States of Even-Even Nuclei in the Near-Harmonic Region: Spectra of Rn^{218}, Rn^{220}, and Rn^{222}†

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Gamma-ray singles and coincidence spectra have been measured in the alpha decay of Ra^{222}, Ra^{224}, and Ra^{226}. Excluding the prominent transitions from the first excited states, the energies (and abundances relative to total α emission) of the observed radiations were: Ra^{222}: 325 kev (8.4×10^{-5}), 475 kev (7×10^{-5}), 525 kev (2×10^{-5}), and 798 kev (2.5×10^{-4}); Ra^{224}: 290 kev (9×10^{-5}), 410 kev (4×10^{-5}), 650 kev (6×10^{-5}); Ra^{226}: 260 kev (0.9×10^{-4}), 420 kev (7×10^{-6}), 450 kev (3×10^{-6}), 610 kev (1.0×10^{-5}). The observed gamma-gamma coincidences were Ra^{222}: 325–325 kev, 325–475 kev, and 325–525 kev; Ra^{224}: 241–290 kev and 241–410 kev; Ra^{226}: 188–260 kev and 188–420 kev. It was not strictly determined that the Ra^{222} radiations were not due to other members of the Th^{226} family. These data have been used to deduce the following levels, spins, and parities in the daughter Rn nuclides: Rn^{218}: 650 kev, 2+; 800 kev, 1−; and, possibly, 850 kev, 4+. Rn^{220}: 530 kev, 2+; and 650 kev, 1−. Rn^{222}: 448 kev, 2+; and 610 kev, 1−. The 2+ states in Rn^{220} and Rn^{222} have been previously assigned by Scharff-Goldhaber. These results are incorporated in a general energy level systematics of even-even nuclei in the heavy element region.

INTRODUCTION

A HIGH degree of regularity has been observed among the energy level patterns of the even-even nuclei in the heavy element region.[1] From the very heaviest elements (californium and fermium) down through the isotopes of radium the only energy levels found below about 1 Mev are those belonging to the ground-state (even parity) rotational band, and to a rotational band (odd parity) based on a state having spin and parity 1−. (The implication that there are no particle states differing from the ground state below ~1 Mev in even-even nuclei has itself been the subject of important theoretical developments.)[2] For elements lower than radium there was little information available, and for this reason the present study of the level schemes of the radon isotopes was undertaken. Due to the proximity of radon (element 86) to the closed shell region of lead (element 82) it was suspected that some of the regularity observed in the heavier elements should begin to break down.

It was known from other work[3] that the first excited states of Rn^{218}, Rn^{220}, and Rn^{222} were at energies of 325, 241, and 188 kev, respectively. These energies are considerably larger than are found for the transuranium even-even nuclei, where all the first excited states lie between 40 and 45 kev. The rise in energy of this presumably analogous state is very regular and is due to the approach to the region of the doubly closed shell. Even within the radon isotopes, themselves, a sharp rise in energy is observed in progressing to lower neutron numbers. The only additional information available on the energy levels of the radon isotopes is some recent work on Ra^{226} decay by Stephens[4] and by Harbottle, McKeown and Scharff-Goldhaber.[5] Stephens (in a preliminary report of this work) observed a gamma ray of 255 kev in coincidence with Ra^{226} alpha particles, and Harbottle et al. showed that this gamma ray had an energy of 260±3 kev and was in coincidence with the 188-kev transition. Thus, a level is defined at 450 kev which Scharff-Goldhaber[6] has suggested to be a state with spin and parity 2+. This state is presumed to be analogous to the 2+ second excited states in those isotopes further down in the periodic table which lie between the spherical (closed shell) and spheroidal (rotational) regions. This has been called the *near-harmonic* region by Scharff-Goldhaber, who has pointed out that it might be expected to occur in the heavy elements in just the region of the radon isotopes.

In the present work we have examined the level schemes of Rn^{218}, Rn^{220}, and Rn^{222} as they are populated by the alpha decay of their respective parent radium isotopes. Since the alpha groups leading to the levels of interest were too weak to observe directly, the level schemes were inferred from studies made on the gamma rays accompanying the alpha decay. Sodium iodide (Tl) crystals $1\frac{1}{2}$ inches in diameter by one inch thick were used in a coincidence array with pulse-height discriminators to analyze the energies. A single-channel pulse-height analyzer receiving the output from one of the crystal assemblies could be used to provide the gating pulse for a 50-channel analyzer connected to the other, making possible a variety of coincidence

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

[1] Information summarized by I. Perlman and J. O. Rasmussen, *Handbuch der Physik* (Springer-Verlag, Berlin, 1957), Vol. XLII.

[2] B. R. Mottelson, *Proceedings of the Rehovoth Conference on Nuclear Structure*, edited by H. J. Lipkin (North-Holland Publishing Company, Amsterdam, 1958), p. 83.

[3] D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 585 (1958). The authors summarize the work of various investigators.

[4] F. S. Stephens, Jr., Ph.D. thesis, University of California Lawrence Radiation Laboratory Report UCRL-2970, 1955, p. 69 (unpublished).

[5] G. Harbottle, M. McKeown, and G. Scharff-Goldhaber, Phys. Rev. **103**, 1776 (1956).

[6] G. Scharff-Goldhaber, Phys. Rev. **103**, 837 (1956).

experiments. This apparatus has been described in detail elsewhere.[4] The principal difficulty in these measurements was in obtaining sufficient separation of the radium isotopes from their decay products in order to see the low-intensity gamma rays belonging with the radium decay. Since the methods of purification employed varied for the different isotopes, they will be discussed individually in the appropriate sections.

EXPERIMENTAL

Rn^{218}

Information on the energy levels of Rn^{218} may be obtained by studying Ra^{222}, a 38-second alpha emitter. Because of this short half-life Ra^{222} cannot conveniently be isolated for study. It may, however, be obtained in equilibrium with its 31-minute parent, Th^{226}, which in turn can be kept in equilibrium with its parent, 21-day U^{230}, or readily separated therefrom. The U^{230} used in this work was prepared by bombarding Th^{232} with 100-Mev protons on the 184-inch cyclotron. The 18-day Pa^{230} so formed was allowed to stand for about a month, after which the U^{230} was removed from it. Protactinium-230 decays by β^- emission[3] to U^{230} to the extent of $\sim 15\%$; the other 85% decays by electron capture.[3]

The chemical purification of the U^{230} consisted of a combination of ion exchange and ether extraction techniques. The thorium target was dissolved in $10M$ HCl to which a few drops of hydrofluosilicic acid had been added. This solution was run through a Dowex A-1 anion-resin column which retains the uranium and protactinium under these conditions, while the thorium passes through. The uranium fraction, free from thorium, was eluted from the column in $1.4N$ HCl, during which a partial separation between uranium and protactinium was also achieved. The resulting solution was evaporated to dryness, and the uranium separated from the protactinium by extracting with ether in contact with an aqueous layer saturated with NH_4NO_3 and slightly acidified ($0.007M$ HNO_3). Re-extraction of the aqueous layer and washing of the ether layers with saturated NH_4NO_3 produced a uranium fraction of good purity. The uranium was removed from the ether by washing with a small portion of distilled water.

