Alpha Decay to Vibrational States

S. BJØRNHOLM,* M. LEDERER, F. ASARO, AND I. PERLMAN
Lawrence Radiation Laboratory, University of California, Berkeley, California
(Received 18 October 1962)

A coincidence apparatus has been developed which permits the measurement of energies and intensities of rare alpha groups leading to excited states of the product nucleus. It has proved possible to measure transitions with intensities as low as 10^{-8}, and the technique has been used in the present study, principally to characterize vibrational states. The energies, relative abundances, and multipole orders of the gamma-ray and conversion electron transitions which de-excite the levels populated by these weak alpha branchings can be determined, and these lead to assignments of spin and parity for the levels. Excited states with spin and parity 0+ (beta vibrations) were confirmed in the energy level spectrum of Th^{230}, U^{234}, Pu^{238}, and Pu^{240}. A corresponding state was observed at 780 keV in the energy level spectrum of U^{235} and was given the assignment 1/2+. The reduced alpha transition probabilities to states of this type were found generally to be about an order of magnitude smaller than the theoretical value for unhindered alpha decay. The characterization of these levels simply as *beta vibrations* appears to be inadequate in that there are distinct differences in the modes of de-excitation for the different nuclides.

I. INTRODUCTION

THE experiments reported here represent an attempt to develop a general method for studying collective states of vibrational character in heavy nuclei. The energy levels of nuclei from Th^{230} to Cf^{250} have been examined.

All of the nuclei in this region are known to have nonspherical equilibrium shapes. Decay-scheme[1–6] and Coulomb-excitation[7–9] studies have disclosed a number of excited states which can be described in terms of quadrupole and octupole vibrations of an axially symmetric ellipsoid. Some of the reasons for this type of description are: (1) These states appear in even-even nuclei below the energy gap defined by the odd-even mass difference; (2) the $E2$ and $E3$ transition probabilities found in Coulomb excitation are enhanced relative to the single-particle estimates; (3) similar levels seem to appear systematically and show a more or less smooth variation of excitation energy with change in mass number. Vibrations of one class, the beta vibrations, are distinguished in their decay by strong electric monopole transitions. Even though it became evident during the course of the present work that these states could *not* be described simply in terms of a pure beta vibration, for ease of description they will be called "beta vibrational states" in the remainder of this paper.

Although something is known about the location of vibrational states and of their characteristics, it seems certain that a much more detailed examination would be fruitful. As an example, these states appear not far below the energy gap, and an interplay between single-particle and collective aspects may be expected. The particle aspects will undoubtedly exhibit considerable variation from nucleus to nucleus and this suggests that a wide range of species be examined.

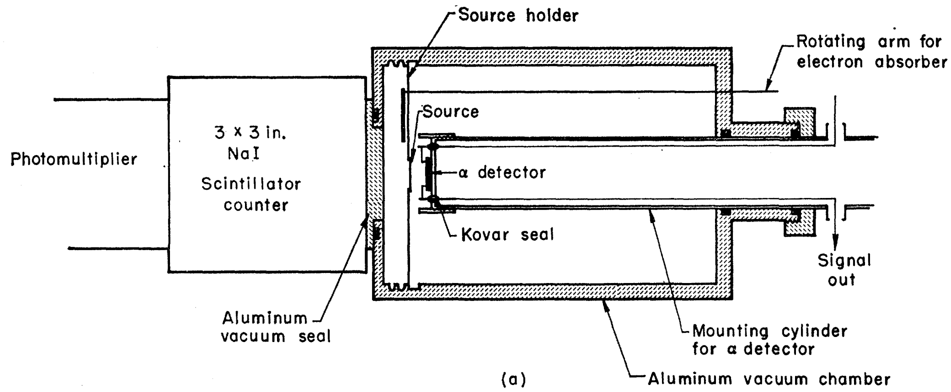
The principal obstacle in studying this class of energy levels is that there exists no widely applicable means of exciting nuclei to these states with the exception of Coulomb excitation. Many nuclei of interest in the heavy-element region do not lend themselves to this method because of their short lifetimes. Such possible methods as the investigation of neutron-capture gamma rays involve the same problem.

The alpha-decay process has much to recommend it for this problem including the large number of alpha emitters with suitable half-lives and the discreteness of alpha-particle energies. Indeed much has already been learned by this route. The limitation here is that states as high as 1 MeV are very poorly populated in alpha decay and, therefore, one must devise methods for measuring alpha groups of very low intensity. In the work reported here, silicon semiconductor detectors used as alpha-particle spectrometers were applied to this problem.

The demands upon the equipment may be gauged by calculating from alpha-decay theory the *maximum* expected intensity of an alpha group leading to a state at some selected energy above the ground state. (The *maximum* intensity has reference to an alpha emission process which is "unhindered," i.e., the reduced alpha transition probability to the specific state is the same

* On leave from the Institute for Theoretical Physics, University of Copenhagen, Copenhagen, Denmark, during the period of these studies.

[1] D. Strominger, J. M. Hollander, and G. T. Seaborg, Rev. Mod. Phys. **30**, 585 (1958).
[2] C. J. Gallagher, Jr., and T. D. Thomas, Nucl. Phys. **14**, 1 (1959).
[3] G. T. Wood, Phys. Rev. **119**, 2004 (1960).
[4] R. G. Albridge and J. M. Hollander, Nucl. Phys. **21**, 438 (1960).
[5] F. Asaro and I. Perlman, Lawrence Radiation Laboratory Chemistry Division Annual Report, 1960 UCRL-9566, 1961 (unpublished); p. 50. I. Perlman, Frank Asaro, B. G. Harvey, and F. S. Stephens, Jr., Bull. Am. Phys. Soc. **2**, 394 (1957).
[6] M. E. Bunker, B. J. Dropesky, J. D. Knight, J. W. Starner, and B. Warren, Phys. Rev. **116**, 143 (1959).
[7] P. H. Stelson and F. K. McGowan, Phys. Rev. **121**, 209 (1961).
[8] F. E. Durham, D. H. Rester, and C. M. Class, *Proceedings of the International Conference on Nuclear Structure at Kingston, Canada, 1960* (University of Toronto Press, Toronto, 1960), p. 594.
[9] R. Diamond, B. Elbek, and F. S. Stephens, Jr. (private communication).



