamma peaks of Fig. 15(b), although the resolution is better. Low-energy tailing, caused by backscattered electrons, almost masks the weaker electron lines around 670 keV. The similarity between the electron and γ-ray spectra in coincidence with the 5.30-MeV alpha groups indicates that the many transitions involved have similar conversion coefficients. On this assumption, the measured conversion coefficients are $\alpha_K = 0.08 \pm 0.03$, $\alpha_{L+M+N+\cdots} = 0.020 \pm 0.006$. The transitions have accordingly been assigned M1.

In the last column of Table III, the four states observed are designated as members of a K = 3/2+[631] Nilsson band. Some of the reasons for this are:

1. The energy of the band agrees well with the expected energy for a "hole" state of that assignment.[72] The same assignment has been given to a state[74] occurring at 312 keV in the level spectrum of U^{233}.
2. The energy spacings of the states agree satisfactorily with predictions for a K = 3/2 band with a rotational constant $\frac{\hbar^2}{2\Im} = 5.6 \pm 0.5$.

Table III. Resolution of the complex 5.30-MeV alpha peak of Cm^{243}.

Energy of the alpha group (MeV)	Excitation energy in Pu^{239} (keV)	Energy separation from α_{759} (keV)	Intensity per Cm^{243} α decay	Hindrance factor	Assignment			
					K	I	π	$[N n_z \Lambda]$
5.338±0.015[a]	735±15	-24±10	$(1.3^{+1.1}_{-0.6})\times10^{-5}$	200^{+200}_{-100}	3/2	3/2	+	[631]
5.315±0.010	759±10	———	$(2.1\pm1.0)\times10^{-5}$	100±50	3/2	5/2	+	
5.274±0.015	800±15	41±5	$(1.0\pm0.4)\times10^{-5}$	110±40	3/2	7/2	+	
5.226±0.015	849±15	90±5	$(3.9\pm1.6)\times10^{-6}$	150±60	3/2	9/2	+	

[a] This alpha group is regarded as questionable, because it is too close to α_{759} to be resolved.

3. The observed M1 transitions to members of the K = 1/2+ [631] band probably fix the K quantum number at 1/2 or 3/2; otherwise these transitions would be K-forbidden. Possible weak transitions to members of the K=5/2+[622] band are also K-allowed.
4. The observed M1 transitions are also allowed by selection rules for asymptotic quantum numbers.[40] The same applies to the possible weak transitions to members of the K = 5/2+[622] band.
5. The relative intensities of the alpha groups populating members of the band are in fair agreement with the values predicted for an $\ell = 2$ alpha wave.
6. Hindrance of the alpha transitions is reasonably low, as can be expected for the odd-neutron transition 5/2+[622]→3/2+ [631].[75,76]

The decay scheme is shown in Fig. 15(d). Besides the groups discussed above, the α-e^- spectrum gives evidence for a possible alpha group at 5.11 MeV, intensity limit, 1×10^{-7}. Another group at 4.94 MeV belongs predominantly to Cm^{244} decay (see Section L); the same limit 1×10^{-7} applies to a possible Cm^{243} group at this energy. Below 4.92 MeV, there are no groups with an intensity 5×10^{-8}.

A study of α particles in coincidence with γ rays > 785 keV failed to detect any transitions more intense than 5×10^{-7}, although there were several doubtful alpha groups below this intensity limit. There are no γ rays over 900 keV in coincidence with α particles with an intensity greater than 3×10^{-7}.

In summary, four alpha groups were identified, and from the data they were assigned as members of a 3/2+[631] Nilsson band. No other alpha groups of Cm^{243} were positively identified. Within the limits stated, there is no alpha decay to a state distinguished by low hindrance factor or strong monopole de-excitation (or both) that might therefore be characterized as a beta vibration.

L. $Cm^{244} \xrightarrow{\alpha} Pu^{240}$

The β^- decay of the 7.3-min Np^{240} populates many states of Pu^{240}, including a 01- state at 597 keV, 00+ and 02+ states at 858 and 900 keV, and a 22+ state at 942 keV.[77] As previously mentioned, the alpha spectrum of Cm^{244} has been studied in a magnetic spectrograph, and alpha groups populating states at 863 keV, $(1.55\pm0.16)\times10^{-6}$, and 903 keV, $(0.50\pm0.08)\times10^{-6}$, were observed.[6] These states were identified with the 00+ and 02+ states mentioned above.

Alpha-gamma and alpha-electron coincidence spectra showed population of levels at (610±20) and (870±15) keV, intensities $(1.1\pm0.2)\times10^{-6}$ and $(2.3\pm0.5)\times10^{-6}$. The group α_{610} is identified with the 01- state, although the peak shape may indicate some population of the 03- member of the octupole band. The group "α_{870}" is identified with the doublet $\alpha_{863} + \alpha_{903}$ mentioned in the last paragraph.

The gamma spectrum in coincidence with "α_{870}" shows not only the expected E2 transition at 825 keV, but also 262-, 570-, and 610-keV transitions, indicating cascade decay via the 01- state, such as occurs in Th^{228}. The conversion coefficient of the 262-keV transition, measured by a comparison of intensities in the cascade, is 0.15±0.08, which is unambiguous evidence that the transition is E1. A γ-γ coincidence experiment confirms the existence of the cascade, which has also been observed in Np^{240} decay to Pu^{240}.[77]

An alpha spectrum in coincidence with the gamma radiation around 260 keV, taken at higher resolution, shows clearly the doublet to the 00+ and 02+ state. In addition to confirming the doublet nature of the peak, this shows that the 02+ level also decays partly via the octupole band, as would be expected.

The alpha-electron coincidence spectrum shows only one peak more intense than 1×10^{-8}, populating the K = 0 + band, intensity $(9.5\pm2)\times10^{-8}$. Most of this intensity, $(8\pm2)\times10^{-8}$, is assigned to an E0 transition to the ground state. Failure to observe α_{610} in the α-e^- spectrum confirms the E1 assignment for transitions from the 610-keV state, and the 1- assignment for the state.

The E0, E2, and E1 transitions de-exciting the 00+ state were previously observed in the β^- decay of Np^{240}.[77] The relative intensities agree well with the values reported here.

The hindrance factor for the 01- state is 100±20; for the 00+ and 02+ states, HF = 3±0.5, and HF = 5±2, respectively. No decay was observed to the 22+ state at 942 keV, which sets a lower limit of 100 on the hindrance factor for α decay to this state.

All this information is summarized in Fig. 16.

M. $Cf^{250} \xrightarrow{\alpha} Cm^{246}$

The source for this investigation had the following composition by alpha activity: Cf^{250} 96%, Cf^{252} 3.6±0.4%, Cf^{249} 0.4±0.1%. The alpha spectrum in coincidence with electrons > 380 keV showed no groups in true coincidence with an intensity $>1.2\times10^{-7}$.

