Radiations from Four Radioactive Isotopes of Platinum*

V. R. Potnis,† C. E. Mandeville, and J. S. Burlew‡
Bartol Research Foundation of The Franklin Institute, Swarthmore, Pennsylvania

(Received September 22, 1955)

The gamma radiations of the 18-hr Pt^{197} have been examined with scintillation counting methods and have been found to consist of 77- and 191-kev quanta in coincidence and a 279-kev gamma ray which is noncoincident with other gamma rays. Beta-gamma coincidence studies have been performed and values of log *ft* have been obtained for the three beta-ray spectra of Pt^{197}. The radiations of Pt^{195m} include gamma rays of energies 31, 99, and 130 kev. The latter gamma ray is not to be confused with the highly converted one emitted in the 3.8-day isomeric transition and is the cross over of the 31-kev–99-kev cascade. Pt^{193m} emits a gamma ray of energy 130 kev. No quantum radiations of higher energies were detectable. Some coincidence measurements have been performed on the complex gamma-ray spectrum of Pt^{191}.

INTRODUCTION

THE radioactive isotopes of platinum have been produced by slow-neutron irradiation of platinum,[1] by fast-neutron bombardment of mercury,[2] and by deuteron bombardment of various heavy elements.[3,4] Three of the radionuclides so produced have approximately the same lifetimes (about 3.5 days). In the above mentioned studies, naturally occurring elements were usually employed as the target materials; so uncertainties existed in the determination of the radiation characteristics of the various radioelements. More recently, Cork *et al.*[5] have studied the radiations of Pt^{191}, Pt^{193m}, and Pt^{195m}, produced by slow-neutron irradiation of isotopically enriched targets of platinum. A magnetic spectrometer and scintillation counter were employed in these investigations.[5] To obtain the present measurements, four enriched isotopes of platinum of even mass number were irradiated by slow neutrons in the Brookhaven pile. The active materials were subsequently purified for the removal of any Hg, Au, and Ir. The radiations of Pt^{197}, Pt^{195m}, Pt^{193m}, and Pt^{191} have been studied with single scintillation spectrometers and by pairs of scintillation spectrometers in coincidence.

Cylindrical crystals of NaI(Tl) of height 3.5 cm and diameter 3.5 cm were employed for detection of gamma rays. Beta rays were counted in an anthracene crystal of thickness 1.16 cm. Uniformly thin beta-ray sources were deposited upon "Nu-Skin" foils of surface density less than 0.5 mg/cm². The scintillation spectrometers for gamma-ray measurements were calibrated at quantum energies of 87, 279, and 661 kev, the monochromatic radiations of Cd^{109}, Hg^{203}, and Cs^{137} respectively. The anthracene beta-ray counter was calibrated at 625 kev by the conversion line of the gamma ray of Cs^{137}.

PLATINUM-197

To obtain quantities of radioactive Pt^{197}, four samples of metallic platinum of weight 6 mg each were irradiated in succession at the Brookhaven pile. The isotopic enrichment of Pt^{196} was 66%. Pt^{197} ($T_{1/2}\sim18$ hr) decays by negatron emission to excited states of Au^{197}. The pulse-height distribution of the subsequently emitted gamma rays is shown in Fig. 1. Three gamma rays are clearly present with energies of 77, 191, and 279 kev, respectively. The relative intensities of the unconverted quantum radiations were calculated to be, in order of ascending energy, 30, 3.9, and 1. These gamma rays have been previously reported by Mihelich and de-Shalit[6] in the decay of Hg^{197} to Au^{197}. Earlier,[7] only the two softer gamma rays had been reported as emitted in the decay of both Hg^{197} and Au^{197}. The two harder quanta have also been observed in Coulomb

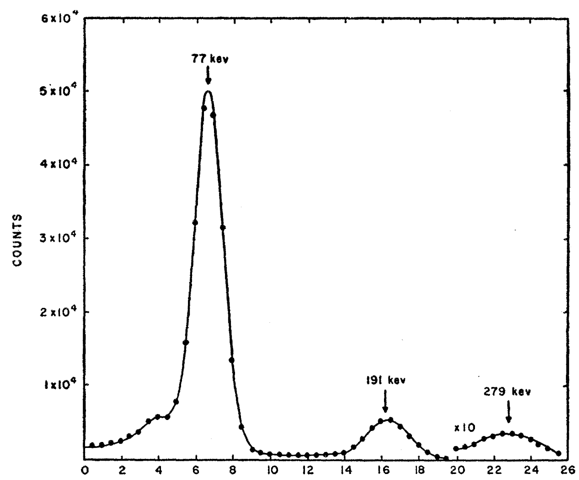

FIG. 1. Spectrum of the gamma rays of Pt^{197}.