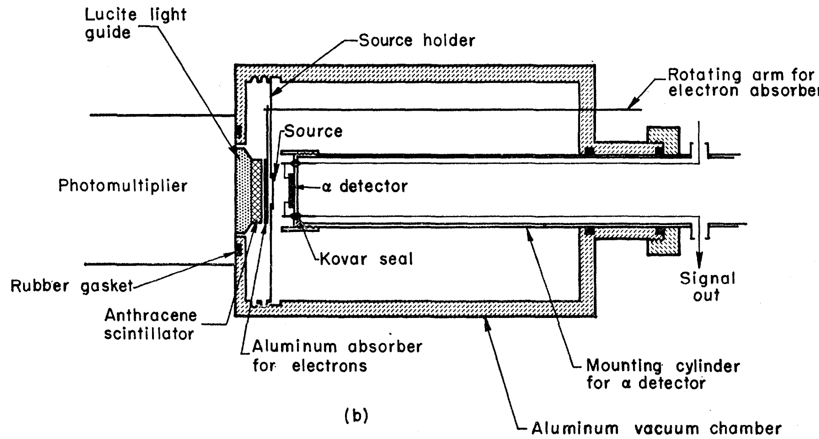


FIG. 1. Source detector arrangement; (a) for alpha-gamma coincidence measurements and (b) for alpha-electron coincidences. The position of the semiconductor alpha detector can be varied relative to the source, whereas the source geometry for the scintillation detectors is fixed. Absorbers can be inserted in front of the scintillation detectors. The electron absorbers is shown in position in 1 (b), although in a normal run it is moved away.

as to the ground state of even-even nuclides.) Table I illustrates these guide lines in a somewhat different form. Here, we have selected alpha-group intensities of 10^{-4} and 10^{-8} relative to the ground-state transition and have listed the corresponding energies of excitation which an unhindered transition would reach. For example, in the decay of Pu^{238}, the alpha intensity of a group leading to a hypothetical state in U^{234} at 620 keV should not be greater than 10^{-4}. If we are looking for a state at about 1170 keV, we should not expect an intensity greater than 10^{-8}.

To observe a sufficient number of events requires alpha-count rates up to 10^5 counts/sec for intervals of the order of several days. The importance of semiconductor counters, compared to gas ionization chambers, lies in the very rapid collection of the charge generated by the alpha particle, which makes it possible to record a higher number of events per unit time without serious loss in energy resolution from pile-up effects. Still, a direct recording of alpha groups of intensities 10^{-4}–10^{-8} relative to the ground-state alpha group is not possible since the weak groups will be masked by the low-energy tail from the main alpha peak. The transition to the excited state, however, occurs in coincidence with high-energy gamma rays or conversion electrons and is hereby distinguishable in alpha-gamma and alpha-electron coincidence measurements. The high solid angle obtainable with semiconductor counters as compared to magnetic spectrometers is here essential.

Coincidence experiments of the type just mentioned will also yield information on the modes of de-excitation of the excited state. Such information will include gamma-ray branching and conversion coefficients, data which will be of importance for the characterization and classification of the state.

Some progress had been made on the identification of vibrational states populated by alpha decay by employing other types of detectors. Perlman *et al.*[5] were able to measure directly the intensities of high-energy gamma rays from a number of alpha emitters. These were ones which could be obtained in states of high isotopic purity and free of other radioactive contaminants. Similarly, several monopole transitions were identified by measuring coincidences between the electron lines (anthracene crystal detector) and K x rays. All of these measurements could be made with intense and thick sources because the alpha particles themselves were not analyzed. In one favorable case, the decay of Cm^{244}, two of the alpha groups leading to the beta vibrational band around 870 keV were seen by magnetic alpha spectroscopy[10] even though the total intensity of the groups was only 2.0×10^{-6}.

[10] F. Asaro and I. Perlman (unpublished data).

TABLE I. Predicted maximum excited state energies for alpha group intensities of 10^{-4} and 10^{-8}.

Parent nuclide	Half-life	Excitation energy (keV) corresponding to an alpha intensity of: 10^{-4}	10^{-8}
Ra^{222}	38 sec	900	1580
Ra^{224}	3.6 day	730	1330
Ra^{226}	1.6×10^{3} yr	560	1040
Th^{226}	31 min	810	1500
Th^{228}	1.9 yr	640	1200
Th^{230}	8.0×10^{4} yr	520	970
U^{230}	21 day	700	1320
U^{232}	74 yr	610	1140
U^{234}	2.5×10^{5} yr	520	980
Pu^{236}	2.8 yr	660	1250
Pu^{238}	90 yr	620	1170
Pu^{240}	6.6×10^{3} yr	580	1060
Cm^{240}	27 day	730	1380
Cm^{242}	163 day	720	1330
Cm^{244}	18 yr	670	1240
Cf^{246}	36 h	810	1520
Cf^{250}	11 yr	700	1290
Cf^{252}	2.2 yr	710	1320
Fm^{254}	3.3 h	880	1650

II. EXPERIMENTAL

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

Figures 1 (a) and 1 (b) show the source-detector arrangement. The sources consisted of thin deposits of the active materials on polystyrene and were prepared as described in Sec. II C. The alpha-particle detector was a phosphorous-diffused, *p-n* junction, silicon semiconductor of the guard ring type.[11] Different detectors with sensitive areas from 0.5 to 2.0 cm in diameter could be inserted; most often one of 1 cm in diameter was used. A solid angle acceptance close to 50% was attainable with small sources, *s*, and the solid angle could be varied by moving the brass cylinder on which the detector was mounted. Normally, no collimation of the alpha beam was necessary. Some of the measurements were made with surface barrier counters[12] and for these, collimation was required to avoid edge effects. The two types of counters were otherwise similar in performance. The bias voltage varied between 100 and 300 V for the diffused junction counters. It was about 25 V for the surface barrier type.

Figure 1(a) shows the mounting of the 3-in.×3-in. NaI crystal. The source geometry relative to the crystal was fixed. Careful calibration (±5%) of the over-all photo- and Compton-peak efficiencies was made with the aid of gamma-ray standards of Na^{22}, 511 and 1276 keV; Cs^{137}, 662 keV; and Co^{60}, 1173 and 1333 keV. Other standards were used for energy calibration. A set of calibrated absorbers inserted in the slots between source and detector were part of the equipment.

The electron detector, shown in Fig. 1(b), was an anthracene crystal 2.5 cm in diameter and 0.3 cm thick. The photomultiplier tube was sealed vacuum tight against a rubber gasket, and a short light guide provided the optical contact. The counter was calibrated with a Cs^{137} source (624-keV electron energy) mounted on polystyrene; the effective solid angle was found to be 33%. In order to measure the response of the anthracene crystal to gamma rays separately, a 0.1-cm aluminum absorber was moved in front of the counter [see Fig. 1(b)]. The source backing was chosen of sufficient thickness so that no alpha particles would reach the electron counter, and polystyrene was selected because it gives a minimum loss of electrons from backscattering.

The electronic equipment is shown in the block diagram, Fig. 2. The amplifiers for the scintillation detectors are of conventional design, as are the single-channel and multichannel analyzers. The preamplifier for the semiconductor detector was designed by Goulding.[11] The decay constant of the output pulse has been lowered from 20 to 0.5 μsec to accommodate higher counting rates. A double delay line amplifier was chosen in order to obtain maximum counting rate although the resolution became decreased by a factor of about 1.5 relative to *RC* pulse shaping.[13,14] The 5-Mc scaling unit with variable scaling factor served to decrease the counting rate before the pulse train reached the biased amplifier and the multichannel analyzer. Output pulses

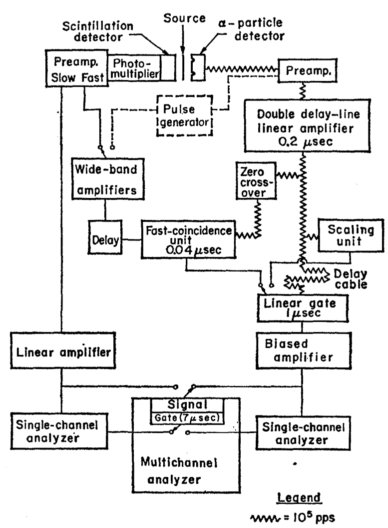


FIG. 2. Simplified block diagram of the electronic apparatus used in the experiments with silicon semiconductor alpha detectors.