Alpha-gamma coincidences were studied by use of the fission anticoincidence technique to reduce the background from Cf^{252} fissions. Weak alpha peaks appeared in coincidence with γ rays > 460 keV at (5.51±0.02) and (5.24±0.04) MeV, intensities 3×10^{-6} and 1×10^{-6}, respectively, per Cf^{250} decay. The 5.51-MeV group was observed to decay partly by a γ ray of about 500 keV, intensity about 1×10^{-6}. This could represent a state of Cm^{246} at about 510 keV. However, it could also be a state of Cm^{245}, populated by the decay of Cf^{249} with an intensity of about 5×10^{-4} per Cf^{249} alpha decay. The 5.24-MeV alpha group is in coincidence with a complex gamma spectrum in the region 500 to 900 keV, indicating that it probably belongs to the decay of Cf^{249}.

With the possible exception of the weak group at 5.51 MeV, there are no alpha transitions to states of Cm^{246} that are above ≈ 500 keV and decay to the ground or first excited state with an intensity $> 8\times10^{-7}$.

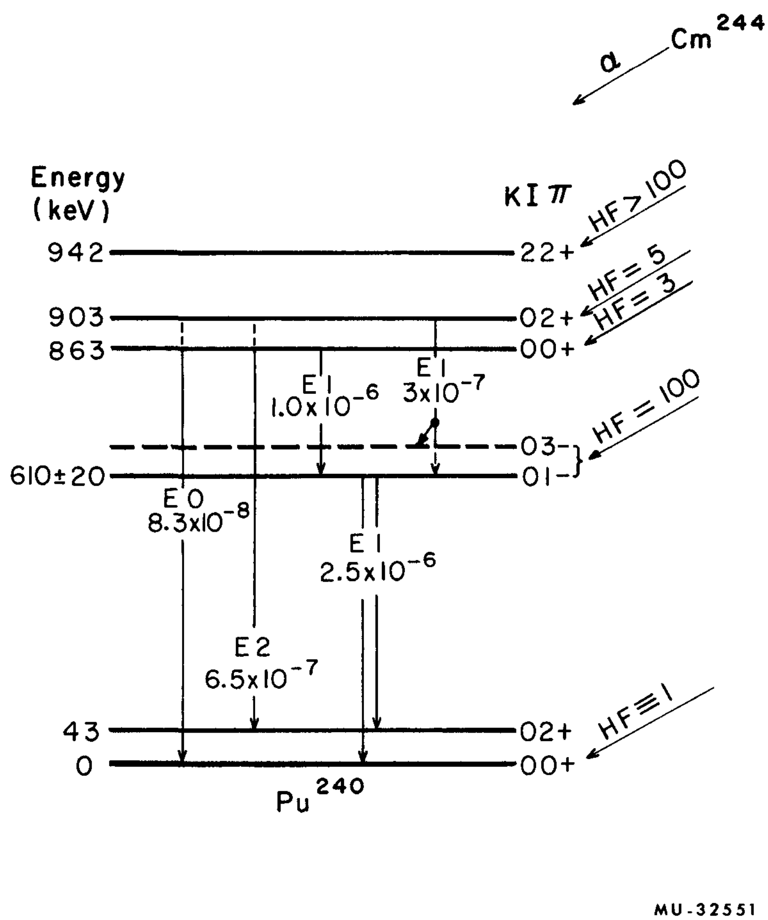

Fig. 16. Decay scheme for Cm^{244}.

N. $Cf^{252} \xrightarrow{\alpha} Cm^{248}$

Previous coincidence measurements set a limit of 6×10^{-7} on alpha's coincident with electrons > 400 keV. No groups were observed in coincidence with γ rays > 225 keV; intensity limit, 2×10^{-5}.

When the fission anticoincidence technique was used, no alpha groups more intense than 8×10^{-7} were observed in coincidence with γ rays > 480 keV. The fission anticoincidence counter thus reduced the background by a factor of about 20.

In a search for the 06+ member of the ground-state band, a γ ray was observed at (160±15) keV in coincidence with 5.7- to 6.0-MeV α particles, intensity $(2.0\pm0.6)\times10^{-5}$. This is identified with the $6+ \xrightarrow{E2} 4+$ transition. The theoretical conversion coefficient for this transition is of 3.3; based on this value, alpha decay populates the 06+ state at (203±15) keV with an intensity $(6.6\pm2.2)\times10^{-5}$, HF=1170±350.

O. $E^{253} \xrightarrow{\alpha} Bk^{249}$

The primary source of information about the levels of Bk^{249} is the alpha decay of E^{253}. Most recent studies[78,79] reveal a large number of levels, many of which have been assigned to rotational bands built on the Nilsson states 7/2+[633], ground state; 3/2-[521], 8.8 keV; and 5/2+[642], 393 keV. Favored alpha decay of E^{253} populates the ground state with an abundance of 90%.

In my experiments, five alpha spectra were taken in coincidence with γ rays, gating at different gamma energies. These show a complicated structure in the region above 600 keV excitation in Bk^{249}. Below this energy, the spectrum of weak alpha groups is masked by more intense transitions to members of the K = 5/2+ band, which decay by prompt M1 transitions to members of the ground-state band. Figure 17 shows one of the alpha-gamma spectra.

Four different gamma spectra were taken in coincidence with α particles, gated on different regions of the alpha spectrum (see PHW's in Fig. 17). Since many alpha groups were only partially

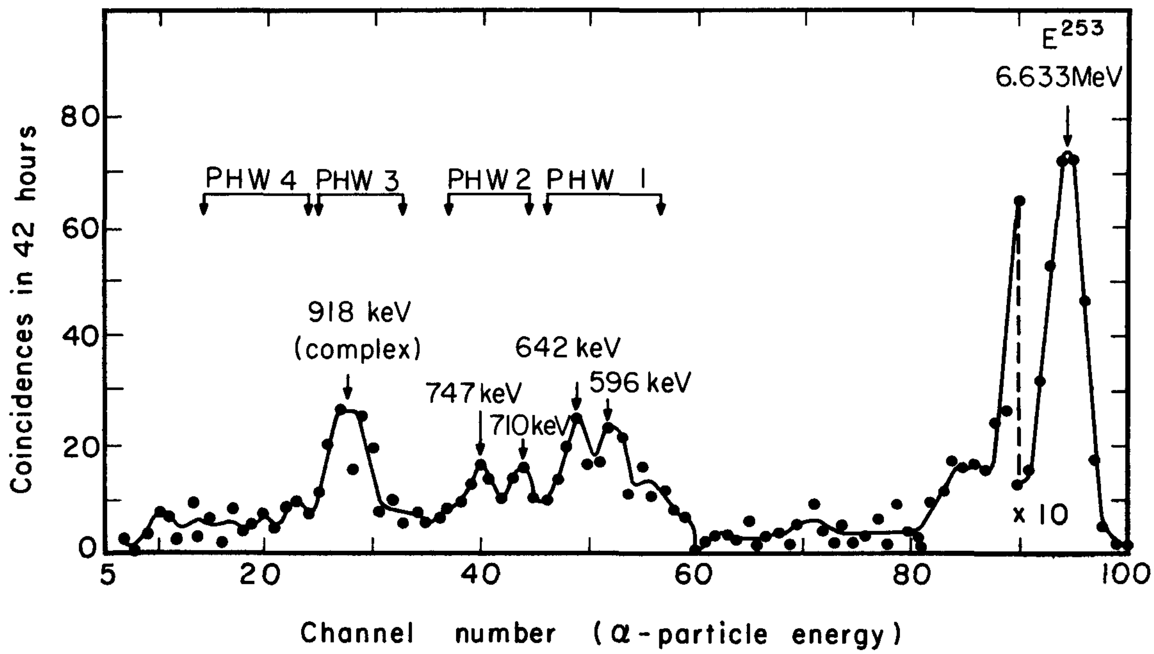

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Fig. 17. E^{253} alpha spectrum in coincidence with γ rays > 570 keV

resolved, each of the gamma spectra represents the de-excitation of a number of states; nevertheless, considerable information was gained about the de-excitation of some of the states.