* Assisted by the joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

† Research Fellow, Bartol. Permanent address, Gwalior (M.B.) India.

‡ The Franklin Institute, Philadelphia, Pennsylvania.

[1] McMillan, Kamen, and Ruben, Phys. Rev. **52**, 375 (1937).

[2] Sherr, Bainbridge, and Anderson, Phys. Rev. **60**, 473 (1941).

[3] J. M. Cork and E. O. Lawrence, Phys. Rev. **49**, 788 (1936).

[4] R. S. Krishnan and E. A. Nahum, Proc. Cambridge Phil. Soc. **37**, 422 (1941).

[5] Cork, Brice, Schmid, Hickman, and Nine, Phys. Rev. **94**, 1218 (1954).

[6] J. W. Mihelich and A. de-Shalit, Phys. Rev. **91**, 78 (1953).

[7] Huber, Humbel, Schneider and Zunti, Helv. Phys. Acta **24**, 127 (1951).

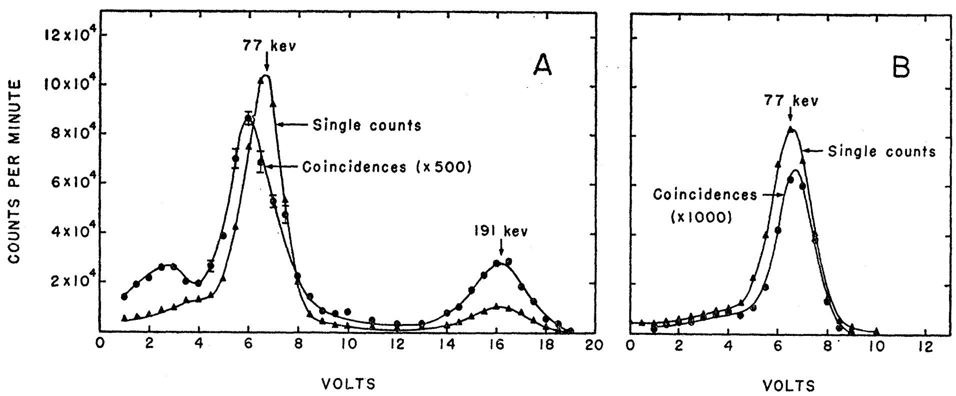

FIG. 2. Coincidence studies of the gamma rays of Pt^{197}; Curves in (A) with one counter fixed at 77 kev; Curves in (B) with one counter fixed at 191 kev.

excitation of gold by protons[8,9] and by alpha particles.[8] On initially considering the various gamma-ray energies and the accuracy of the measurements, it might be concluded that the 279-kev radiation is in actuality the cross-over transition of the 77-kev to 191-kev cascade. These latter two gamma rays have been previously reported to be in coincidence.[7] To determine accurately the energy of the hardest gamma ray, a careful comparison of the position in energy of its photopeak was made with that of the photopeak of the monochromatic 279-kev gamma ray of Hg^{203}. The positions were found to be identical. Precautions were taken to eliminate any possible gain shifts which might arise from differing counting rates of the two sources by equalizing their integral rates. From a consideration of the channel width of the scintillation spectrometer, it was concluded that the energies of the two gamma rays must differ at most by 3 kev. This fact, combined with the previously accurately determined values of the energies of the two softer gamma rays,[6,7] led to the conclusion that the 279-kev gamma ray could not be emitted in the cross over transition. A search was made for gamma rays of higher energies by setting the channel of the analyzer so that only gamma rays of 500 kev or more could be counted. No harder quanta were detected. From the background counting rate of the spectrometer, it was possible to conclude that any gamma rays present with a quantum energy of about 550 kev[9] could be emitted only with a probability of less than 10^{-4} per disintegration.

TABLE I. Experimentally determined conversion coefficients of the gamma rays of Pt^{197}.

Energy (kev)	77.4	191	279
Multipole order	$M1+E2$	$M1$	$M1$
α_T	3.66[a]	2.92[b]	0.315[a]
Relative intensity of the transition	139.8	15.0	1.32

[a] See reference 6.
[b] Present measurements.