[11] Fred S. Goulding and William L. Hansen, Nucl. Instr. Methods 12, 249 (1961).

[12] The surface barrier counters were kindly provided by Dr. R. M. Latimer of the Lawrence Radiation Laboratory.

[13] E. Fairstein, Nuclear Science Services Report No. 32, National Academy of Science, National Research Council Publication 871 (unpublished).

[14] D. A. Landis, M. S. thesis, University of California Lawrence Radiation Laboratory Report UCRL-10001, 1961 (unpublished).

from any of the decade units could trigger the linear gate, and with this arrangement it was possible to measure coincident alpha spectra and singles alpha spectra without change of counting rate and amplifier settings. A zero crossover pick-off unit[15] was used to extract a time signal for the fast coincidence circuit. A pulse generator with variable amplitude fed into the first stage of the preamplifier. A synchronous pulse could be used to trigger the fast coincidence unit. This arrangement proved useful during the development of the system. The pulse generator was stable and linear, and the arrangement was used routinely for energy calibration and stability tests.

The entire alpha counting equipment had to meet very strict requirements with respect to resolution (<0.8%) and stability (<0.2% drift for periods of days).

B. Gamma-Ray Spectra Equipment

Total gamma spectra were measured with a 3-in. ×3-in. NaI crystal coupled to a 100-channel analyzer. Gamma-gamma coincidence measurements were made with two 3-in.×3-in. crystals coupled to a standard fast-slow coincidence arrangement. The resolving time of the fast circuit was 0.06 μsec.

Coincidences between electrons and K x rays were measured with a slow coincidence unit (2–4 μsec). Detectors were a 0.6-in.-diam×0.1-in.-thick anthracene crystal and a 1.5-in.×1-in. NaI crystal covered by a beryllium window.

C. Source Preparation

Three types of sources were employed. A sample of U^{234} was available which had been prepared by vacuum sublimation onto a nickel foil (22 mg/cm² thickness) and covered an area 2.5 cm in diameter. Other substances with relatively low specific activity (Pu^{238}, Pu^{239},

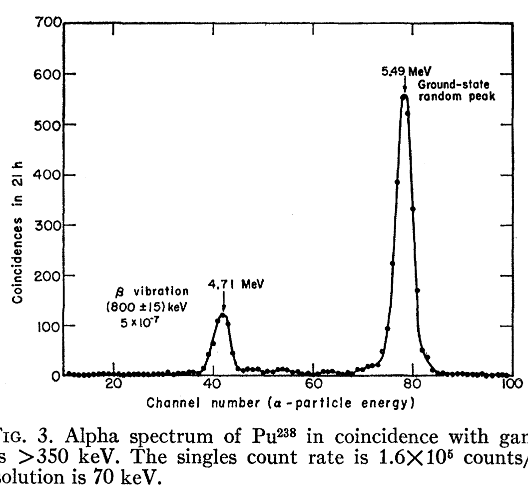


FIG. 3. Alpha spectrum of Pu^{238} in coincidence with gamma rays >350 keV. The singles count rate is 1.6×10⁶ counts/sec. Resolution is 70 keV.

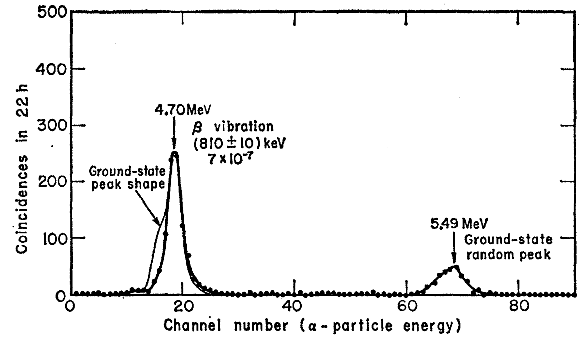


FIG. 4. Alpha spectrum of Pu^{238} in coincidence with conversion electrons >450 keV. Singles count rate 0.93×10⁵ counts/sec. Resolution 45 keV. The shape of the alpha groups to the ground-state band as recorded in the singles spectrum has been drawn at the position of the 4.70-MeV peak for comparison (thin line).

and Cm^{244}) were likewise mounted by vacuum sublimation of the oxides or chlorides, but the backing material for these was polystyrene of 5 or 10 mg/cm² thickness. The activities covered areas of 0.8-cm diam. The shorter lived activities (Cm^{242}, Cf^{252}, E^{253}, and Fm^{254}) were deposited from solution into a thin ion-exchanger film (10–15 μg/cm²) prepared on the surface of the polystyrene by controlled sulfonation with fuming sulfuric acid.[16] The diameters of these sources ranged from 0.3 to 0.8 cm.

III. PROCEDURE

Typically, the gamma-ray or electron singles spectrum was measured first, employing standards for energy calibration as already mentioned. Next, measurements were made of the alpha-particle singles spectrum to establish the source strength and to obtain an energy calibration. The energy calibration was aided by the precisely linear pulse generator.

In order to avoid excessive counting rates in the biased amplifier and pulse-height analyzer in the singles measurement, only those pulses gated by the output of the scaling unit were allowed to pass through the linear gate and into the remainder of the equipment. The scaling factor was adjustable in powers of 10 and ranged from 1 to 10^4.

After the singles measurements, the delay on the scintillation counter input to the fast coincidence unit was adjusted using either the source under investigation or a Cm^{243} standard source.

As can be seen from Fig. 2, in the coincidence measurements the 1-μsec linear gate allowed alpha pulses to pass on to the biased amplifier if a fast coincidence had occurred. Using either of the single-channel analyzers we had the choice of recording the alpha spectrum in coincidence with a particular gamma energy band or the gamma-ray spectrum in coincidence with a selected alpha energy. The coincidence measurement normally

[15] F. S. Goulding and R. A. McNaught, Nucl. Instr. Methods **9**, 282 (1960).

[16] S. Bjørnholm and M. Lederer, Nucl. Instr. Methods **15**, 2331 (1962).