A single alpha spectrum in coincidence with electrons > 500 keV contained no peaks more intense than 3×10^{-8}. There is thus no indication that any of the states observed decays by strong monopole transitions.

Table IV lists the states observed and the information obtained about them. A state at (617±10) keV, populated with an intensity $(2.4\pm1.0)\times10^{-6}$ in coincidence with γ rays, is assigned as the 13/2+ member or the K = 5/2+[642] rotational band. Allowing for M1 conversion of the γ rays de-exciting this state to members of the ground-state band, one finds the total alpha population is $(3.4\pm1.4)\times10^{-6}$. Asaro has predicted a group populating this state at about 610 keV with an intensity of about 5×10^{-6}. The other levels listed in Table IV have not been assigned, because the data are not sufficient to make unambiguous choices among the many Nilsson states in this region.

The hindrance factors listed in the table are all high with the exception of the value 60 for the 921-keV level. Relative to the least hindered transition, this hindrance factor is only 30. However, the conversion coefficient for the (895±15)-keV γ ray de-exciting the state is < 3%. If this transition is to be predominantly E2, rather than E1, it can have an E0 component no greater than 1%. This rules out the possible interpretation of the 921-keV state as a beta vibration, in spite of the relatively low hindrance factor. As noted in the table, the alpha transition to this "state" appears to be complex, so that the actual hindrance factor could be higher by a factor of 1.5 or so.

An alpha spectrum of E^{253} has shown the existence of a group[79] at 6.39 MeV, 250 keV excitation in E^{253}, intensity about 2×10^{-5}. It was thought that this might be the 7/2-[514] Nilsson level, which should decay to the ground-state band by E1 transitions. Table V gives the negative results of a search for this alpha group in coincidence with

Table IV. States of Bk^{249} observed in the alpha decay of E^{253}.

Excitation energy in Bk^{249} (keV)	Intensity per E^{253} α decay in coinc. with γ rays	Hindrance factor	De-excitation: Gamma rays, energy (keV)	De-excitation: Conversion coefficient	De-excitation: Assignment	Assignment of state
617±10	$(2.4\pm1.0)\times10^{-6}$	550±250	Not measured	Not measured	(M1)	5/2 13/2 +[642]
(600±10)	$(6\pm3)\times10^{-7}$ [a]	4000±2000	Complex spectrum of γ rays. Possible transitions: 556, 585, 640 keV. No γ rays > 640 keV.	Not measured		
(642±10)	$(2\pm1)\times10^{-7}$ [a]	7000±3500				
(687±10)	$(3\pm1.5)\times10^{-7}$ [a]	2800±1400				
(712±10)	$(2\pm1)\times10^{-7}$ [a]	3000±1500				
748±10	$(2.7\pm0.8)\times10^{-7}$	1400±400	746±10	< 0.04	E2, or E1	
((790))	4×10^{-8}	6000	799±10			
921±10 (complex)	$(8\pm3)\times10^{-7}$	60±20	895±15 ((920))	< 0.03	E2, or E1	
((966))	1×10^{-7}	240	Not observed	< 0.10		
((1010))	1×10^{-7}	120		< 0.10		
((1060))	4×10^{-8}	180		< 0.40		
((1110))	8×10^{-8}	50		< 0.15		

[a]The intensities of these transitions may be higher if the states decay partially by γ rays < 500 keV. No parentheses around the energy means that an α or γ transition is definitely established. Single parentheses means that there is some intensity, but the peak position is uncertain. Double parentheses indicate that the transition is doubtful.

Table V. Limit on possible coincidences between 6.39-MeV α particles and prompt gamma radiation (< 0.06μsec).

Energy of coincident γ radiation (keV)	Limit per E^{253} α decay
125 to 170	2×10^{-6}
170 to 200	5×10^{-7}
200 to 300	1×10^{-7}

gamma radiation. It is concluded that the alpha group does not populate the expected 7/2- state. It could be an alpha branch from the β^- emitter E^{255} or the 37-hour β^- emitter E^{254}, or an E^{253} alpha group in coincidence with delayed or highly converted transitions.

P. $Fm^{254} \xrightarrow{\alpha} Cf^{250}$

No alpha groups are observed in coincidence with high-energy radiation. The alpha spectrum in coincidence with electrons > 300 keV establishes an intensity limit of 1×10^{-7} on possible transitions. In coincidence with γ rays > 400 keV, a limit of 1×10^{-6} is established.

A 1032-keV 22+ state of Cf^{250} has been observed in the β^- decay of Bk^{250}.[80] From my results, the hindrance factor for alpha decay to this state is > 18.

Alpha decay to the 06+ member of the ground-state band was observed in another experiment. It decays by a γ ray at (151±5) keV to the 04+ state with an intensity of $(1.0\pm0.3)\times10^{-5}$. This places the 06+ state at (290±5) keV. The theoretical conversion coefficient for this E2 γ ray is 3.95. Assuming that value, one finds the total alpha intensity to the 06+ state is $(4.0\pm1.2)\times10^{-5}$, or HF = 1300±400.

V. DISCUSSION

Interesting features of the level structure and decay schemes for some of the nuclides studied are considered in this section. Systematics of the levels is considered in Section VI.

Levels of Rn^{218}

This nucleus falls into the near-harmonic region — deformable nuclei that have low-energy vibrational structure, but whose equilibrium shape is thought to be spherical. Although there is expected to be an abrupt change in structure when one goes from spherical to axially symmetric nuclei, there is considerable evidence that the properties of some levels vary smoothly across the boundary around neutron number 135.[23] An example of this is the behavior of the 1- states (see Fig. 19). Particularly interesting are the relative intensities of the E1 transitions de-exciting these states to the lower-lying 0+ and 2+ states. For a nucleus with axial symmetry, the intensity ratio, after correction for the third-power energy dependence of the E1 transition rate, depends only on vector addition coefficients.[81] For the K = 0 states, the predicted ratio is

$$\frac{B(E1, 1- \rightarrow 2+)}{B(E1, 1- \rightarrow 0+)} = 2.$$

For nonspherical nuclei, all measured values of this ratio are within experimental error of 2. Surprisingly, the rule appears to hold also for spherical nuclei, for which K is not a good quantum number.

If the 1- assignment for the 792-keV state of Rn^{218} is correct, this is the first case in which the rule breaks down; the measured ratio is 11±3. It should be noted, however, that if the assignment is wrong, and the 846-keV level is the 1- state, then the ratio is 3.2±0.9, close to the value found for other 1- states.