The results of gamma-gamma coincidence measurements are shown in Fig. 2. To obtain the data of Fig. 2(A), the channel of one spectrometer was fixed at the photopeak of the 77-kev radiation while the channel of the other was moved over the entire region between 1 and 20 volts. Radiation falling in the fixed channel is clearly coincident with the 191-kev gamma rays. Coincidences were also observed as shown between the fixed channel and the K x-rays of Au which follow internal conversion of the 191-kev gamma ray. An escape peak is also in evidence at $\sim$38 kev on the coincidence curve, resulting from escape of the iodine K-line after detection of the x-rays of gold. To observe the coincidence curve of curves in Fig. 2(B), the channel of one spectrometer was fixed at 191 kev while the channel of the second was moved through the region of the 77-kev radiation. Coincidences were observed which are peaked at 77 kev with a few coincidences at lower energies which are related to escape radiation of the 77-kev quanta. Thus, the curves of Fig. 2 have established with complete certainty by the scintillation counter method the cascade relationship between the 191-kev and 77-kev radiations. The previously measured value[7] of 1.7 for the K-shell conversion coefficient of the 191-kev gamma ray has been shown to be incorrect by Mihelich.[6] This coefficient can be estimated from the curves of Fig. 2(A) and is calculated to have the value $\alpha_K=2.5$. This result was obtained by dividing the area on the coincidence curve under the photopeak of the 191-kev gamma ray into that portion of the area under the x-ray peak which results from coincidences between 77-kev gamma rays counted in the fixed channel and gold K-x-rays counted in the moving channel. This value of α_K is somewhat greater than the theoretically expected value[10] of $\sim$1. Were the 191-kev radiation an $E2-M1$ mixture, the K-shell conversion coefficient would be less than the theoretically indicated value. Thus, an assignment of $M1$ as the nature of the 191-kev transition is favored and furthermore agrees with the measured K/L ratio obtained by Cork *et al.*[11] The

[8] N. P. Heydenberg and G. M. Temmer, Phys. Rev. **93**, 906 (1954).
[9] Class, Cook, and Eisinger, Phys. Rev. **96**, 658 (1954); W. I. Goldburg and R. M. Williamson, Phys. Rev. **95**, 767 (1954).
[10] Rose, Goertzel, Spinrad, Harr, and Stone, Phys. Rev. **83**, 79 (1951).
[11] Cork, LeBlanc, Stumpf, and Nester, Phys. Rev. **86**, 415 (1952).

relative intensities of the three transitions involved may be calculated with the use of the previously quoted intensities of the unconverted quantum radiations and the total conversion coefficients given in Table I. The transition intensities so obtained are also included in Table I.

An intensive search was conducted to determine whether the 279-kev gamma ray might be in sequence with any other gamma rays. No coincidences between it and any other radiations were detected.

Using an anthracene counter as a beta-ray detector, beta-gamma coincidences were measured between the beta rays and each of the three gamma rays of Pt^{197}. To obtain coincidences, the gamma-ray channel was fixed upon a single photopeak while the beta-ray channel was moved over the beta-ray spectrum in differential intervals. The three sets of data are shown in Fig. 3

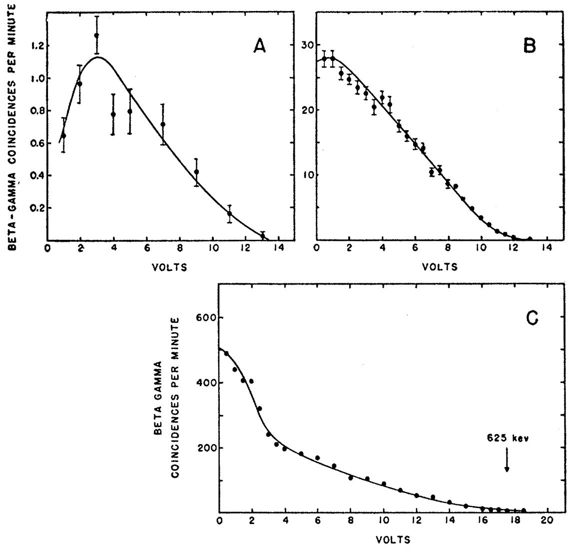

FIG. 3. Beta-gamma coincidences in the decay of Pt^{197}; Curve A, with the gamma-ray counter fixed at 279 kev; Curve B at 191 kev; Curve C at 77 kev.

where the beta-gamma coincidence curves are lettered in order of decreasing quantum energy. The end points of the beta-spectra are 468, 479, and 670 kev, the spectra terminating at levels in Au^{197} which are 279, 268, and 77 kev above the ground state. The relative intensities of these beta-ray spectra can be obtained from a consideration of the related gamma-ray transition intensities. These data are shown together with their respective values of $\log ft$ in Table II.