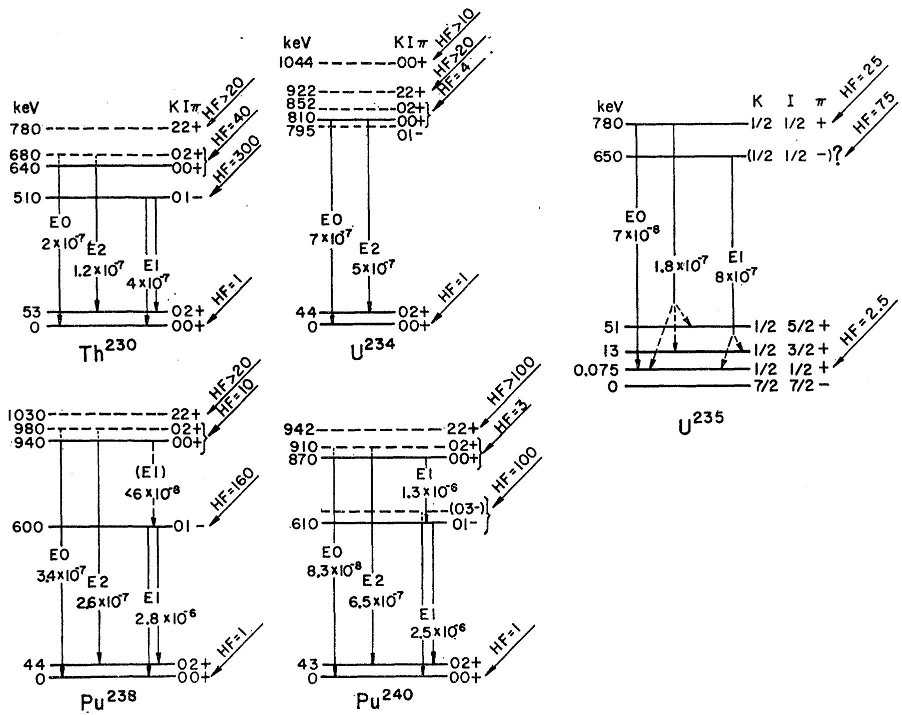


FIG. 5. Summary of decay schemes studies in the present investigation.

ran for one or more days. After completion of the coincidence measurement, the singles spectra, the energy calibrations, and the measurement of the delay curve were repeated to check the stability. The coincidence runs were then resumed if the stability proved satisfactory and better counting statistics were required.

The intensity of a peak in the coincidence spectrum was calculated by comparing it to the accumulated number of alpha pulses in the ground-state peak determined from the singles spectrum and the length of the counting interval. The intensity had to be corrected for absorption, solid angle, and detection efficiency of the scintillator. A small correction for losses from pile-up effects was also required, and finally a correction was made for the efficiency of the fast coincidence circuit. For this purpose, we used Cm^{243} which has abundant alpha groups of well-known intensity[1] in coincidence with 210-, 228-, and 278-keV $M1$ transitions. We also used the more indirectly determined intensities of gamma rays and conversion electrons de-exciting the 810 keV (00+) state[17] in U^{234} seen in the decay of Pu^{238}.[5] In both of these cases we found that coincidence efficiency was $(75\pm5)\%$ whether gating with electrons or gamma rays. This efficiency was unexpectedly low and was doubly puzzling in that it did not depend on the scintillator used. However, the result was found to be reproducible.

[17] The designation of vibrational states will consist of the K quantum numbers followed by the spin and parity. The beta vibrational state 00+ and the first rotational member 02+ are distinguished from the ground-state band with the same symbols by the context in which they appear.

IV. RESULTS

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

Vibrational states in U^{234} obtained by studying the beta decay of Pa^{234} and electron capture decay of Np^{234} have been reported[2,3] as follows: 810 keV (00+), 850 keV (02+), 795 keV (01−), 922 keV (22+), and 1044 keV (00+).

The spectrum shown in Fig. 3 displays the alpha groups which are in coincidence with gamma rays of energy >350 keV. By setting the gamma-ray gate at this energy, the relatively intense alpha transitions to the ground-state rotational band were excluded. However, in all spectra so taken the intense ground-state transition always appeared because of chance coincidences. It is seen that only a single alpha group appeared in true coincidence, this to a state in U^{234}, (800 ± 15) keV above the ground state. This level is identified with 00+ state at 810 keV mentioned above. There is no evidence for a transition to the 02+ member of this band although the resolution is not sufficiently good to set a very low limit.

Somewhat better resolution was obtained when the alpha spectrum was measured in coincidence with electrons (see Fig. 4). Here again, the only alpha group seen was that to a state of ~800 keV. The peak shape superimposed in Fig. 4 is that which would have been obtained if the population to the 02+ vibrational state was relatively the same as to the analogous 02+ state of the ground-state band, and if the $E0$ transition probability from the 02+ state were the same as from the

00+ state. It may be worth noting that with this particular nuclide, Durham *et al.*[8] failed to see any $E0$ transition in their otherwise successful attempts to observe 02+ states by Coulomb excitation (Th^{230}, Th^{232}, U^{236}, and U^{238}).

In the present work, the total population of the two levels in the beta band is 1.2×10^{-6}, in good agreement (as mentioned earlier) with the intensity determined by gamma-ray singles and K x-ray-electron coincidence measurements.[5] The alpha intensity corresponds to a hindrance factor (HF) of 4. No attempt is made at present to obtain a lower limit for hindrance factor for the transition to the 02+ state.

For the two higher lying levels found in Pa^{234} decay, upper limits can be established on the alpha population and from these, minimum values for hindrance factors. We find for the alpha decay to the 22+ state at 922 keV, HF > 20; to the 1044-keV level, HF > 10. This information is summarized in Fig. 5 along with that from other studies.

In searching for extremely rare events accompanying the alpha-decay process, spurious gamma rays arising from nuclear reactions of the alpha particles may be encountered. An intense sample of Pu^{238}, which showed clearly the 765-keV gamma ray (from the 810-keV state to the 44-keV state), also had a gamma ray of ~880 keV which grew in intensity as the sample aged. This was finally deduced to be the 875-keV transition in O^{17} excited by the reaction, $N^{14}(\alpha,p)O^{17}$; the Pu^{238}, which was initially wet, gradually dried permitting air to come in contact with the alpha particles. The same gamma ray had been noted in a number of other cases in which the source mounting permitted alpha particles to come into contact with air. Figure 6 shows the gamma spectrum of Pu^{238}: In the top section an air gap of 1/8 in. was permitted between the source and a protecting foil; in the lower section a 1-mil nickel foil was placed in loose contact with the source. It is seen that the peak at 880 keV is strongly diminished.

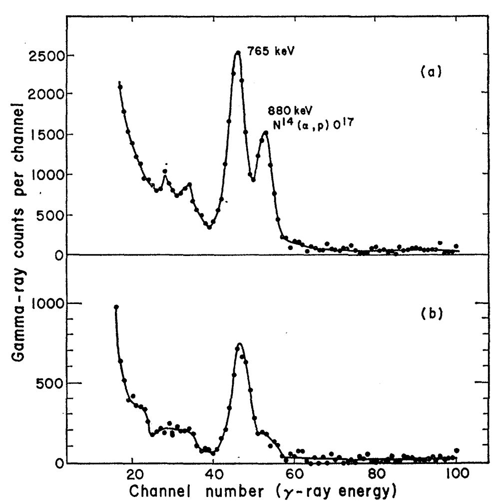


FIG. 6. Pu^{238} high-energy gamma-ray spectra (a) with 1/8-in. air gap between sample and cover, (b) with 0.001-in. Ni foil on top of sample (loose contact).

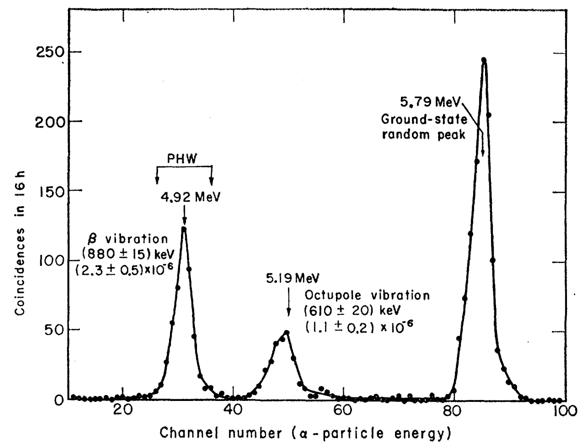


FIG. 7. Alpha spectrum of Cm^{244} in coincidence with gamma rays >480 keV. PHW refers to the pulse-height window used when recording the gamma rays in coincidence with the 4.92-MeV alpha groups (see Fig. 8).