Levels of U^{234}

As mentioned in the introduction, γ-ray singles and electron—K x-ray coincidence spectra established the existence of 0+ states in

U^{234} and Pu^{238}, excited by the alpha decay of Pu^{238} and Cm^{242}. In both cases, the 0+ states were characterized by low hindrance factors ($\lesssim 20$) and de-excitation to the first two members of the ground-state band by E0 and E2 transitions of roughly equal intensity. These characteristics have been used to define a "regular beta vibrational state."[16]

A surprising result of my work is the observation that the 02+ member of the beta band in U^{234} is poorly populated in alpha decay. It seems more reasonable to expect the relative population of the 00+ and 02+ members of the beta band to be similar to the ratio for alpha decay to the corresponding members of the ground-state band. For comparison of different nuclides, including odd-mass ones, it is convenient to define the quantity

$$R_\alpha(\beta/f) = \frac{W_\alpha(\text{2nd member of beta band})/W_\alpha(\text{1st member of beta band})}{W_\alpha(\text{2nd member of favored band})/W_\alpha(\text{1st member of favored band})},$$

where the W_α's are the alpha intensities to the states in parenthesis and the favored band is the band containing the state to which alpha transitions are only slightly hindered, i.e., the "base" state for a beta vibration observed in alpha decay.

For the beta band of U^{234}, $R_\alpha(\beta/f) < 0.26$, compared with an expected value of ≈ 1.

<u>Levels of Th^{228}</u>

The 02+ member of the beta band in Th^{228} also is populated with unexpectedly low intensity. This state has never been observed, but it must certainly lie about 58 keV above the 00+ state. For this nucleus, $R_\alpha(\beta/f)$ is again < 0.26.

The 00+ state of Th^{228} de-excites about 90% by an E1 γ ray to the lower-lying 01- "octupole vibrational state." Although this 497-keV E1 transition has an estimated single-particle lifetime much shorter than the competing E2 transition to the 58-keV 02+ state, it is forbidden by the vibrational model. The lifetimes of the states are not measured in my experiment, but one does obtain a useful parameter for comparison of different cases, the ratio of the reduced transition probabilities:[82]

$$\frac{B(E1)}{B(E2)} = 1.05\times10^{-6}\ A^{2/3}\ \frac{(E_{E2})^5}{(E_{E1})^3}\ \frac{W(E1)}{W(E2)}\ , \qquad (1)$$

where A is the atomic number of the nucleus,

E_{E2} and E_{E1} are the energies of the competing transitions, in MeV,

and $W(E1)/W(E2)$ is the experimentally observed intensity ratio for the two transitions.

In this case, $\frac{B(E1)}{B(E2)} = (1.8^{+\infty}_{-0.9})\times10^{-3}$. Values of B(E2) measured in the Coulomb excitation of other beta vibrational states all fall in the range 1 to 5 single-particle (s. p.) units.[34,83] The plausible assumption that this holds roughly true in all cases makes it possible to estimate the absolute transition probability for the E1; in this case, $B(E1) \gtrsim 10^{-3}$ s. p. units. Since E1 transitions in heavy elements are quite generally retarded by at least two or three orders of magnitude, this becomes a rather fast E1. Nevertheless, since the matrix elements are small, the presence of this E1 is not necessarily in serious disagreement with the vibrational model. A small impurity in the wave function of either the 00+ (beta vibrational) or the 01- state could account for the relative speed of the transition.

It was mentioned in Section IV.B that the beta vibrational band of Th^{228} was expected to lie at about 520 keV,[31] on the basis of measured distortion of the ground-state rotational band, presumably caused by vibration-rotation interaction. There are now indications that the ground-state band and beta band interact more strongly than previously expected.[83,84] For this reason, it is not surprising to find the beta band of Th^{228} at a higher energy.[84]

It is nevertheless interesting to consider the significance of failure to observe any alpha population of a hypothetical state. One possible approach is based on the fact that the outgoing α particle can excite the nucleus by electromagnetic interactions when it is outside the range of specifically nuclear forces. This process is closely analogous to Coulombic excitation by accelerated ions, and unlike that for nuclear interactions, the probability for Coulomb excitation can be calculated precisely (I am indebted to Dr. John A. Rasmussen for showing me this calculation), as follows:

The motion of a backscattered ion in Coulomb excitation is symmetrical about t = 0, the time of closest approach. An α particle emerging from a decaying nucleus travels the second half of this trajectory, so that the excitation amplitude is in this case just half that calculated for an α-particle projectile, with the same c.m. energy, in a 180° orbit. The excitation cross section, which is proportional to the square of the amplitude, is just 1/4 the value calculated for a projectile, or (for an electric excitation)[85]

$$\sigma_{E\lambda} = \left(\frac{z}{\hbar v_i}\right)^2 a^{-2\lambda+2} \frac{df_{E\lambda}(180°, \xi)}{d\Omega} B(E\lambda)$$

where

$$a = \frac{2ze^2}{m_0 v_i v_f},$$

z and m_0 are the charge and reduced mass of the daughter nucleus,

v_i and v_f are the velocities of the α particle at radius ∞ before and after exciting the nucleus,

$B(E\lambda)$ is the reduced transition probability for the electric transition of multipole order λ,

$$\xi = \frac{2ze^2}{\hbar}\left(\frac{1}{v_f} - \frac{1}{v_i}\right),$$

and $\frac{df_{E\lambda}}{d\Omega}$ is a tabulated function.

The probability that a given alpha decay will excite the nucleus is obtained by dividing this expression by the Rutherford cross section:

$$\frac{W_{\alpha'}(f)}{W_{\alpha}(i)} = \left(\frac{2e}{\hbar v_i}\right)^2 a^{-2\lambda} \frac{df_{E\lambda}(180°, \xi)}{d\Omega} B(E\lambda, i\rightarrow f) . \qquad (2)$$

Consider the hypothetical 517-keV 0+ state of Th^{228}. Thirty-two percent of U^{232} alpha decays populate the 58-keV 2+ level of Th^{228}. The emerging α particle can then excite the daughter nucleus from this state to the 0+ state by an E2 interaction. From formula (2), the fraction of decays to the 2+ state that will be followed by further E2 excitation is

$$\frac{W_\alpha(0+')}{W_\alpha(2+)} = 5.4\times10^{60} B(E2,\ 2+ \rightarrow 0+') .$$

The experimental limit on alpha branching to the hypothetical 0+ state is 5×10^{-8}, or

$$\frac{W_\alpha(0+')}{W_\alpha(2+)} < \frac{5\times10^{-8}}{0.32} = 1.56\times10^{-7} .$$

Therefore,

$$B(E2, 2+ \rightarrow 0+') < 2.9\times10^{-68}\ cm^4 = 3\ s.p.\ units.$$

The calculation shows that there is no state at 517 keV that has an E2 transition probability to the 2+ member of the ground-state band greater than 3 s.p. (single-particle) units. Since beta vibrational states have E2 transition rates of just this order, the calculation does not rule out the existence of a 517-keV beta vibration.