The U^{230} produced and isolated in this manner contains appreciable amounts of U^{232} from the decay of Pa^{232} formed in the bombardment along with the Pa^{230}. Uranium-232, itself, did not interfere with the measurements, but the daughters of this isotope (Th^{228} and family) are very prolific gamma-ray emitters and did interfere. Since the half-life of Th^{228} is 1.9 years and that of Th^{226} is 31 minutes a tractable ratio could be achieved by cleaning the uranium of thorium (by ion exchange separation) a short time before measurements were to be made. The U^{230} source prepared by this method had an intensity of about 10^6 alpha disintegrations per minute.

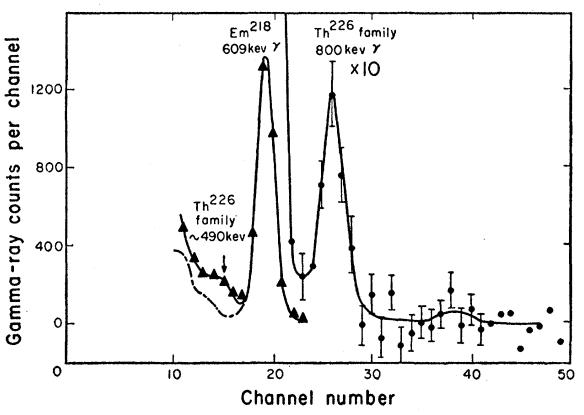


FIG. 1. High-energy gamma-ray spectrum of the U^{230} family. —·— Cs^{137} γ-ray distribution normalized to the 609-lev peak.

The alpha-particle and gamma-ray spectra of U^{230} and its daughters have been the subject of a previous paper.[7] and will not be reviewed in detail here.

Briefly: the decay of U^{230} is accompanied by several gamma rays of energy less than 250 kev all of which have been well placed in a level scheme; a closely-analogous level structure of Ra^{222} was deduced from the alpha and gamma spectra of Th^{226}; the spectra of Ra^{222} revealed a single excited state in Rn^{218} at 325 kev and from Rn^{218} decay the 609-kev state in Po^{214} (previously known from Bi^{214} beta decay) was seen. The present work is concerned with new low-intensity gamma rays belonging to the excited states of Rn^{218}, in addition to the 325-kev gamma ray which leads from the first excited state (2+) to ground.

Figure 1 shows the gamma-ray spectrum of the U^{230} family for the energy interval 400 to 1000 kev. In addition to the gamma ray of 325 and 609 kev, which have already been shown to belong with the decay of Ra^{222} and Rn^{218}, respectively, a gamma ray of 798 ± 5 kev is seen. Also there is an indication of a peak around 500 kev, although its energy and shape are not well defined. In a sample of Th^{226} separated from U^{230}, these same two peaks appeared, proving that these gamma rays are not due to U^{230} decay, itself.

Coincidence measurements were also made, and in Fig. 2, the photon spectrum in coincidence with the 325-kev gamma ray of Ra^{222} decay is shown. There a peak of the same energy as the gating photon appeared (325 kev) and also a complex peak around 500 kev, which is best resolved into a gamma ray at 475 kev with a less intense component, at 525 kev. The intensities of these gamma rays are shown in Table I and are based upon the value 3.6% for the intensity of the 325-kev γ.[7] It is seen that the abundance of the peak at 500 kev in the gamma-ray spectrum corresponds

[7] F. Asaro and I. Perlman, Phys. Rev. **104**, 91 (1956).

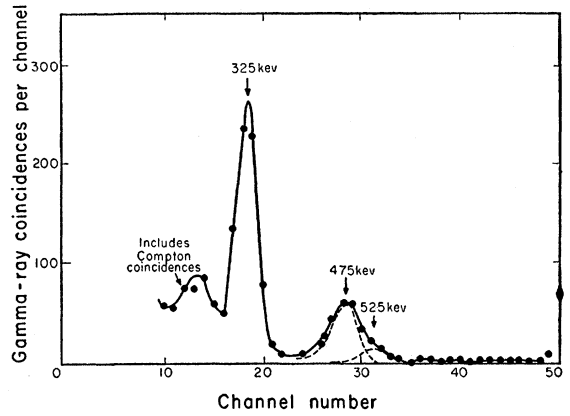


FIG. 2. U^{230} family gamma-ray spectrum in coincidence with the 325-kev gamma ray. — Resolved peaks. ◆ Integrates all events above channel 49.

well with the sum of the abundances of the 475- and 525-kev photons observed in the coincidence run.

It has been shown fairly conclusively that the 800- and 500-kev gamma rays belong in the Th^{226} family, since they were repeatedly observed in samples of Th^{226} milked from U^{230} sources, and in every case decayed with the 31-minute half-life of Th^{226}. It was not possible, however, to show unambiguously by these measurements to which member of this family they belong. The series is essentially terminated at 22-year Pb^{210}; so four possibilities exist: Th^{226}, Ra^{222}, Rn^{218}, and Po^{214}. Since the coincidences observed were with a gamma ray known to be in the Ra^{222} decay scheme, it is assumed that the three gamma rays seen in this manner belong also to Ra^{222} decay. The 798-kev gamma ray is nearly exactly the energy of the sum of the 475- and 325-kev transitions, which have been shown to be in coincidence with each other, and hence it seems likely that this gamma ray represents the crossover transition. It should be emphasized that these assignments are not proved experimentally, but it will be seen that if they are correct they lead to a readily interpretable level scheme of Rn^{218}, and one which resembles very closely the level schemes of Rn^{220} and Rn^{222}.

Once it has been concluded that the gamma rays belong in the decay of Ra^{222}, construction of the decay

TABLE I. Gamma rays in the Th^{226} family.