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

The alpha spectrum in coincidence with gamma rays of energy >480 keV, Fig. 7, shows two peaks with energies corresponding to the 01− state at 610 keV and the 00+ state at 870 keV.[1,6] It appears that the alpha group to the 610-keV "state" is irregular and broad indicating a considerable population of the higher lying 03− rotational state.

Figure 8 shows the gamma-ray spectrum which is in coincidence with the alpha group (4.92 MeV) which populates the beta vibrational state at 870 keV. It reveals unexpected complexity. There is seen not only an 825-keV ($E2$) transition, but also the gamma rays de-exciting the 01− state plus a 262-keV transition. This can only mean that the 00+ level de-excites via the 01− state. A gamma-gamma coincidence experiment confirmed qualitatively and quantitatively the existence of the cascade. A portion of these cascading gamma rays will be recorded simultaneously by the NaI crystal, because of the close geometry, and contribute as a sum peak to the 825-keV events. Correcting for this we find the intensities shown in Table II.

It is possible to determine the total conversion coefficient of the 262-keV transition by a comparison of intensities in the cascade. They should be equal if no

TABLE II. Cm^{244} γ rays in coincidence with α_{870}.

Energy (keV)	Type	Intensity per Cm^{244} α decay
262	$E1$	$(1.2\pm0.3)\times10^{-6}$
570, 610	$E1$	$(1.4\pm0.3)\times10^{-6}$
825	($E2$)	$(6.5\pm2)\times10^{-7}$

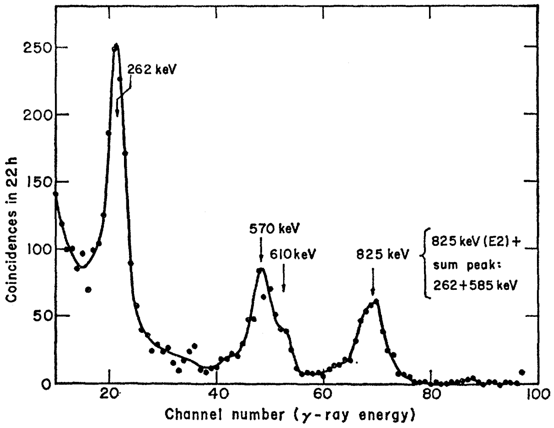


FIG. 8. Gamma spectrum of Cm^{244} in coincidence with alpha particles of the 4.92-MeV peak (see Fig. 7).

internal conversion takes place. Using the theoretical conversion coefficient of the 570–610 keV $E1$ groups, we find for the 262-keV transition a total conversion coefficient of 0.15±0.08. The theoretical coefficient is 0.06 for an $E1$ transition, 0.33 for $E2$, and much higher values for other multipolarities.

The coincidence spectrum between alpha particles and electrons of energy greater than 450 keV is shown in Fig. 9. The only alpha group with intensity greater than 1×10^{-8} was that to the 870-keV state. In particular, the upper limit can be applied to the states at ~600 keV and confirms the $E1$ assignment for the transitions to the ground-state band.

The intensity of all electron coincidences from the 870-keV level is $(9.5\pm2)\times10^{-8}$. If we assume the 825-keV transition to be $E2$, its contribution to the conversion electrons is 1.2×10^{-8} and we are left with $(8.3\pm2)\times10^{-8}$ for the 870-keV monopole transition. This interpretation is consistent with the assumption that the 870-keV level is a 00+ state. If this were not so, the 825-keV gamma ray could be an $M1$ transition in which case all of the intensities would be different and the 870-keV transition could not be $E0$. However, the 870-keV state is most likely the same as the 867-keV state found in the beta decay of Np^{240} which was shown definitely to decay by an $E0$ transition.[6]

The total population of the 01− band is $(1.1\pm0.2)\times10^{-6}$, HF=100±20; and of the 00+ band $(2.3\pm0.4)\times10^{-6}$, HF=3±0.5. As mentioned before, a direct measurement with a magnetic alpha spectrograph showed[10] two alpha groups which were identified as populating the beta band. The intensities were $(1.55\pm0.16)\times10^{-6}$ for the 00+ state at 863 keV and $(0.50\pm0.08)\times10^{-6}$ for the 02+ state at 903 keV. The present determination compares reasonably well with the sum, $(2.05\pm0.18)\times10^{-6}$.

A 22+ state has been reported at 942 keV.[6] The absence of a corresponding alpha group in the present study permits us to set a lower limit on the hindrance factor for this group: HF>100. All of the information discussed here is summarized in Fig. 5.

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

In previous measurements of the Cm^{242} high-energy singles gamma-ray spectrum,[5,18] gamma rays of 562, 605, and 890 keV were observed. Electron spectra measurements[5] indicated a (941.4±2)-keV transition with an intensity of 5×10^{-7}. The 605- and 562-keV transitions were characterized as $E1$ transitions originating from a 01− state at 605 keV and decaying to the ground and first excited states of Pu^{238}. The 890-keV photon and the 941-keV electron transitions were characterized as $E2$ and $E0$ transitions, respectively, decaying from a 00+ state to the first excited state and ground state of Pu^{238}. Albridge and Hollander[4] measured the conversion electron spectra in the beta decay of Np^{238} to Pu^{238} and tentatively assigned the 00+ state at (943.1±1) keV.

Our measurements with Cm^{242} were complicated by the presence of a few percent by activity of Cm^{243} in the source. Thus, it was not possible from our measurements alone to establish which groups belong to Cm^{242} and which to Cm^{243}.

The alpha spectrum in coincidence with gamma rays showed groups of 5.51, 5.27, and 5.18 MeV (see Fig. 10). The alpha spectrum in coincidence with electrons showed a predominant peak at 5.17 MeV with very little evidence for the 5.27- or 5.51-MeV groups (see Fig. 11). The gamma-ray spectrum in coincidence with the 5.18-MeV group showed gamma rays of 755 and 900 keV and weaker ones of 470 and 580 keV. In

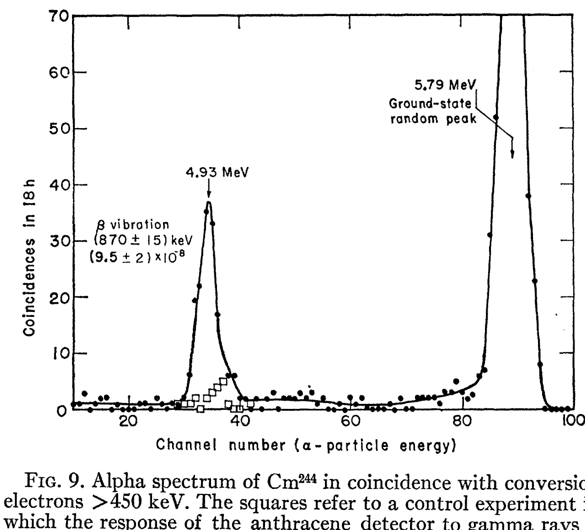


FIG. 9. Alpha spectrum of Cm^{244} in coincidence with conversion electrons >450 keV. The squares refer to a control experiment in which the response of the anthracene detector to gamma rays is recorded separately.