More practically, the calculation shows that the excitation of 0+ states in alpha decay is not due to Coulomb interaction occurring when the α particle is outside the nucleus. For the 825-keV state of Th^{228}, as well as the other 0+ states found in this work, Coulomb excitation accounts for less than 1% of the alpha population.

Levels of U^{232}

A low hindrance factor and strong monopole decay place the 693-keV 0+ state of U^{232} in the class of "regular" beta vibrations. However, alpha decay fails to populate the 735-keV 2+ member of the band, as was also observed for the beta bands of U^{234} and Th^{228}. In this case, favorable circumstances made it possible to set a low limit:

$$R_\alpha(\beta/f) < 0.075.$$

The 00+ state was not observed to decay by E1 transitions to the 565-keV 01- state. The ratio of reduced transition probabilities, B(E1)/B(E2), is less than 1×10^{-4}, at least an order of magnitude lower than the value for the corresponding transitions in Th^{228}.

Levels of Np^{237}

The Am^{241} α-γ coincidence spectrum gives evidence for population of the second member of the beta band. The measured value of $R_\alpha(\beta/f)$ is 0.6±0.3.

The 5/2- member of the beta band decays partly by γ-ray transitions to the first two members of the 5/2-[523] band at 60 and 103 keV. The relative intensities of these transitions should depend only on the transition energies and the appropriate vector addition coefficients.[81] The second column of Table VI lists the experimental intensities, normalized to a sum of 1. Columns 3 and 4 give the theoretical intensities for M1 and E2 transitions. The agreement is reasonable for M1's, but it is unsatisfactory for E2's.

It is well known that mixing of two rotational bands (in this case the beta band and the band on which it is based) can have a considerable effect on the relative intensities of transitions between them. Calculations have been carried out for the effect of mixing on the E2 branching ratios from 22+ states,[86,87,66] and more recently, for E2 branching from gamma vibrational states of odd-mass nuclides.[9]

It is not difficult to do a semi-empirical calculation for the beta vibrational bands. (I am indebted to Dr. Frank S. Stephens, Jr., for showing me these calculations.) If one assumes Coriolis mixing, the amplitude of the beta vibrational state of spin I admixed into the base state of the same spin is

$$-[I(I+1) - K(K\pm1)]\epsilon .$$

In this expression, ϵ is a parameter to be fixed by experiment, and the choice of + or - depends on whether the mixing is effected through an intermediate state having $K' = K + 1$, or $K' = K - 1$. The choice of sign is unimportant, because the sign as well as the dependence on

Table VI. M1 - E2 γ-ray branching from the 721-keV 5/2 5/2- state of Np^{237}.

Final state				Relative intensity (normalized to a sum of 1)			
K	I	π	Energy (keV)	Experimental	M1	E2	E2, corrected for mixing
5/2	5/2	-	60	0.84±0.10	0.76	0.46	0.29
5/2	7/2	-	103	0.16±0.10	0.24	0.43	0.49
5/2	9/2	-	158	< 0.05	——	0.11	0.22

K drop out of the expressions for the branching ratios. Since the value of ϵ is experimentally determined, only the dependence on I is a result of the specific choice of Coriolis forces. The amplitude of the base state mixed into the beta vibrational state with the same spin is

$$[I(I+1) - K(K\pm 1)]\epsilon .$$

Corrected branching ratios are obtained by multiplying the uncorrected values by a factor chosen from Table VII. Here Q_g and $Q_{\beta-g}$ are the intraband (rotational) and interband reduced transition amplitudes.

Current studies of E2 branching from the 02+ members of the beta bands of Th^{232} and U^{238} (reference 83) indicate a value of $\epsilon(Q_g/Q_{\beta-g}) \approx +0.05$. Column 5 of Table VI gives the corrected E2 branching ratios for the 721-keV state of Np^{237}, assuming this value for $\epsilon(Q_g/Q_{\beta-g})$. There is no experimental information on which to base corrections for M1 transitions, although these are probably smaller.

The corrected E2 branching ratios are farther from the experimental values than the uncorrected ones. Unless the sign of $\epsilon(Q_g/Q_{\beta-g})$ in Np^{237} is different from the same sign in Th^{232} and U^{238}, the mixing corrections can serve only to widen the gap between observed branching ratios and the ratios predicted for E2 transitions.

Table VII. Coriolis mixing corrections to theoretical E2 branching ratios for transitions between two rotational bands with $\Delta K = 0$.

Transition	Correction factor
$I \rightarrow I - 2$	$\left[1 - (4I - 2)\epsilon \frac{Q_g}{Q_{\beta-g}}\right]^2$
$I \rightarrow I - 1$	$\left[1 - 2I\epsilon \frac{Q_g}{Q_{\beta-g}}\right]^2$
$I \rightarrow I$	1
$I \rightarrow I + 1$	$\left[1 + (2I + 2)\epsilon \frac{Q_g}{Q_{\beta-g}}\right]^2$
$I \rightarrow I + 2$	$\left[1 + (4I + 6)\epsilon \frac{Q_g}{Q_{\beta-g}}\right]^2$

The transitions are therefore assigned M1 multipolarities with reasonable certainty. The failure to observe a transition to the $I = 9/2$ level (see the last row of Table VI) is further evidence that the transitions are M1.

The M1 transitions de-exciting the "beta vibrational" state of Np^{237} contradict a specific prediction by the vibrational model. The data in fact give no indication that the expected E2 transitions occur, and these are probably lower in intensity than the M1's by a factor of at least five. On the assumption that the E2's are on the order of a few s. p. units, the M1's are at least a few hundredths of a s. p. unit. The breakdown of the vibrational model is in this case more serious than that indicated by the E1 transitions that de-excite the beta vibrational states of Th^{228} and Pu^{240}.

The E1 transitions that also de-excite the 5/2- beta vibration in Np^{237} are anotheer curious feature of the decay scheme. If one again assumes that the beta vibrational state de-excites by E2 transitions of 1 s. p. unit, and by M1 transitions that are at least five times as fact, then these E1 transitions have a reduced transition probability of at least 3×10^{-5} s. p. units.

In this case, the E1 transitions connect the "beta vibration" with a state of different intrinsic structure. This is indicated in Fig. 18 by schematic "vibrational-model wave functions." (Rotational functions and symmetrization have been omitted for simplicity.) The lifetime of the 60-keV E1 transition between the 5/2 5/2- [523] $\psi_{vib}(n_\beta = 0)$ state and the 5/2 5/2+ [642] $\psi_{vib}(n_\beta = 0)$ state has been directly measured and found to be 6.3×10^{-8} second, corresponding to a B(E1) of 8×10^{-6} s. p. units. From my work, it appears that the reduced rate of the transition 5/2 5/2- [523]$\psi_{vib}(n_\beta = 1) \overset{E1}{\rightarrow}$ 5/2 5/2+ [642] $\psi_{vib}(n_\beta = 0)$ is actually larger. Although this is unreasonable if one assumes a vibrational model, it should be borne in mind that the E1 matrix elements are still very small, so that the transition rates can be affected by very small impurities in the states.