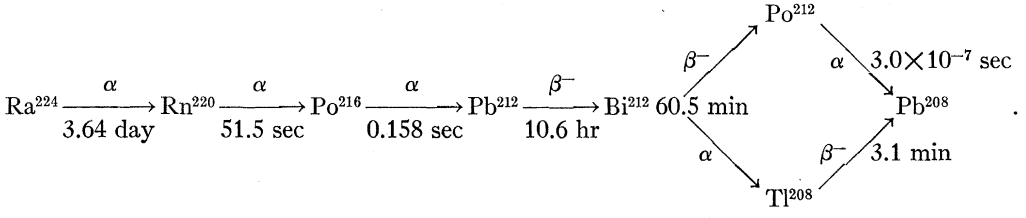
	Gamma energy (kev)	Intensity per alpha; based on a single alpha emitter
Gamma-ray spectrum	798±5	$(2.5\pm0.4)\times10^{-4}$
	~500	$(1\pm0.3)\times10^{-4}$
In coincidence with 325-kev gamma ray	325±5	$(8.4\pm1.5)\times10^{-5}$
	475±10	$(7\pm2)\times10^{-5}$
	525±10	$(2\pm1)\times10^{-5}$

scheme is not difficult. The gamma rays of energy 325, 475, and 525 kev, all of which are in coincidence with the 325-kev transition, must terminate at the 325-kev first excited state of Rn^{218}. That no two of these can be in cascade with each other is convincingly proved by considering alpha-decay theory. If, for example, the two 325-kev and the 475-kev transition were in cascade a level would be defined at ~1125 kev. The intensity of the 475-kev gamma ray of 7×10^{-5} would demand that this level be populated through alpha decay of Ra^{222} to at least that extent. This required alpha branching is more than an order of magnitude greater than would be expected for *unhindered* alpha decay characteristic of ground-state transitions of even-even nuclei. "Superallowed" alpha transitions defined in these terms have never been observed.

The level scheme of Rn^{218} which is consistent with the information at hand is shown in Fig. 3 and consists of states at 650, 800, and 850 kev in addition to the first excited state at 325 kev previously observed. The spin and parity assignments indicated will be discussed later.

Rn^{220}

The energy levels of Rn^{220} may be studied from the alpha decay of Ra^{224}. This isotope has a 3.6-day half-life, and is a member of the naturally occurring thorium family in which its immediate parent, 1.9-year Th^{228} serves as a convenient source. The separation of Ra^{224} from Th^{228} is accomplished readily, and its half-life (3.64 days) is convenient for measurement of the radiations. However, the descendents of Ra^{224} grow in rapidly and among these are products with gamma rays in high intensity. The decay series is as follows:



In first approximation Rn^{220}, Po^{216}, and Po^{212} are not troublesome because their gamma rays are in low intensity but Pb^{212} and Bi^{212} must be removed rigorously. It will be seen, however, that once these radiation sources are eliminated the relatively low intensity 545-kev gamma ray accompanying Rn^{220} decay becomes

limiting. Since we are concerned with gamma rays from Ra^{224} decay with intensities of a few parts in 10^5 or less it can readily be shown that some type of continuous purification would be necessary.

Of several continuous flow processes tried, the most successful will be described. The Ra^{224} was first milked from Th^{228} using a Dowex-50 cation resin column. The mixture of activities was put on the column in about $0.5N$ HNO_3 and elution with $4N$ HNO_3 brought the radium through in the first few column volumes leaving the thorium on the column quantitatively. The radium solution (containing lead and bismuth) was then evaporated to dryness and redissolved in about 0.2 ml of a $1.5N$ HCl solution. This solution was run through a very small Dowex A-1 anion resin column which was washed with an equal volume of $1.5N$ HCl. Under these conditions over 90% of the radium passed through the column while the lead and bismuth were retained quantitatively. This step served to purify the Ra^{224}, but in order to keep it pure continuously during measurement of the gamma rays the HCl solution was fed directly into a small *cation* resin column (1 inch long$\times\frac{3}{16}$-inch outer diameter) which was mounted between two sodium iodide crystals. By passing $1.5M$ HCl through the column at a rate of 30–50 drops per minute the bismuth and lead, as they grew in, were continuously removed. (The radium also passed through the column slowly but was retained long enough to permit measurement.) The flow of dilute HCl also kept the 52-sec Rn^{220} at a below-equilibrium level.

The gamma-ray spectrum taken in this manner is shown in Fig. 4(a). In addition to the prominent gamma ray of Ra^{224} at 241 kev, peaks are seen at 545 and 650 kev, with some evidence of an unresolved peak around 400 kev. A detailed resolution of this 400-kev peak was not attempted, since it was found to be much easier to study this gamma ray in the coincidence spectra, as will shortly be seen. A gamma ray of 542 kev has been reported in the decay of Rn^{220} to Po^{216},[8] and for this reason, it was suspected that the 545-kev peak in Fig. 4(a) was due to this isotope. This suspicion was confirmed when the flow of acid through the column was stopped and after 3 to 4 minutes the gamma-ray spectrum retaken. This spectrum is shown in Fig. 4(b), and has been normalized to the same running time as Fig. 4(a). The intensity of the 545-kev peak has increased by a factor of roughly five, while the 241- and 650-kev peaks have remained essentially constant. This indicates that the 545-kev peak grows into the radium sample, and is very probably the Rn^{220} gamma ray. On the other hand this measurement shows conclusively that the

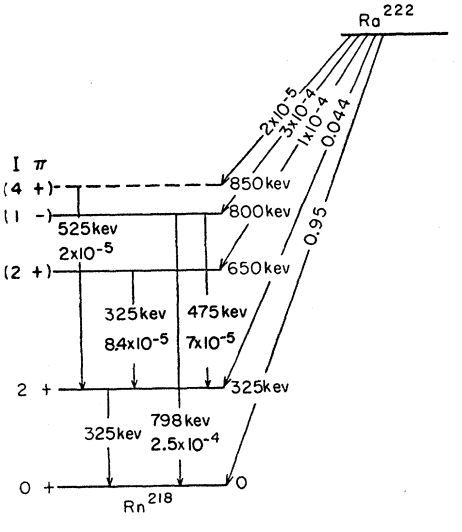


FIG. 3. Decay scheme of Ra^{222}.