[18] F. Asaro, B. G. Harvey, and I. Perlman (unpublished) as quoted by F. S. Stephens, Jr., F. Asaro, and I. Perlman, Phys. Rev. **100**, 1543 (1955).

coincidence with the 5.27-MeV group was predominantly the gamma ray at 755 keV (see Fig. 12).

The 5.51-MeV group agrees well with the expected energy for populating the previously assigned 01− state at 605 keV. Its intensity, $(2.8\pm0.5)\times10^{-6}$, is likewise in good agreement with the sum of the intensities of the 562- and 605-keV radiations previously observed,[5] 3.2×10^{-6}. Comparison of the intensity of the 5.51-MeV group in coincidence with gamma rays and electrons establishes a maximum conversion coefficient for the ~600-keV gamma rays of 1%. This value is consistent only with an $E1$ assignment for these transitions and confirms the spin and parity assignment of 1− for the 605-keV level. The hindrance factor for the alpha population to this level is 160±30 from our measurement.

The 5.17-MeV alpha group which we observed in coincidence with electrons agrees well with the expected energy for populating the 00+ state at ~940 keV. The observed intensity, $(3.4\pm0.8)\times10^{-7}$, is in reasonable agreement with the previous measurement of the conversion electron intensity, 5×10^{-7}. The intensity of the 900-keV gamma ray in coincidence with the 5.18-MeV alpha groups, $(2.6\pm0.8)\times10^{-7}$, is appreciably smaller than the intensity of the 890-keV gamma ray observed in the previous gamma-ray singles measurement, 8×10^{-7}. For the reasons given in the earlier section on Pu^{238} results, the singles measurement probably contained a considerable portion of an 875-keV gamma ray resulting from a $N^{14}(\alpha,p)O^{17}$ reaction. The intensity of the conversion electron de-exciting the 940-keV state relative to the gamma rays is only consistent with the presence of an $E0$ transition. Thus, our results confirm the 00+ assignment for the ~940-keV state in Pu^{238}. The total population to the beta band is $(6\pm1.5)\times10^{-7}$ giving a HF of 10±3. No population of the gamma vibrational state (22+) at 1030 keV[4] is observed and a lower limit of 20 can be set for the hindrance factor.

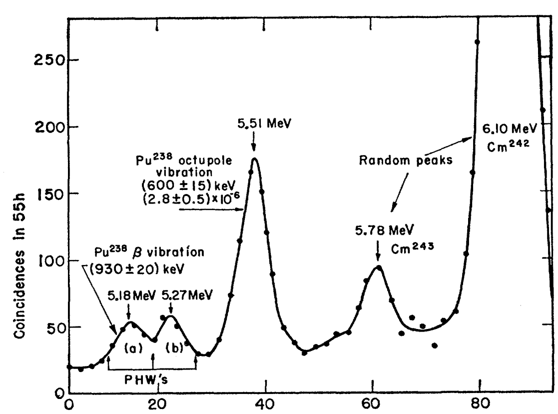


FIG. 10. Alpha spectrum of Cm^{242} containing a few percent Cm^{243} in coincidence with gamma rays >400 keV. In two subsequent experiments the gamma rays in coincidence with the alpha peaks at 5.18 and 5.27 MeV have been recorded (see Fig. 12).

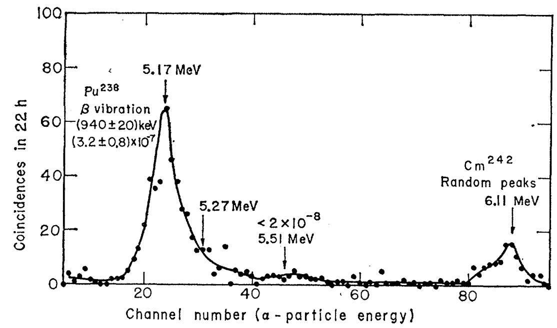


FIG. 11. Alpha spectrum of Cm^{242}, containing a few percent Cm^{243}, in coincidence with electrons >350 keV.

The 580-keV gamma ray observed in coincidence with the 5.18-MeV alpha group could represent a small amount of decay of the 00+ state through the 01− state but it must be caused at least in part by a small overlap of the relatively intense 5.51-MeV alpha group in the 5.18-MeV gate. Therefore, the intensity of the 580-keV gamma ray, 6×10^{-8}, represents only an upper limit to the branching from the 00+ through the 01− state (≤10%). This relatively small value is in contrast to the corresponding situation in Pu^{240} where this branching is over 60%.

The 755-keV gamma ray was observed to be in coincidence with both the 5.18- and 5.27-MeV alpha groups. No gamma ray of this energy has been observed in either the singles gamma ray spectrum of Cm^{242} alpha decay or the beta decay of Np^{238}.[4] If the 5.27-MeV alpha group belonged to Cm^{243} (intensity $\sim2\times10^{-5}$ per Cm^{243} alpha decay), it would populate a state ~800 keV above the ground state of Pu^{239}. This state could then de-excite to the known state of Pu^{239} at 57 keV by an ~750-keV transition, in good agreement with the observed gamma-ray energy of 755 keV. The 755-keV gamma ray in coincidence with the 5.18-MeV alpha group, as well as the 470-keV transition may also belong to the decay of Cm^{243}.

More recent measurements[19] on a pure Cm^{242} sample prove that the 5.27-MeV alpha group and the 470- and 755-keV gamma rays do not belong to the decay of Cm^{242}. The limit on the El cascade transitions has been reduced to 1% of the total population of the 00+ state.

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

As mentioned in the introduction, the characteristic association of strong electric monopole transitions with beta vibrational states might make it possible to identify one in an odd-A nucleus where the high density of particle states makes the identification of vibrational states difficult. Figure 13 shows the result of an alpha-electron coincidence measurement with Pu^{239}. Forty-six events were observed in as many hours, but they still

[19] M. Lederer, F. Asaro, and I. Perlman (unpublished).