It is interesting to note that Diamond et al. have recently observed "forbidden" M1's de-exciting gamma vibrational bands of Tb^{159} and Tm^{169}. These authors have also noted the existence of a fairly

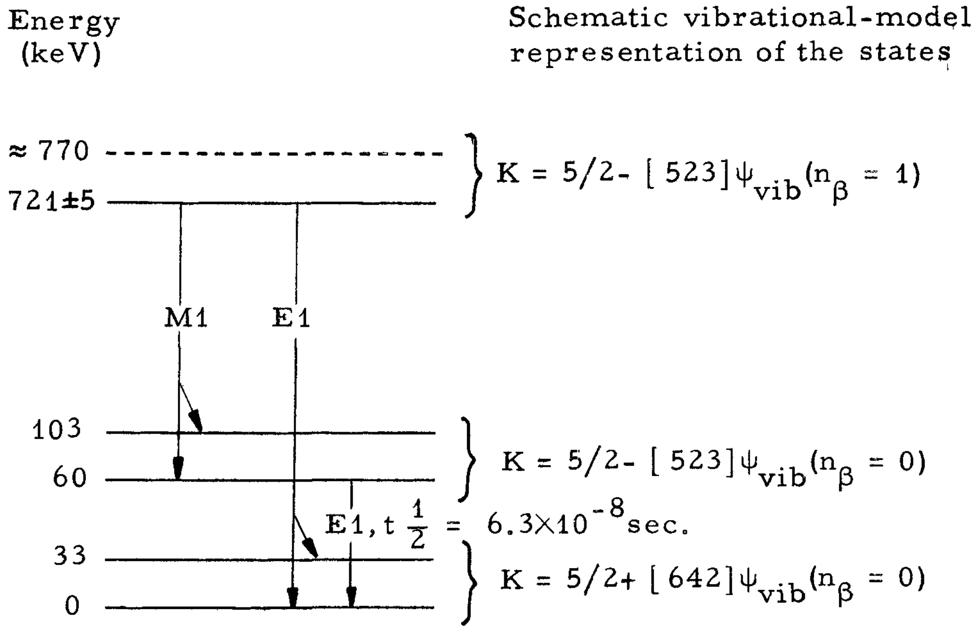

Fig. 18. Gamma-ray transitions in Np^{237}.

fast E1 transition between one of the gamma vibrational bands of Ho^{165} and another state of different intrinsic structure.[9]

Levels of Np^{239}

The beta vibrational states found in U^{235} and Np^{239} may also de-excite partly by M1 transitions to the base states, but in these cases it was not determined whether the transitions are M1 or E2. The transitions from the beta vibration to its base state in Np^{239} are much weaker than the E1 transitions to the ground state, in contrast to the corresponding beta vibrational state of Np^{237} where the transitions to the base state are twice as fast as the E1's to the ground state.

It would be interesting to know whether this is because transitions to the base state in Np^{239} are slower than those which occur in Np^{237} (possibly because the M1 component is lower in Np^{239}), or because the E1 transitions are faster. In connection with the latter possibility, it may be worth while to note that the lifetime of the E1 from the 76-keV 5/2 5/2- [523] state to the ground state of Np^{239} is $\approx 10^{-9}$ sec.[88] This represents a reduced transition probability more than 30 times that of the corresponding 60-keV E1 in Np^{237}. This could be related to (possibly) enhanced E1 transitions from the beta band of Np^{239}.

Levels of Pu^{239}

The assignment of a new Nilsson level and the failure to observe a beta band have already been discussed. This failure is disappointing, but not alarming; a beta band with about the same energy (above the band favored in alpha decay), hindrance factor, and E0/E2 ratio as the 940-keV 00+ state of Pu^{238} would fall just below the experimental intensity limits.

It should be possible to Coulomb-excite a beta vibration based on the ground state of Pu^{239}. This should be particularly interesting, in light of the very different characteristics of the 00+ states of Pu^{238} and Pu^{240}. Coulomb excitation of the odd-mass nuclei U^{235} and Np^{237} also offers interesting possibilities. In both cases, the beta vibration based on the ground state, which should be strongly populated in Coulomb excitation, is not the one observed in alpha decay. It might prove highly enlightening to compare two beta bands of the same nucleus based on different intrinsic states. Np^{237}, whose K = 5/2- [523] beta band displays irregular de-excitation, would be a good nucleus in which to make such a comparison.

Levels of Pu^{240}

The 02+ member of the beta band in Pu^{240} is populated. From the measurements by Asaro and Perlman[6] $R_{\alpha}(\beta/f) = 1.1\pm0.2$. This is more in line with the expected value ≈ 1, but in marked contrast to the low values for Th^{228}, U^{232}, and U^{234}.

In other respects, the beta band of Pu^{240} is highly irregular. It decays partly by E1 transitions to the 610-keV 01- state. The value of B(E1)/B(E2) is $(1.7\pm0.3)\times10^{-3}$, so that these E1 transitions are probably on the order of 10^{-3} s.p. units. Corresponding E1 transitions were not observed in the neighboring nucleus Pu^{238}, for which B(E1)/B(E2) is less than 7×10^{-6}, lower by a factor of more than 200.

A more striking difference between the beta bands of Pu^{238} and Pu^{240} is the relative strength of the monopole transition. This is conveniently expressed by the parameter[89] $\mu_K(0' \rightarrow 2) = W_K(E0)/W_\gamma(E2)$, where the W's are the K-electron and gamma intensities de-exciting the state. For the beta vibration of Pu^{240}, $\mu_K = 0.1\pm0.03$, compared to a value 1.0 ± 0.3 for Pu^{238}.

In spite of this strange behavior, the beta band in Pu^{240} has a very low hindrance factor. This may indicate that a low hindrance factor is characteristic of a broader class of 0+ states than "regular" beta vibrations. As mentioned in Section IV.A, it appears that the 1046-keV 0+ state of U^{234} may be populated by alpha decay with an intensity slightly below 1×10^{-8}, corresponding to a hindrance factor of ≈ 10.

Nuclei heavier than Pu^{240}

Since the beta vibrational states appear to be populated strongly in most of the nuclei studied, it is surprising that none were found in the very heavy nuclei. "Regular" beta bands, as defined near the beginning of Section V, do not exist below 940 keV in Cm^{246}, 830 keV in Cm^{248}, 1280 keV in Bk^{249}, or 1150 keV in Cf^{250}. Therefore, the energies, the hindrance factors, or both are higher in these nuclides. Neither have 1- states been observed in nuclei heavier than Pu^{240}.

Although it would be interesting to investigate the heaviest nuclides more thoroughly, it is generally difficult to produce them in sufficient quantity. Cm^{246} is available in sufficient amounts, but it must be isotope-separated from large quantities of Cm^{244}. Pure Cf^{249} is now available, or will be soon. Cf^{246} probably offers the best hope. This isotope can be prepared rather easily by an $(\alpha, 2n)$ reaction on Cm^{244}.