The disintegration scheme of Pt^{197} is shown in Fig. 4. The ground-state spin of Au^{197} has been previously measured[12] as 3/2, consistent with a shell model prediction of $d_{3/2}$. The same model moreover indicates $d_{5/2}$ as a possible orbital assignment for the 279-kev level. The two spin assignments for the above two levels are

[12] R. M. Elliot and J. Wulff, Phys. Rev. 55, 170 (1939).

TABLE II. Beta spectra of Pt^{197}.

Energy (kev)	Relative intensity (%)	$\log ft$
468	0.9	7.80
479	10.6	6.88
670	88.5	6.36

consistent with designation of the 279-kev transition as $M1$. Assignment of spins 1/2+ and 3/2+ to the 77-kev and 268-kev levels is also in agreement with shell model considerations and with the assigned multipole orders of the 77-kev and 191-kev gamma rays. The values of $\log ft$ for the various beta spectra are consistent with a first forbidden classification in each case. This forbiddenness is consistent in each case with that indicated by the spin values of the decay scheme. The absence[13] of a beta-ray transition to the ground state of Au^{197} ($p_{1/2}\rightarrow d_{3/2}$) is difficult to explain if only the last odd particle is considered as having a role in determining the nature of the transition. The orbitals of the ground state transition are such as to suggest a first forbidden transition of an intensity comparable with that of the three observed spectra. In a beta transition such as that of Pt^{197}, the last eight neutrons participate in a rearrangement of the core, $(i_{13/2})^8\rightarrow(p_{1/2})^2(i_{13/2})^6$. However, this alteration very likely also occurs in the transitions to the excited states, and therefore cannot explain the absence of the ground-state transition, unless it is for some reason more difficult to bring about in the case of the ground state. This large change of the orbitals of a pair of neutrons, $(i_{13/2})^2\rightarrow(p_{1/2})^2$ may offer some clue as to the absence of the transition.

PLATINUM-195m

The radiations of Pt^{195} have been observed in the decay[5,13] of Pt^{195m} ($T_{1/2}=3.8$ days) as well as in de-

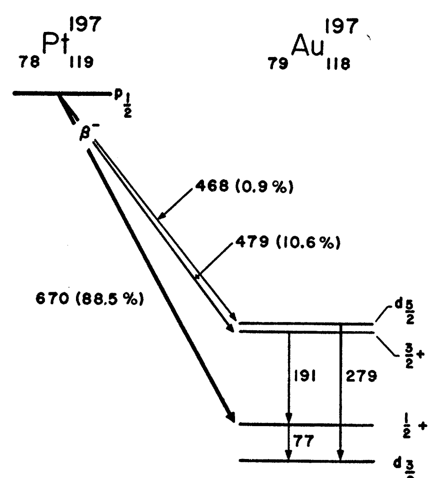

FIG. 4. Disintegration scheme of Pt^{197}.

[13] de-Shalit, Huber, and Schneider, Helv. Phys. Acta 25, 279 (1952).

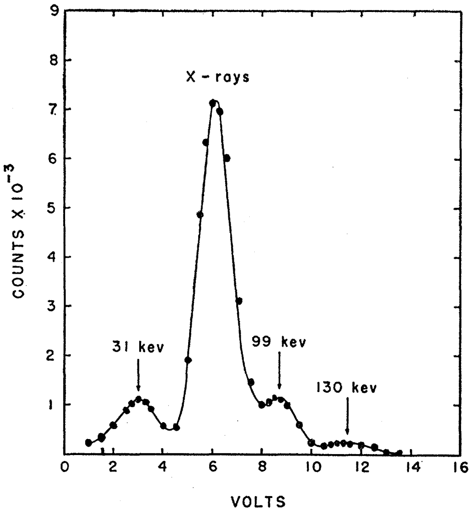

FIG. 5. Gamma rays of Pt^{195m}.