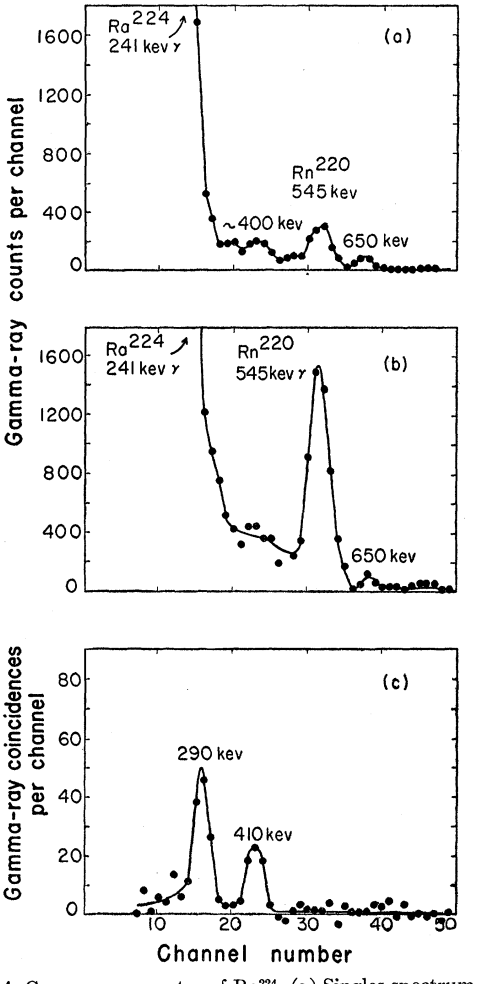


FIG. 4. Gamma-ray spectra of Ra^{224}. (a) Singles spectrum taken with $1.5N$ HCl continuously purifying the radium. (b) Singles spectrum taken 3–4 min after the acid flow was stopped. The spectrum was normalized to the same running time as in Fig. 4(a). (c) Gamma-ray spectrum in coincidence with the 241-kev gamma ray of Ra^{224}. The radium was purified continuously during the measurement.

[8] L. Madansky and F. Rasetti, Phys. Rev. **102**, 464 (1956).

650-kev gamma ray cannot belong to Rn^{220}, or to Po^{216} which will be in equilibrium with Rn^{220}. Additional spectra showed that there were very prominent gamma rays of Pb^{212} and Bi^{212} which grew into the radium sample a short time after the spectrum of Fig. 4(b) was taken. Since the most intense of the lead and bismuth peaks are not seen in Figs. 4(a) or 4(b), there can be no observable lead or bismuth contributions in these spectra. Thus the only member of the Ra^{224} family to which the 650-kev gamma ray can belong is Ra^{224} itself.

It was possible to obtain confirmatory evidence that the 650-kev gamma ray does belong to the Ra^{224} family. It was mentioned earlier that at a flow rate of 30 to 50 drops per minute the radium remained on the column only for a limited time (about 30 minutes). The time at which the radium eluted off the column could be determined precisely by watching the 241-kev gamma ray. It was found that in each of several experiments the 650-kev peak disappeared at just the time the radium left the column. Furthermore, in these experiments the radium was obtained by successively milking the same Th^{228} source, so that the 650-kev gamma ray must belong to an isotope which repeatedly grows into this source. It therefore seems very likely that this gamma ray belongs in the decay of Ra^{224}.

It is difficult to calculate the abundance of the 650-kev gamma ray from these experiments. Comparison of peak heights of the 650- and 241-kev photons could not yield accurate results because absorbers were used to attenuate strongly the 241-kev photons in order that the high counting rate not overload the electronic system. It is, however, possible to obtain a good intensity ratio between the 545-kev peak of Rn^{220} and the 650-kev peak of Ra^{224}, since for energies this high the absorber corrections are not large. The 650-kev photons are thus calculated to be $4.8\pm0.5\%$ of the 545-kev photons when the radon is in equilibrium with the radium [Fig. 4(b)]. In order to convert this ratio into an abundance for the 650-kev gamma ray, the abundance of the radon gamma ray must be known. The two values in the literature for this latter abundance are 0.3%[9] and 0.025%[8] per alpha disintegration of Rn^{220}. The first measurement was the intensity of the alpha group leading to the 650-kev state and the second was based upon a measurement of the gamma-ray abundance. These values are, unfortunately, in poor agreement. For this reason an attempt was made to redetermine the abundance of the 545-kev gamma ray. Because of difficulties which will not be described here, these experiments were not very successful; however, the number 1.2×10^{-3} photon per Rn^{220} alpha disintegration was obtained. This number is probably accurate to within a factor of 2. The abundance of the 650-kev gamma ray may then be calculated to be 6×10^{-5} per Ra^{224} alpha decay.

[9] F. Asaro and I. Perlman, reported in reference 3.

The gamma-ray spectrum in coincidence with the 241-kev photons was also examined. This spectrum is shown in Fig. 4(c), where peaks of 290 ± 5 and 410 ± 5 kev are seen. These two peaks, in about this relative intensity, appeared on each of several experiments using different methods of purification. Furthermore, when the continuous purification was stopped, it was observed that these peaks did not increase in intensity, and, in fact, after the daughters had grown in sufficiently it was no longer possible to distinguish them. For these reasons the peaks are believed to belong to Ra^{224}. From the number of 241-kev photon gates and a knowledge of the solid angle and counting efficiencies for the 290- and 410-kev peaks it was possible to calculate the abundances for these gamma rays as 9×10^{-5} and 4×10^{-5} per Ra^{224} alpha disintegration, respectively. The limits of error here should again be about a factor of two and are large due to uncertainties in the geometry of the continuous flow arrangement.

The decay scheme of Ra^{224} deduced from the experiments described here is shown in Fig. 5. The 650-kev level is defined rather unambiguously by the 410- and 650-kev gamma rays, and intensity considerations similar to those mentioned previously in discussing the Ra^{222} decay scheme rule out the possibility that the 290-kev transition terminates at the 650-kev level. Thus there seems to be no reasonable alternative to the decay scheme shown in Fig. 5.

Rn^{222}

The energy levels of Rn^{222} are populated in the alpha decay of naturally occurring Ra^{226}. The Ra^{226} used apparently was of high isotopic purity. Any Ra^{228} present as an impurity was below the limit of detection in these experiments, since no gamma rays that could be attributed to this isotope or its family were observed.