<u>Other lighter alpha emitters</u>

A number of lighter alpha emitters reamin to be studied by this technique. Cm^{240} can be produced in good yield by the bombardment of Pu^{239} with α particles, although the Cm^{241} also produced emits intense γ rays which may interfere seriously with coincidence studies. Pu^{234} can be produced by the reaction $U^{233}(\alpha, 3n)Pu^{234}$, but its small alpha branching is a drawback. Pu^{242} and U^{233} may be profitably studied in spite of their low specific activities. Th^{230} is a good candidate for study. Among the isotopes lighter than this, serious difficulties often develop from the growth of daughter activities into the sample. Of course, more information about the alpha emitters already studied may be obtained by this technique. Time and temporary experimental limitations were important factors in defining the scope of the experiments reported here.

VI. SUMMARY AND CONCLUSIONS

Most of the new information presented in this paper concerns the 00+ states and analogous states in three odd-mass nuclei, the "beta vibrations." No gamma vibrations have been observed in alpha decay. In one case, the alpha decay of Cm^{244}, the hindrance factor for decay to the 22+ state must be larger than 100.

Some new knowledge of octupole vibrations has been obtained. This includes possible observation of alpha decay to a 01- state of U^{234} with a rather low hindrance factor, and the observation of a state in U^{235} which is probably an octupole vibration of the same character, also with a low hindrance factor. In Rn^{218}, a probable 1- state has been observed whose E1 branching ratios give the first indication of a breakdown in the K quantum number. Most interesting is the observation of a possible K = 2- octupole band in U^{236}.

The energies and known hindrance factors for the lowest members of the octupole bands are shown in Fig. 19. The smooth trends in the 1- states are quite obvious.

The energies and hindrance factors for the beta vibrations are shown in Fig. 20. The energies of the 0+ states in Th^{232} and U^{238} are taken from the work of Stephens and Diamond.[83] All other points are taken from my work.

Both the energies and the hindrance factors show small variations which cannot be explained by a simple vibrational model. The hindrance factors appear to decrease somewhat with increasing mass number, and the energy plot seems to have a broad minimum around mass number 230. The particularly low energies of the beta vibrations in the neptunium isotopes may be caused by coupling to the odd proton. The effect of the odd neutron in U^{235} appears to be smaller.

It is surprising that the energies of the beta vibrations are not much lower in the lightest nuclides studied. These nuclei are supposedly very "soft" to β deformations, those which preserve axial symmetry. Th^{228} is supposed to be particularly deformable, but its beta vibration occurs at rather high energy. The 0+ states of Th^{226} and Ra^{222} probably

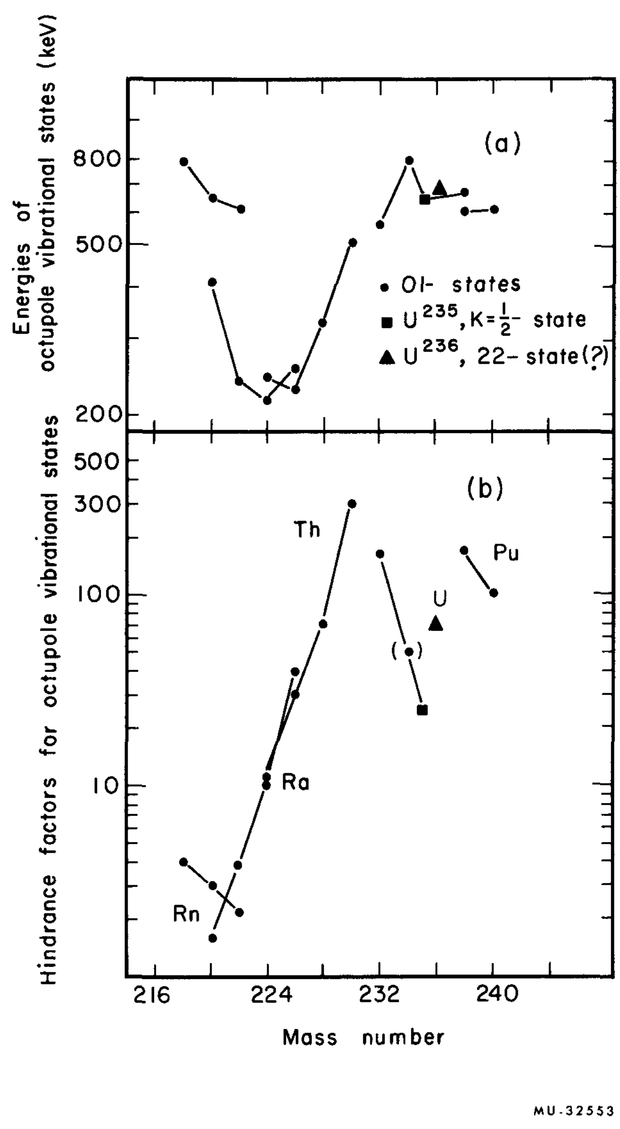

Fig. 19. (a) Energies of the "octupole" states. Many of the points are taken from other literature.

(b) Hindrance factors for alpha transitions to the "octupole" states. The hindrance factor for the state assigned K = 1/2- in U^{235} has been divided by 2.5, the hindrance factor for the favored transition. Many of the points are taken from other literature.

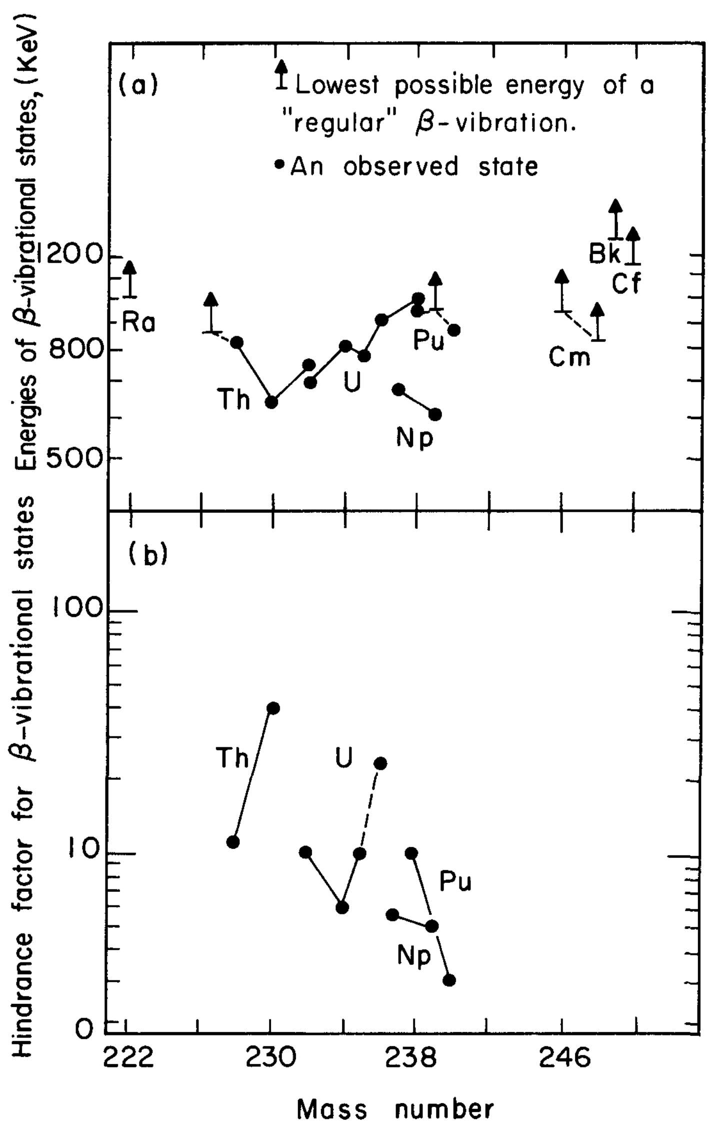

Fig. 20. (a) Energies of the "beta vibrational" states.
(b) Hindrance factors for alpha transitions to the "beta vibrational" states. Values for odd-mass nuclei are relative to the least hindered transition.

lie even higher. Although not enough is known about the 0+ states in the mass region 220 to 230, it already appears that the current ideas about deformation energies may not be entirely correct. It should also be noted that the "softness" toward odd-parity deformations, as indicated by the low energies of the 1- states in this region, has not been satisfactorily explained.