excitation of Pt^{195} formed as the decay product[13–15] of Au^{195}. The various gamma-ray energies reported have been 31.1, 99.1, and 129.9 kev in triple cascade[5]; 29, 97, and 129 kev in triple cascade and a 126-kev gamma ray as cross-over transition of the 29-kev–97-kev cascade[13]; 95-kev and 129-kev gamma rays noncoincident[14]; and 31, 99, and 129 kev, the latter transition being considered within the limits of error of the measurements, to be the cross-over transition of the two softer gamma rays in cascade.[15] Because of the energetics of the decay of Au^{195}, only one of the two 129-kev gamma rays could possibly be observed in the decay of that element.[13–15]

In the present investigations, sources of Pt^{195m} were obtained by irradiating quantities of Pt^{194} (isotopic enrichment 65%) in the Brookhaven pile. The observed gamma-ray spectrum is shown in Fig. 5 where photopeaks are in evidence corresponding to quantum energies of 31, 65, 99, and 130 kev. The 65-kev radiation is interpreted as x-radiation following internal conversion of some of the gamma rays in platinum. A large portion of the area of the 31-kev peak may be ascribed to the presence of escape radiation associated with detection of the 65-kev x-rays. It might be expected that about 10% of the x-rays give rise to pulses which would fall in the escape peak.

Some coincidence studies of the decay of Pt^{195m} are shown in Fig. 6. To obtain the curves of Fig. 6(A) one channel of the coincidence arrangement was fixed at the peak location of the 31-kev radiation while the other channel was moved over the region of higher energies. The coincidence peak at 31 kev arises from coincidences between any 31-kev gamma rays and the pulses of the escape peak of the x-radiation. Coincidences between coincident x-ray escape peaks also contribute heavily to the coincidence rate observed in this region. Precautions were taken by the use of a proper geometry and a copper absorber of thickness 0.6 g/cm² to eliminate any coincidences which might arise from detection of pulses of the x-ray escape peak in one counter and the iodine $K\alpha$ escape radiation in the other. The coincidence peak at 65 kev derives from coincidences between radiations near 31 kev and the 65-kev x-rays. In the case of these particular measurements, the coincidence peak appears to be shifted slightly to a lower energy than that at which appears the peak of single counts. This effect results from the presence of some 77-kev gamma rays of Pt^{197} which were also present in the exposed sample. Curve A of Fig. 6 also indicates the presence of coincidences between the 31-kev radiations and the 99-kev gamma ray. Beyond these coincidences lie a few arising from coincidences between 130-kev radiation and radiations in the vicinity of 31 kev. These particular coincidences are thought to be due to detection of 130-kev gamma rays (cross overs of the 31-kev–99-kev cascade) and the x-ray escape radiation of x-rays which are emitted following internal conversion of the additional 130-kev gamma ray of the 3.8-day isomeric transition. In the curve of Fig. 6(B) are presented coincidences between the 65-kev x-rays and the remainder of the gamma-ray spectrum. To obtain coincidences between the x-rays and the harder gamma rays, absorbers of carbon and copper were introduced before the counters so that the integral counting rates could be maintained at a satisfactory level, though a strong source was employed to provide a sufficient number of the hard gamma rays. From Fig. 6(B) it is clear that (x-ray)-(x-ray) coincidences are present as well as coincidences between x-rays and the harder gamma rays. Below the (x-ray)-(x-ray) coincidence peak in energy appears an additional peak of coincidences which apparently arises from detection of the full energy of the x-ray in one counter and the escape peak of the 65-kev x-ray in the other.

The gamma-ray spectrum of Fig. 5 shows the presence of a considerable amount of unconverted radiation at

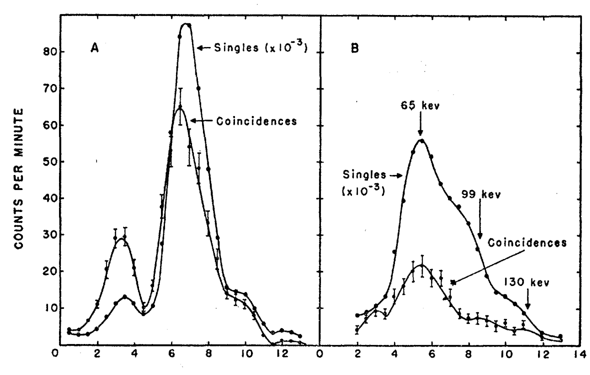

FIG. 6. Gamma-gamma coincidences in the decay of Pt^{195m}; (A): with one counter fixed at 31 kev; (B): with one counter fixed on the $K\alpha$-line of gold.