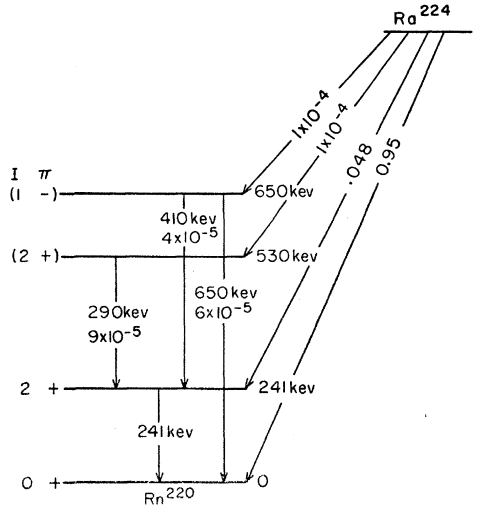


FIG. 5. Decay scheme of Ra^{226}.

The decay sequence of Ra^{226} is shown below:

$$Ra^{226} \xrightarrow[1622\ yr]{\alpha} Rn^{222} \xrightarrow[3.8\ day]{\alpha} Po^{218} \xrightarrow[3\ min]{\alpha} Pb^{214} \xrightarrow[27\ min]{\beta^-} Bi^{214} \xrightarrow[20\ min]{\beta^-} Po^{214} \xrightarrow[1.6\times10^{-4}\ sec]{\alpha} Pb^{210}.$$

The technique for singling out the gamma rays associated with Ra^{226} decay itself is somewhat different than for the other radium isotopes discussed. Here the radon is not a serious problem because of its relatively long half-life and because its most prominent gamma ray (510 kev) is in low abundance (7×10^{-4}). However, Pb^{214} and Bi^{214} have abundant gamma rays and grow into any Rn^{222} present rather quickly. The most successful process devised is as follows.

The bulk of the radioactivity due to the radon, polonium, bismuth, and lead was removed from the radium simply by evaporating the radium solutions to dryness in a centrifuge cone, and allowing the cone to remain heated for a period of several hours. This removed the radon gas and permitted the lead and bismuth to decay. The radium (usually around a milligram) was then dissolved in a minimum of 1.5*N* HCl and passed through a small Dowex A-1 anion resin column. The radium passed through but the bismuth and lead were adsorbed quantitatively. The radium solution from the column was diluted to several milliliters with a solution buffered to a *p*H of 5.0 to 5.5 with sodium acetate, and this buffered solution was then introduced into the continuous flow solvent extraction apparatus. In this case the continuous purification was effected by extracting with a 0.4*M* thenoyltrifluoroacetone (TTA) solution in benzene. Bismuth and lead readily extract into the benzene phase at this *p*H, whereas radium does not. The mixing chamber was a small sintered glass funnel (with vertical walls) into which the benzene phase was introduced from the bottom. The tiny bubbles of benzene solution emerging upward from the sintered glass disk caused a rather thorough mixing between the two phases, and the fact that the sintered glass disk was wet by the benzene prevented the aqueous radium solution from penetrating into it. The rate of flow was adjusted so that the benzene phase became continuous just at the top of the funnel and hence could be drawn off leaving a small hold-up of that phase in the zone where radiation detectors were placed. With funnels one to two inches in diameter, flow rates of the order of 30 ml per minute could be used. The limiting feature of this process was a slow build-up of lead and bismuth activity on the glass parts of the funnel, so that after about an hour it was generally no longer useful to continue the runs. The exact cause of this build-up was never determined, but the troublesome activity clearly remained in the funnel after the flow rate was increased to the point where the aqueous radium solution was carried over into the waste container. The gamma-ray counters were arranged and shielded so that they could observe only the funnel.

One of the best gamma-ray spectra of Ra^{226} obtained in the manner described above is shown in Fig. 6. Gamma rays ascribed to Ra^{226} are observed at energies of 260, 420, 610, and probably 450 kev. We can be certain that none of these peaks is due to daughter activities by the following reasoning. The only (and therefore most intense) gamma ray known in either Rn^{222} or Po^{218} decay is the one of 510 kev belonging with Rn^{222}. Since no peak in Fig. 6 at this energy is observed, no interference would be expected from either of these two isotopes. When the flow rate of TTA solution was stopped, the first thing observed was the extremely rapid growth of peaks at 300 and 350 kev belonging to Pb^{214}. Within a few minutes these peaks overshadowed everything else in this energy region; so that it is clear that none of the other peaks in Fig. 6 could be due to Pb^{214}.

Bismuth-214 could also be observed to grow into the radium when the benzene flow was stopped, but this occurred much more slowly than the growth of Pb^{214}. In the energy region shown in Fig. 6, the only bismuth peak which could be observed was at 609 kev, which energy was not distinguishable from that of the 610 kev peak shown in Fig. 6. There are two rather conclusive arguments, however, by which we can show that the 610-kev peak in Fig. 6 is not due to Bi^{214}. The first argument hinges on the counting rate registered in an integrating channel which totalled all pulses whose energy was larger than the region shown in Fig. 6. There are a number of higher-energy gamma rays in Bi^{214} decay which would be recorded in this

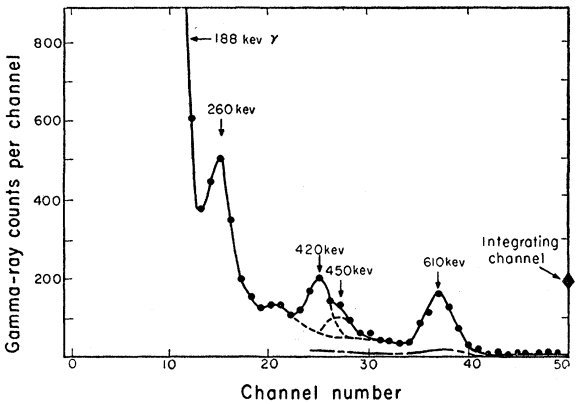


FIG. 6. Gamma-ray spectrum of Ra^{226} taken while the radium was continuously purified. —·— Maximum Bi^{214} gamma-ray spectrum normalized to the integrating channel. — Resolved peaks.

TABLE II. Gamma rays of Ra^{226}.