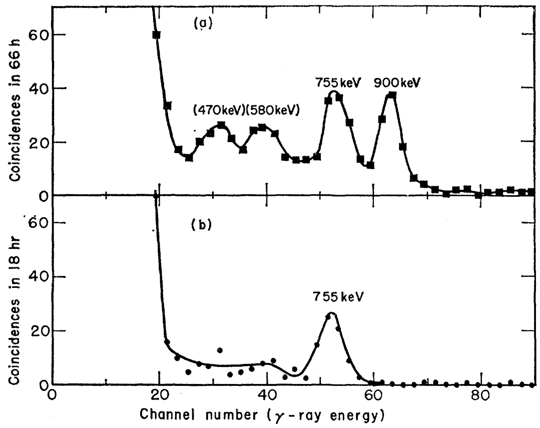


FIG. 12. Gamma-ray spectrum of Cm^{242}, containing a few percent Cm^{243}, in coincidence with alpha particles (see Fig. 10): (a) in coincidence with 5.18-MeV alpha particles, (b) in coincidence with 5.27-MeV alpha particles.

establish very clearly an alpha group to a state or a band at 780 keV, de-exciting by electron emission of intensity $(7\pm2)\times10^{-8}$. This agrees reasonable well with the value found by K x-ray electron coincidences[20], $(1.5\pm0.8)\times10^{-7}$. In the alpha spectrum in coincidence with gamma rays of energy above 680 keV, the peak is found again with intensity $(1.8\pm0.7)\times10^{-7}$. The total intensity is $(2.5\pm0.8)\times10^{-7}$, corresponding to HF$=25\pm8$. The level decays by 28% electron emission and 72% gamma emission. This demonstrates the occurrence of an $E0$ transition and, thus, of a beta vibrational state (or band). The least hindered alpha group in Pu^{239} has HF$=2.5$, so relative to this group the HF for the alpha decay to the 780-keV state is only 10 ± 3. The least hindered group populates the well-known $1/2+$ isomeric level in U^{235}, a few electron volts above the $7/2-$ ground state. The logical assignment to the 780-keV level is then $1/2+$. It is interesting to consider the possibility of another beta vibrational band based upon the ground state of U^{235}, one which could be reached by Coulomb excitation.

If the discriminator level for the gamma counter is lowered to 580 keV, a peak at 4.51 MeV is observed corresponding to a level at 650 (±20) keV. Gamma radiation in this energy range has been previously observed.[21] Our intensity $(8\pm3)\times10^{-7}$, corresponds to a HF of 75 ± 30, or 25 ± 8 relative to the least hindered alpha group. The 4.51-MeV peak is not seen in the electron run; the conversion coefficient derived is equal to or less than 0.01 which allows an unambiguous $E1$ assignment. We have assigned the state as $K=1/2-$ in analogy to the $1-$ states in even-even nuclei (see Fig. 5). A similar type of assignment has been made in the energy levels of Ac^{225} populated by Pa^{229} alpha decay,[22] and for levels in U^{239} at 690 keV and at 665 keV deduced from measurements of gamma rays following thermal neutron capture in U^{238}.[23]

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

Several levels above the ground-state rotational band were previously known in Th^{230}: a $01-$ state[24,25] at 508 keV, a $00+$ state[5,25,26] at 634 keV, and a $22+$ state[25] at 783 keV. Figure 14 shows the alpha spectrum found in coincidence with electrons in the present work. The energy of the state in Th^{230} populated by the 4.12-MeV alpha particles, (650 ± 20) keV, is in good agreement with the value deduced from U^{234} gamma ray[24] and electron spectra,[5] ~635 keV, and a more precise value of 634 keV, measured in the electron capture decay[25] of Pa^{230}. The value for the intensity of α_{650} in coincidence with electrons, $(2.1\pm0.5)\times10^{-7}$, is higher than previously reported,[5] 6×10^{-8} for the K line. The gamma spectrum measured in coincidence with $3.98\rightarrow4.38$ MeV alpha particles (see Table III) was in good agreement with the results of a measurement of the singles gamma spectrum[24] of U^{234}.

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

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

The measurements were complicated by the presence of spontaneous fissions (3% of all Cf^{252} alpha decays). These produced abundant electrons and gamma rays which contributed not only chance coincidences with alpha particles but also true coincidences with the fission fragments making it difficult to observe very

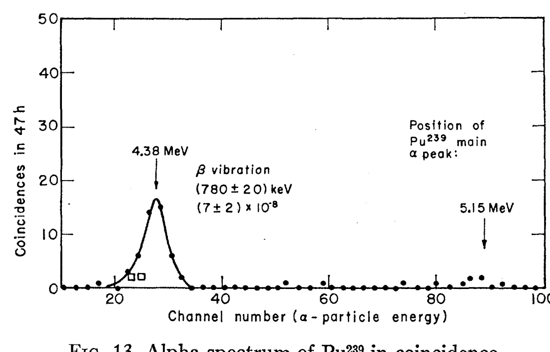


FIG. 13. Alpha spectrum of Pu^{239} in coincidence with electrons >400 keV.

[20] S. Bjørnholm and F. Asaro, Lawrence Radiation Laboratory Chemistry Division Annual Report UCRL-9566, 1960 (unpublished), p. 46.

[21] E. L. Murri and J. E. Cline, MTR-ETR Technical Branches Quarterly Reports, 4th Quarter, 1960, IDO-1665 (unpublished) and 1st Quarter, 1961, IDO-16695 (unpublished).

[22] M. W. Hill, Ph.D. thesis, University of California Lawrence Radiation Laboratory Report UCRL-8423, 1958 (unpublished).

[23] G. T. Emery, N. F. Fiebiger, and W. R. Kane, Bull. Am. Phys. Soc. **7**, 11 (1962).

[24] F. S. Stephens, Jr., F. Asaro, and I. Perlman (unpublished) as quoted by D. Strominger, J. M. Hollander, and G. T. Seaborg, Rev. Mod. Phys. **30**, 585 (1958).

[25] E. Arbman and O. B. Nielsen (to be published).

[26] F. S. Stephens, Jr. (unpublished) reported in reference 5.

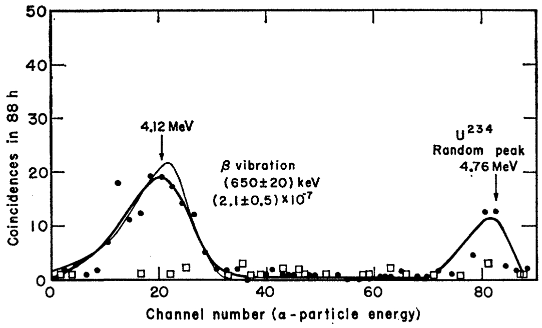


FIG. 14. Alpha spectrum of U^{234} in coincidence with electrons >320 keV. The shape of the alpha groups to the ground state as recorded in the singles spectrum has been drawn at the position of the 4.12-MeV peak for comparison (thin line). The squares refer to a control experiment in which the response of the anthracene detector to gamma rays is recorded separately.

low-intensity alpha groups. Saturated pulses from the high-energy fission fragments triggered the zero crossover pick-off unit later than those from the lower energy alpha pulses. It was, thus, possible to stop most of the high-energy fission pulses from passing through the linear gate and flooding the biased amplifier which was sensitive to high count rates.

The alpha spectrum in coincidence with gamma rays of energy greater than 225 keV gives indication of an alpha group of abundance 1×10^{-5} going to an excited state at 680 keV. More prudently, it can be said that there is no evidence for the population of excited states above 400 keV decaying directly to the ground-state band by transitions of intensity greater than 2×10^{-5}. The corresponding limit for an alpha electron measurement, discriminating at 400-keV electron energy, is 6×10^{-7}.

From this, it is easily calculated that there are no unhindered alpha transitions to levels in Cm^{248} between 820 and 400 keV. There are no alpha transitions with HF$\leqslant$10 to levels between 660 and 400 keV, etc. If we define a "regular" beta vibrational state as having HF$\leqslant$10 and decaying by fast $E0$ and $E2$ transitions in equal intensity, we can say that there is no "regular" beta band in Cm^{248} between 860 and 400 keV.