[14] Steffen, Huber, and Humbel, Helv. Phys. Acta 22, 167 (1949).
[15] Gillon, Gopalakrishnan, de-Shalit, and Mihelich, Phys. Rev. 93, 124 (1954).

130 kev. Absorption measurements were performed to show that the peak at 130 kev did not merely result from additive simultaneous detection in the crystal of the 99- and 31-kev radiations. The source strength was measured by noting the intensity of the 99-kev radiation and correcting for its total conversion coefficient which has been previously measured as having values ranging from 3.1[14] to 9.[13] Using these values of $(\alpha_T)_{99}$ to obtain estimates of the source strength, the total conversion coefficient of the 130-kev radiation was calculated to lie between values of 10 and 23 with the assumption that one 130-kev transition occurs in each disintegration. In other words, it was for the moment assumed that the 130-kev radiation detected in the curves of Fig. 5 was actually emitted in the 3.8-day transition rather than in the cross-over transition of the 31-kev–99-kev cascade. With this assumption, a coincidence experiment was performed in which coincidences were sought between the presumed 130-kev gamma rays of $\alpha_T \sim 23$ and the 99-kev gamma. The calculated coincidence rate was $\sim$300 coincidences per minute whereas the actual observed coincidence rate was 0.3 ± 0.3 coincidence per minute. In this manner it was established that the observed unconverted quantum radiations of Pt^{195m} at 130 kev are not emitted in the isomeric transition but more probably in the cross over transition of the 31-kev–99-kev cascade. These results are consistent with the characterization of the isomeric transition as $M4$ and having a correspondingly large conversion coefficient so that its quantum radiations are not detectable. The cross-over transition has been previously reported with an intensity of 10%.[13] It appears not to have been detected in more recent studies.[11,15]

The disintegration scheme of Pt^{195m} is shown in Fig. 7. The spin of the ground state of Pt^{195} has been previously measured[16] as 1/2. The shell model would indicate a ground state orbital of $p_{1/2}$ in agreement with this measurement. If the excited states of Pt^{195} are also considered to be single particle levels, the 99-, 130-, and 260-kev levels would have respectively orbitals of

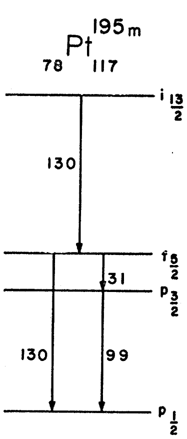

FIG. 7. Decay scheme of Pt^{195m}.

[16] B. Jaeckel and H. Kopfermann, Z. Physik 99, 492 (1936).

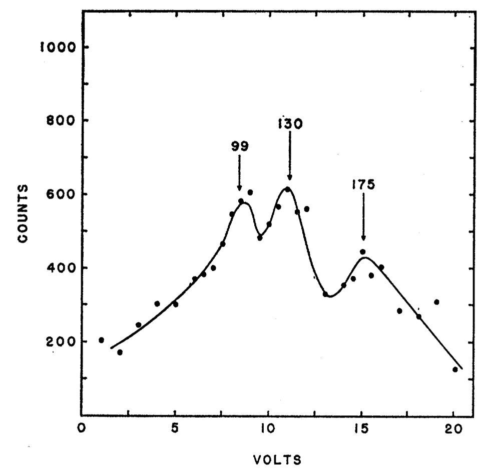

FIG. 8. Some gamma rays from neutron capture in platinum. The 130-kev peak is from Pt^{193m}. This spectrum was observed through 1.3 g/cm² of cadmium and 1.5 g/cm² of copper to reduce x-ray intensity.

$p_{3/2}$, $f_{5/2}$, and $i_{13/2}$. The 31-kev gamma ray is placed above the 99-kev transition in the decay scheme, because it has been previously observed that the 31-kev gamma ray is the less intense as noted[15] in the K-capture decay of Au^{195}. From a consideration of conversion coefficients, lifetimes, etc., the 31- and 99-kev transitions have been classified[11] as $M1$ in accord with the decay scheme. The measured conversion coefficients[13,14] of the 130-kev cross-over transition, though not completely in agreement with, are, nevertheless, not totally inconsistent with classification of the transition as $E2$. The isomeric transition of energy 130 kev and lifetime 3.7 days has been identified by its K/L ratio[17] as $M4$, again consistent with the shell model prediction that the isomeric state have an orbital assignment of $i_{13/2}$.