Energy (kev)	188	260±5	420±5	450±10(?)	610±5
Relative intensity	1	2.9×10^{-3}	2.1×10^{-4}	9×10^{-5}	3.3×10^{-4}
Absolute intensity per alpha	3.2×10^{-2} [a]	$(0.9\pm0.2)\times10^{-4}$	7×10^{-6}	3×10^{-6}	1.0×10^{-5}

[a] From previous data (see text).

integrating channel. Thus, once the bismuth had grown to the point where the spectrum of Fig. 6 was no longer discernible, the ratio of the peak height of the 609-kev gamma ray to the integrating channel was constant, as would be expected since both are due predominantly to Bi^{214}. If, then, we measure this ratio, we can come back to the spectrum of Fig. 6 and obtain a maximum height of the Bi^{214} 609-kev gamma ray. In fact, we can actually obtain a maximum height of the entire Bi^{214} spectrum, and this maximum has been indicated in Fig. 6. Below about 350 kev this has no significance since Pb^{214} is the chief contaminant, and the two are clearly not in equilibrium. The second argument that the 610-kev peak in Fig. 6 is not due to Bi^{214} comes from consideration of the expected background from energy gamma rays of Bi^{214}. Under the conditions of the experiment these will give rise to a rather flat distribution at energies above 610 kev of about half of the intensity of the 609-kev peak. At all times when the Bi^{214} spectrum was observed this continuum was present, but it is clearly not present in Fig. 6. Since the half-life of Po^{214} is so short, it would always be in equilibrium with Bi^{214}, and the arguments given above against any significant contribution of Bi^{214} to the spectrum of Fig. 6 also pertain to Po^{214}.

Although it is rather easy to show that the peaks in Fig. 6 are not due to the daughter of Ra^{226}, it is not easy to prove that they are not due to any other possible contaminant. Perhaps the best evidence in this regard is that these same peaks appeared when a different chemical purification was used (the column technique described in the section on Ra^{224}). The spectra were not nearly as good as that of Fig. 6, but there was good evidence that at least the 260-, 420-, and 610-kev peaks were present. Also, as we will mention later, some of these gamma rays are tied together by coincidence measurements. Finally, we can say that it was not possible to think of any impurity whose chemical and nuclear properties could have given these results. Thus we feel reasonably confident in assigning the gamma rays to Ra^{226}.

As in the case of Ra^{224} decay, the determination of the gamma-ray intensities was a difficult problem. The 188-kev photon could function as a standard but large amounts of absorber were necessary to reduce its intensity to the point where the counting equipment worked well, and the corrections are always uncertain where large attenuations are involved. The best relative intensities have been collected in Table II. In order to convert these to absolute intensities, it is necessary to know the intensity of the 188-kev photons. Using an alpha abundance of 5.7% to the 188-kev level,[10] and a conversion coefficient of 0.80 for the 188-kev transition,[11,12] the abundances given in the third row of Table II were calculated. The intensity of the 260-kev gamma ray could be determined with reasonable precision (estimated 20%) because its relatively high abundance permitted its comparison with the 188-kev gamma ray using small samples with little or no absorber. Concerning the higher-energy gamma rays, it would be very difficult to obtain a meaningful limit of error. We would estimate, however, that the absolute abundances should be accurate to within a factor of two. The relative intensities of these three gamma rays should be somewhat better than this.

It was difficult to make coincidence measurements on Ra^{226} because of the severe counting-rate limitations of the equipment. We can say definitely, however, that the 188- and 260-kev transitions are in coincidence, as was found by Harbottle et al. Also there was good indication that the 420-kev peak was in coincidence with the 188-kev transition; whereas the 610-kev peak was not. It is not possible to be certain of these conclusions, however, and no data could be obtained on the 450-kev transition.

All of the observed gamma rays fit well into a level scheme of Rn^{222}. Harbottle et al. have already concluded that there must be a level at 448 kev, and the present data strongly support that conclusion. It is very unlikely that the 610-kev transition terminates anywhere but the ground state, because of the super-allowed-alpha-transition arguments which have been mentioned previously. Thus, a level is defined at 610 kev, and the 420-kev transition fits well between this level and the one of 188 kev. If the 450-kev gamma ray is real, it seems likely that it represents the cross-over transition from the 448-kev level to the ground state. The level pattern of Rn^{222} from the decay of Ra^{226} is shown in Fig. 7.

[10] F. Asaro and I. Perlman, Phys. Rev. 88, 129 (1952).

[11] L. A. Sliv and I. M. Band, Leningrad Physico-Technical Institute Reports, 1956 [translation: Report 57 ICCKI and Report 58ICCL1 issued by Physics Department, University of Illinois, Urbana, Illinois (unpublished)].

[12] M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958), p. 145. This work contains calculated M-shell conversion coefficients. We assumed N and higher order shells would be 40% of the total M conversion coefficient.

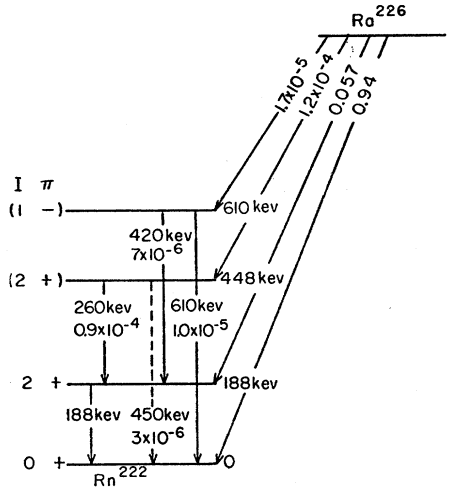


FIG. 7. Decay scheme of Ra^{226}.

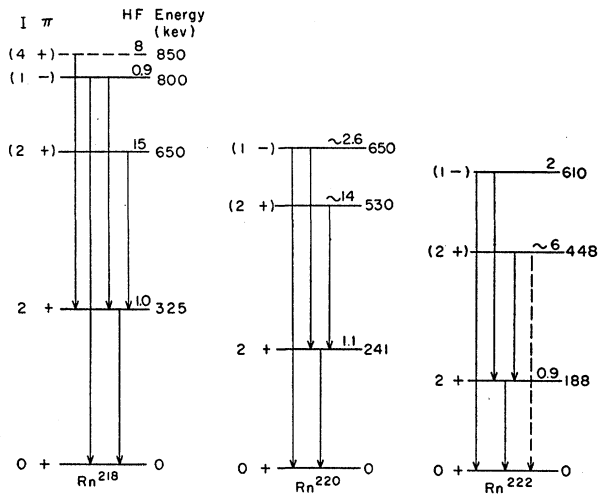


FIG. 8. Energy level assignments for Rn^{218}, Rn^{220}, and Rn^{222}. HF—Hindrance factors for alpha population to the various levels.