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

From an alpha spectrum measured in coincidence with gamma rays of energies above 460 keV there is questionable indication of a level at 980 keV populated in an intensity 2×10^{-6}. More safely a limit of 4×10^{-6} is established. A measurement with electrons of energy above 400 keV gives a limit of 1×10^{-7}. The limits apply to levels above 700 keV. Below this energy the spectrum is obscured by a rotational band built on a 388 keV intrinsic state and decaying by $M1$ transitions.

It follows that no unhindered alpha groups populate states in Bk^{249} (decaying within 0.02 μsec by radiation of energy greater than 460 keV) between 1070 and 700 keV. No alpha transitions with HF<10 goes to levels between 900 and 700 keV, etc. It can also be said that there is no "regular" beta band below 1120 keV.

TABLE III. U^{234} gamma rays in coincidence with alpha particles.

γ energy (keV)	Intensity per U^{234} α decay	Previous work[a]
460 } 510 }	$(4\pm1)\times10^{-7}$	$\sim 3\times10^{-7}$
585	$(1.2\pm0.5)\times10^{-7}$	$\sim1.6\times10^{-7}$
(740)	$<0.3\times10^{-7}$	···

[a] See references 24 and 26.

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

A limit of 7×10^{-6} can be established from an alpha spectrum measured in coincidence with gamma energies above 540 keV. A limit of 3×10^{-6} resulted from a measurement with electron energies above 500 keV.

There is, thus, no unhindered alpha transition to levels in Cf^{250} between 1080 and 540 keV, and no alpha group with HF<10 populates levels between 880 and 540 keV, etc. There is no "regular" beta band below 930 keV.

V. SUMMARY AND DISCUSSION

Figure 5 summarizes the results. The information obtained for the octupole bands and the gamma vibrational bands does not go substantially beyond what is already known and will need no discussion here.

A quantity of interest[27] for the characterization of the 00+ beta vibrational excitation is the ratio of the $E0$ K-conversion electron intensity to the $E2$ gamma intensity:

$$\mu_K(0'\rightarrow2)=W_K(E0)/W_\gamma(E2).$$

The experiment measures the total $E0$ intensity. The ratio of L to K conversion has been consistently found to be ≈0.2 in accord with theory. Allowing for conversion in higher shells, we will use $K/(K+L+M)\cdots=0.77$ as a correction factor in order to obtain the K-conversion intensity. Unfortunately, the energy resolution has not in general been sufficient to resolve the 00+ level from the 02+ rotational state. It is a reasonable assumption though, supported by the experiment, that at least 70% of the alpha decay goes to the 00+ level. The measurement, therefore, relates largely to the 00+ state. The possible error introduced in the experimental $\mu_K(0'\rightarrow2)$ is accordingly of the order of 20–40% at most. With this reservation, the data can be presented as shown in Table IV.

The alpha-decay hindrance factors are low with the exception of Th^{230}. No beta vibration have been observed in the nuclei lighter than Th^{230}. It would be interesting to know whether this has to do with a

[27] A. S. Reiner, Nucl. Phys. 27, 115 (1961).

TABLE IV. Summary of data for even-even alpha emitters.

Daughter nucleus	Excited state energy (keV)	Intensity $E2(\gamma)$	Intensity $E0(Ke^-)$	$\mu_K(0' \to 2)$	α hindrance factor (HF)
Th^{230}	650 ± 20	$(1.2\pm0.5)\times10^{-7}$	$(1.5\pm0.4)\times10^{-7}$	1.3 ± 0.6	40 ± 10
U^{234}[a]	810 ± 10	5×10^{-7}	5.4×10^{-7}	1.1	4
U^{235}	780 ± 20	$(1.8\pm0.7)\times10^{-7}$	$(5.4\pm1.2)\times10^{-8}$	0.3 ± 0.1	10 ± 3[b]
Pu^{238}	940 ± 20	$(2.6\pm0.8)\times10^{-7}$	$(2.6\pm0.6)\times10^{-7}$	1.0 ± 0.3	10 ± 3
Pu^{240}	870 ± 15	$(6.5\pm1.5)\times10^{-7}$	$(6.4\pm1.5)\times10^{-8}$	0.1 ± 0.03	3 ± 0.5

[a] No errors have been quoted as the intensities were used for calibration of the apparatus.
[b] Relative to the least hindered transition.

systematic increase in the hindrance factor and/or the excitation energy.

The 780-keV state in U^{235} is similar to the 00+ state in the neighbor, U^{234}, although μ_K is somewhat smaller. The well-established interpretation of the low-energy level spectrum of a deformed odd-A nucleus as resulting from a single particle coupled to an even-even core in which the particle can be excited independently can thus, very logically, be extended to include low-energy excitations of the core. It would be interesting, and it seems possible, to investigate other odd-A alpha emitters in the same mass region.

A startling feature of present studies is the drop in μ_K by a factor of 10 occurring between Pu^{238} and Pu^{240}. It is very difficult to understand this in terms of a purely collective model of adiabatic vibrations. It gives evidence for an inability to distinguish the collective vibrational aspect from the particle aspect in any general way. Keeping in mind the lack of success in identifying "regular" beta vibrations in the still heavier nuclei, one might speculate that the well-established vibrational states found in the region from Th^{230} to Pu^{238} may not be general for all nuclei in the regions of stable deformations. An alternative description in terms of quasiparticles may be fruitful[28–31]; e.g., Voros *et al.*[29] have calculated that a 0+ quasiparticle state should lie as low as 1.2 MeV for Pu^{240} and Cm^{244}. It is interesting to note that a recent theoretical treatment of the vibrational quadrupole excitations by Marshalek[30] shows that the prevalent collective nature of the vibrational states found in the region from Th^{230} to Pu^{238} is considerably weakened for those nuclei which have higher masses, but remain below the 152 neutron subshell.

[28] C. J. Gallagher, Jr. and V. G. Soloviev, Kgl. Danske Videnskab. Selskab, Mat. Fys. Skrifter **2**, No. 2 (1962).
[29] T. Voros, V. G. Soloviev, and T. Siklos (unpublished).
[30] M. G. Urin and D. F. Zaretsky, Nucl. Phys. **35**, 219 (1962).
[31] E. R. Marshalek, Ph.D. thesis, University of California Lawrence Radiation Laboratory Report UCRL-10046, 1962 (unpublished).

ACKNOWLEDGMENTS

It is a great pleasure to acknowledge the extensive and cordially extended help which has been received from many sources: the mechanical workshop, accelerator technicians, the secretarial staff, and the Information Division among others.

The experiment has relied heavily on the development of adequate electronic equipment and on the availability of the semiconductor counters. We are profoundly indebted to the electronics staff under the direction of F. Goulding and M. Nakamura for the development of the alpha counting equipment and to W. L. Hansen and his group for the development of the alpha detectors.

The Cf^{252}, E^{253}, and Fm^{254} were made available at the time of our measurements from a campaign in which heavy elements were separated following a prolonged neutron irradiation of 100 mg of Cm^{244}. We are indebted to Dr. Sherman Fried, Patrick Howe, and members of the Lawrence Radiation Laboratory Health Chemistry Division for their considerable efforts in carrying out these separations.

One of us, S. B., would like to express his gratitude to the Lawrence Radiation Laboratory for the hospitality he had enjoyed and for many stimulating discussions with members of its staff.