Figure 21 shows the observed ratios of K electrons to E2(+M1) γ rays de-exciting the first members of the beta bands. For even-even nuclides, conservation of angular momentum forbids M1 transitions, so this ratio is equal to $\mu_K(0' \to 2)$. For odd-mass nuclides, the quantity plotted can be lower than μ_K if M1's occur. In the case of Np^{237}, M1's have actually been shown to de-excite the beta band. Points for Th^{232} and U^{238} are again taken from the work of Stephens and Diamond.[83]

Similar values of μ_K are observed in Th^{228}, Th^{230}, U^{232}, U^{234}, and Pu^{238}. The value of μ_K is lower for the beta vibrations of Th^{232} and U^{238}, observed in Coulomb excitation, and it is especially low in Pu^{240}. It may be low for the 00+ state of U^{236}, but this point is based on questionable observations.

On the basis of a vibrational model μ_K should depend mainly on β, the equilibrium deformation of the nucleus, which varies only slightly over most of the mass region studied.[90, 89] The observed fluctuations of μ_K, particularly the sharp drop between Pu^{238} and Pu^{240}, are evidence for some noncollective character in the 0+ states. Vibrational-model calculations[89] actually show μ_K to be slightly higher in Pu^{240} than in Pu^{238}.

Recent theoretical calculations by Marshalek[91] show the energies of the beta vibrations approaching the energy gap at Pu^{240} and heavier nuclides. This may be related to the low value of μ_K in Pu^{240}, as well as the failure to observe beta vibrations in Cm^{246}, Cm^{248}, Bk^{249} and Cf^{250}. A description of the "beta vibrations" as particle states might shed some light on the behavior of these states in the heaviest nuclides, and could also explain anomalies such as occur in the de-excitation of the beta vibration in Np^{237}. Voros et al. have calculated that two quasi-particle 0+ states should lie as low as 1.2 MeV in Pu^{240} and Cm^{244}.[92]

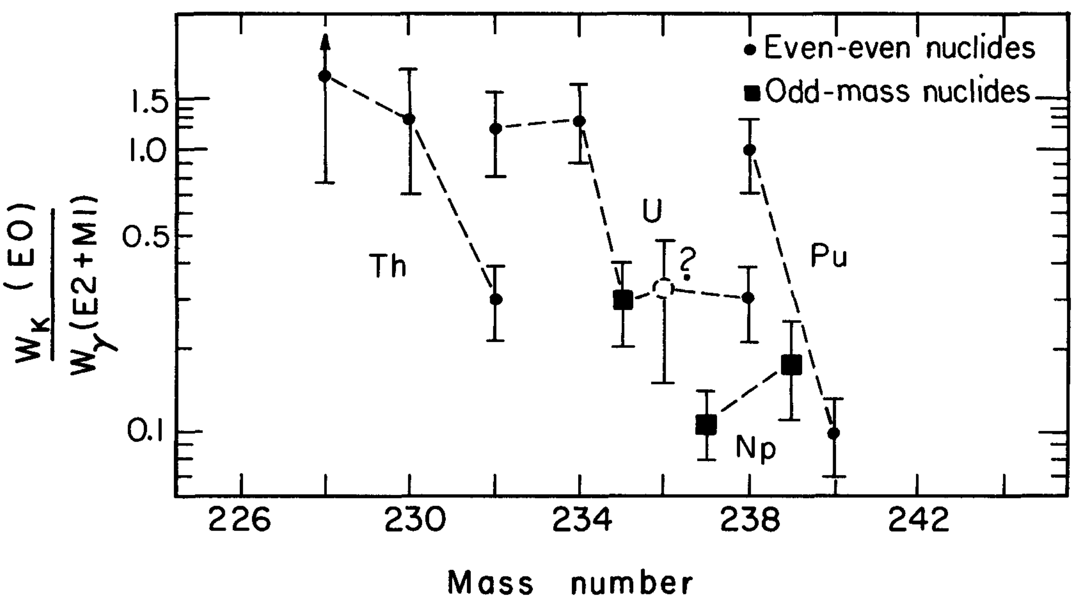

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Fig. 21. $W_K(E0)/W_\gamma(E2 + M1)$ for the "beta vibrational" states. For even-even nuclei this quantity is equal to $\mu_K(0' \rightarrow 2)$. For odd-mass nuclides, the quantity may be lower than μ_K if the "beta vibrational" state de-excites partially by M1 transitions. This is actually known to occur in Np^{237}.

The similarities between a majority of the "beta vibrations" must nevertheless be accounted for. Perhaps the states can best be described as a mixture of vibrations with one or several quasi-particle states.

ACKNOWLEDGMENTS

These experiments by their very nature involve a cooperative effort. I owe a debt to Sven Bjørnholm for the development of the entire experiment. I am grateful to Don Landis and Fred Goulding for the opportunity to share in the use of their fine electronic equipment, even while it was still under development, and for their frequent advice and assistance in the actual experiments; also, to Duane Mosier for his constant help with electronic problems. Michiyuki Nakamura, Mel Brown, George Killian, Jim Hunter, Frank Duarte, Dick Jared, and other members of several electronics groups have also helped with the design, operation, and maintenance of the equipment.

The experiments have relied heavily on the availability of semiconductor alpha detectors, which were kindly provided by William Hansen and members of his group. The lithium-drifted electron detectors were manufactured by Jack Elliott and the Accelerator Technicians group.

Frank Asaro has provided much help with chemical procedures, sometimes in the wee hours; Helen Michel has also helped me on more than one occasion. I am also grateful to V. Subramanyam for providing me with a U^{230} sample, and to Sherman Fried, and his co-workers, and members of the Health Chemistry Division for the Cf^{252} and E^{253} samples from their heavy-element program. Several valuable samples were graciously supplied to me by James Wallmann. Members of the Health Chemistry Division have provided continuous assistance with all matters of handling and safety concerning radioactive materials.

I am grateful to many members of the Chemistry Division, especially to Frank Stephens, for many fruitful discussions. Most important, I am indebted to Frank Asaro, Sven Bjørnholm, and my advisor, Professor Perlman, for the conception of the experiment, and for their patient tutelage and constant advice.

This work was done under the auspices of the U. S. Atomic Energy Commission and with the assistance of a graduate fellowship from the National Science Foundation.

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