SPIN ASSIGNMENTS

In addition to the data previously mentioned, Fig. 8 contains the suggested spin assignments to be discussed here, and the "hindrance factors" (HF) for alpha decay to the various levels. A hindrance factor for alpha decay in an even-even nucleus is defined as the ratio of the experimental alpha half-life to a given level, divided by the half-life which is calculated using the one-body model of alpha decay and assuming decay to the ground state is unhindered (HF=1).[13] Thus, in effect, the one-body model of alpha decay is used to remove the energy dependence for alpha emission, and a "reduced alpha transition probability" is obtained whose inverse is the hindrance factor. The hindrance factor, as defined here, is independent of the spin of the excited state. Since all even-even nuclei undoubtedly have the same spin and parity (0+), one might expect that the hindrance factors for decay from this level to a given type of level in the daughter nucleus would vary in a systematic manner with the parent nucleus. This criterion in addition to gamma-ray data will be applied in making spin assignments for the radon isotopes.

The first excited states of each of the radon isotopes[14,15] has been previously shown to have spin and parity 2+ from alpha particle-gamma ray angular correlation measurements. These assignments are supported, again in each case, by conversion coefficient data[7,16,17]; so that the 2+ character of the three first excited states seems firmly established. The hindrance factors for alpha decay to these levels are 1.0, 1.1, and 0.9 for Ra^{222}, Ra^{224}, and Ra^{226}, respectively. Since the three levels involved here seem to be analogous levels in each of the three radon isotopes, it is not surprising that these hindrance factors are similar.

Scharff-Goldhaber has suggested that the 450-kev level of Rn^{222} is a state analogous to the second excited state of other even-even nuclei in the near-harmonic regions. This assignment is based on the ratio of the energy of this state to that of the first excited state (2.40), and on the mode of de-excitation of the level (predominantly to the first excited state). Two factors in the present work tend to support this assignment. The first is the possible weak gamma ray to the ground state, which rule out spins of 0+ or 4+ for the 450-kev level. The second factor is that levels analogous to this one seem to appear in Rn^{218} and Rn^{220} at 650 and 530 kev, respectively. In these cases the ratios of the energy of the level to that of the first excited state are 2.00 and 2.25, respectively, and again the observed decay of the level is to the first excited state. Thus, although the evidence is not entirely convincing, we shall tentatively assign spin and parity 2+ to these three levels.

The alpha hindrance factors to these second 2+ states are 15, ~14, and ~6 for Ra^{222}, Ra^{224}, and Ra^{226} decay, respectively. Although these vary by a factor of three, this is not considered disturbing. Other cases are known in which hindrance factors to analogous levels in adjacent even-even nuclei differ by a factor of two. It should also be pointed out that both in even-even nuclei and in odd nucleon cases, hindrance factors to nonanalogous states can differ by factors of tens, hundreds, and thousands. In this respect, there is no inconsistency with assigning similar character to the three levels. It is perhaps of interest to point out

[13] M. A. Preston, Phys. Rev. **71**, 865 (1947).
[14] F. S. Stephens, Jr., F. Asaro, and I. Perlman, Phys. Rev. **96**, 1568 (1954).
[15] J. C. D. Milton and J. S. Fraser, Phys. Rev. **95**, 628(A) (1954).
[16] S. Rosenblum, M. Valadares, and M. Guilott, J. phys. radium **15**, 129 (1954).
[17] G. Victor, J. Teillac, P. Falk-Vairant, and G. Boussieres, J. phys. radium **13**, 565 (1952); S. Rosenblum and M. Valadares, Compt. rend. **234**, 2359 (1952); M. K. Juric and D. M. Stanojevic, Bull. Inst. Nuclear Sci. Boris Kidrich **5**, 15 (1955).

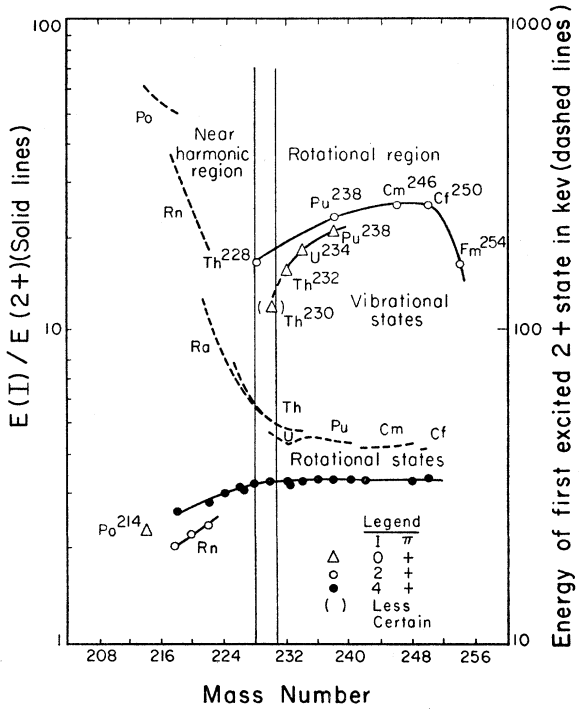


FIG. 9. Even parity states in even-even nuclides.

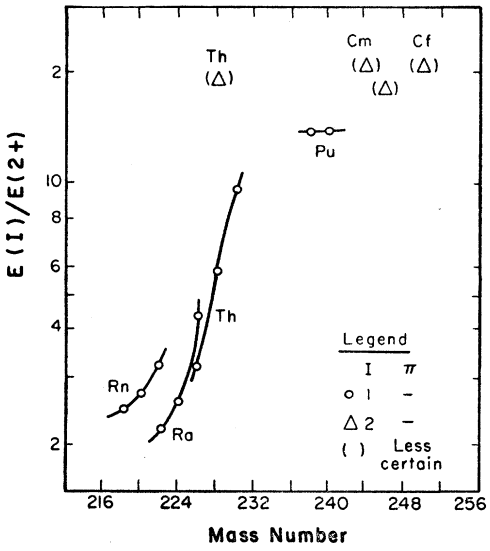


FIG. 10. Odd parity states in even-even nuclides.

that if the 450-kev gamma ray of Rn^{222} is real and does represent the transition from the second 2+ state (2′+) to the ground state, then the reduced transition probability ratio is:

$$B(E2)_{2'+\rightarrow 0+}/B(E2)_{2'+\rightarrow 2+}=2\times 10^{-3}.$$

This assumes, in addition, that both transitions are predominantly $E2$, as is generally found to be the case with near-harmonic nuclei. Although it should be clear that this number is quite tentative, it is, nevertheless, in the range found for other near-harmonic even-even nuclei.

In each of the radon isotopes there is a level which decays both to the ground state and to the first-excited state. These levels lie at energies of 800, 650, and 610 kev for Rn^{218}, Rn^{220}, and Rn^{222}, respectively, and have alpha hindrance factors of 0.9, ~2.6, and 2. These similarities again suggest that we are dealing with an analogous level in each of the three isotopes. Since each of these states de-excite to states of spins 0 and 2, the only reasonable values for the spin would be 1 or 2. Furthermore, since each is undoubtedly populated directly by alpha decay (from a 0+ parent ground state), it must have spin and parity either 1− or 2+ ($l=1$ or $l=2$ alpha wave). Of these possibilities we prefer a 1− assignment for the following reasons. States having spin and parity 1− are known to occur systematically in radium and thorium isotopes of this region. The hindrance factors for alpha decay to these levels decrease quite uniformly with decreasing mass number from a high of around 500 for U^{234} decay (heaviest uranium isotope known to populate a 1− state) to 4 for Th^{226} decay (lightest thorium isotope for which information is available).[18] Thus hindrance factors in the range 0.9 to ~2.6 for the decay of the radium isotopes to these 1− states seem quite reasonable. One of the most characteristic features of the 1− states is their de-excitation to the 0+ and 2+ states, respectively, with $E1$ reduced transition probability ratio always 0.50 within experimental uncertainty.[18] This has been interpreted to indicate that K, the projection of the spin on the nuclear symmetry axis, is zero for these levels.[18] If we examine the data on this point for the radon levels under consideration here, we find that $B(E1)_{1\rightarrow 0}/B(E1)_{1\rightarrow 2}$ is 0.7±0.2, 0.4±0.2, and 0.5±0.2 for Rn^{218}, Rn^{220}, and Rn^{222}, respectively. Again these data support the 1− assignments.

The only level not yet considered is the one at 850 kev in Rn^{218}. The ratio of the energy of this level to that of the first excited state (2.6), and the alpha hindrance factor (8) suggest that this may be the 4+ member of the ground-state rotational band. We have tentatively made this assignment.

DISCUSSION

There is now available a considerable amount of information on the excited states of even-even nuclei in the heavy-element region. Most of this information is summarized in Figs. 9 and 10. Figure 9 is a plot pertaining to some of the even-parity states and consists of the ratios of energies of these states to the first excited (2+) state. Plots similar to this have been given by Scharff-Goldhaber[6] and Sheline.[19] In the

[18] F. S. Stephens, Jr., F. Asaro, and I. Perlman, unpublished data; Phys. Rev. **100**, 1544 (1955).

[19] R. K. Sheline, *Proceedings of the University of Pittsburgh Conference on Nuclear Structure, 1957*, edited by S. Meshkov

rotational region, the familiar 4+/2+ ratio of 3.3 showing the $I(I+1)$ energy dependence is seen (lower curve). Higher members of this ground-state rotational band are not shown. At higher energies two different intrinsic states are seen and these have been attributed to beta vibrational states (0′+) and gamma vibrational states (2′+) predicted at about these energies by Bohr and Mottelson.[20] (Rotational levels based upon these states are not plotted.) A reason for the apparent sharp drop in energy for the gamma vibrational band in Fm^{254} has not been suggested, however.

The region of particular interest here is that between mass number 216 and 228. Scharff-Goldhaber has suggested that a rather sharp transition in the character of these energy levels occurs between proton numbers 86 (radon) and 88 (radium). However, if our assignment of the 4+ state in Rn^{218} is correct, the behavior of this level through the above region is rather uniform. It cannot be ascertained whether a sharp change in the 2′+ or 0′+ states occurs until the position of these levels in some of the radium isotopes is known. If the downward trend of the 0′+ state in Th^{230} is correct, a sharp break in the position of this level seems rather unlikely. Furthermore, the energy of the first-excited state (broken lines in Fig. 9) is quite smooth throughout this region. Our conclusion, then, is that a sudden change in the character or energy of the levels does not seem likely, although there are not yet sufficient data available to be certain of this conclusion.

Figure 10 is similar to Fig. 9 except that odd-parity levels are considered. The data on the 2− levels are fragmentary, and it is not yet at all certain that any of these levels really have spin and parity 2−, so that they will not be discussed. The 1− states are quite well established, however. In Fig. 10 the lines connect the points for a given element, and there is evidence of a break between the radon and radium isotopes. On the other hand, a break of similar magnitude occurs between radium and thorium, so that probably this only indicates that the position of these levels depends on both the proton and the neutron number. It has been suggested that these 1− levels are due to collective octopole vibrations; however, a satisfactory explanation for the sharp dip in the energy of these levels in the region of radium and thorium has not yet been given.

(University of Pittsburgh and Office of Ordnance Research, U. S. Army, 1957), p. 494.

[20] A. Bohr, Kgl. Danske Videnskab. Selskab, Mat-fys. Medd. **26**, 34 (1952); A. Bohr and B. R. Mottelson, *Beta- and Gamma-Ray Spectroscopy*, edited by Kai Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), p. 474.

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Electron Pair Production in π^-+d Capture*†

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The internal conversion coefficient $\rho(p)=(dW_{2e}/dp)/(dW_\gamma/dp)$ relating the π^-+d capture processes yielding $2n+e^++e^-$ and $2n+\gamma$ is calculated as a function of the n-n relative momentum p. It is found to be a slowly varying function of p, insensitive to the strength of the n-n force. The spectrum of the electron pair energies (or of the momentum p) therefore depends sensitively on the n-n scattering length, just as Watson and Stuart found to be the case for the photon spectrum. Thus, observation of the pair production process is an alternative method of measuring the n-n scattering length.

I. INTRODUCTION

IN the process

$$\pi^-+d\rightarrow 2n+\gamma, \qquad (1.1)$$

the relative frequency of high-energy photons depends on the strength and sign of the n-n interaction.[1] Thus, the more attractive the interaction, the more likely it will be for the two neutrons to be emitted with low relative momentum; and low neutron energy corresponds to high photon energy. An attempt has been made to determine the ^1S n-n scattering length from a measurement of the photon spectrum,[2] but only very wide limits could be set. This scattering length cannot be obtained from n-p or p-p data by the arguments of charge independence or charge symmetry, since it is very sensitive to slight differences in the nucleonic interactions such as may be expected to result from electromagnetic effects. It is, in fact, true that the (effective nuclear) scattering lengths for the n-p and p-p systems are significantly different, being -24f and -16f,[3] respectively; this difference corresponds to a difference in well depth of between one and three per-

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[2] R. Phillips and K. Crowe, Phys. Rev. **96**, 484 (1954).