PLATINUM-193m

The radiations of the 4.3-day Pt^{193} have been previously examined, with varying results.[5,18,19] In the present investigation, the isotopic enrichment of Pt^{192}, the target material, had been increased to 13%. After exposure in the Brookhaven pile, the platinum fraction was separated from gold, mercury, and iridium. The gamma-ray spectrum subsequently obtained is shown in Fig. 8. Absorbers of cadmium, copper, and carbon were employed to reduce the x-ray intensity. The remaining three photopeaks were interpreted as being emitted in the decay of Pt^{195m} (99 kev), Pt^{193m} (130 kev), and Pt^{191} (175 kev). Coincidences were sought between the various possible pairs of photopeaks, but none were found, consistent with the various assignments given

[17] M. Goldhaber and A. W. Sunyar, Phys. Rev. 83, 906 (1951).
[18] G. Wilkinson, Phys. Rev. 73, 252 (1948).
[19] Mandeville, Scherb, and Keighton, Phys. Rev. 74, 601 (1948). Earlier references to measurements on Pt^{193} may be found in this paper.

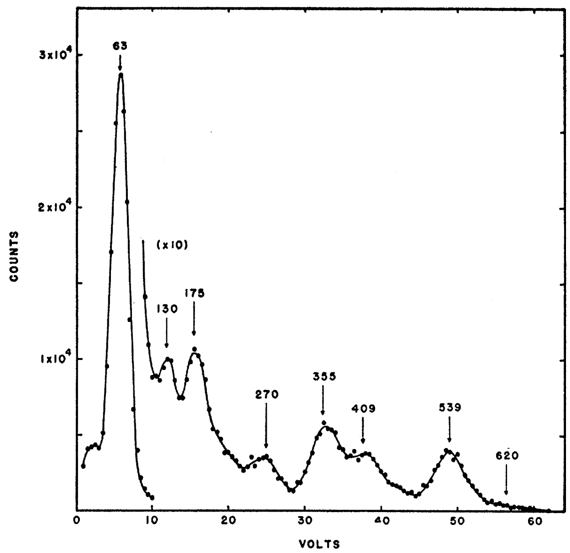

FIG. 9. Gamma-ray spectrum of Pt^{191}.

above. The 130-kev gamma ray is heavily converted and has been classified as emitted in an $M4$ transition.[5]

A search was carried out to determine whether any gamma rays of quantum energies greater than 700 kev might be present in the decay of Pt^{193m}. None were detected, and from the source strength and crystal detection efficiency it was conservatively estimated that if any gamma rays are present on the energy interval 0.7 Mev $< h\nu <$ 2 Mev, they must be emitted with a frequency of less than one in every three thousand disintegrations. Gamma rays of energy 1.6 to 1.7 Mev have been previously reported.[5,18]

PLATINUM-191

The radiations of Pt^{191} have been investigated previously[5,18,20,21] and as many as twenty-six gamma rays have been detected with the use of high resolution magnetic spectrometers. For the present study, Pt^{190} (isotopic abundance enriched to 0.76%) was irradiated by neutrons in the Brookhaven reactor. In the scintillation spectrometer of the present investigation, it was possible to resolve only seven photopeaks, shown in Fig. 9. Present also is an intense x-ray peak at ~63 kev. Its position in energy is perhaps shifted somewhat by the presence of nuclear gamma rays. Virtually all of these peaks displayed half-widths larger than would be expected from the known resolution. This observation is in agreement with the reported complexity of the gamma-ray spectrum.[21]

With the resolution available, it was, nevertheless, decided to investigate insofar as was possible the cascade relationships among the gamma rays. Such measurements also at times serve to resolve the two or more gamma rays of a complex photopeak. The coincidence curves relating to Pt^{191} are shown in Fig. 10. In Fig. 10(A) are plotted coincidences with one channel fixed at the photopeak of the 130-kev gamma ray while the other was moved over the spectrum. The curve shows this gamma ray to be in cascade with radiation at 220 and 409 kev. The 220-kev line was not evident in the gamma-ray spectrum of Fig. 9. The coincidences of Fig. 10(B) are between the 175-kev gamma and the remainder of the spectrum, showing peaks at ~175 kev and 355 kev. These measurements have demonstrated the complexity of the 175-kev line. The results of this study of Pt^{191} are in substantial agreement with a previously proposed decay scheme.[5]

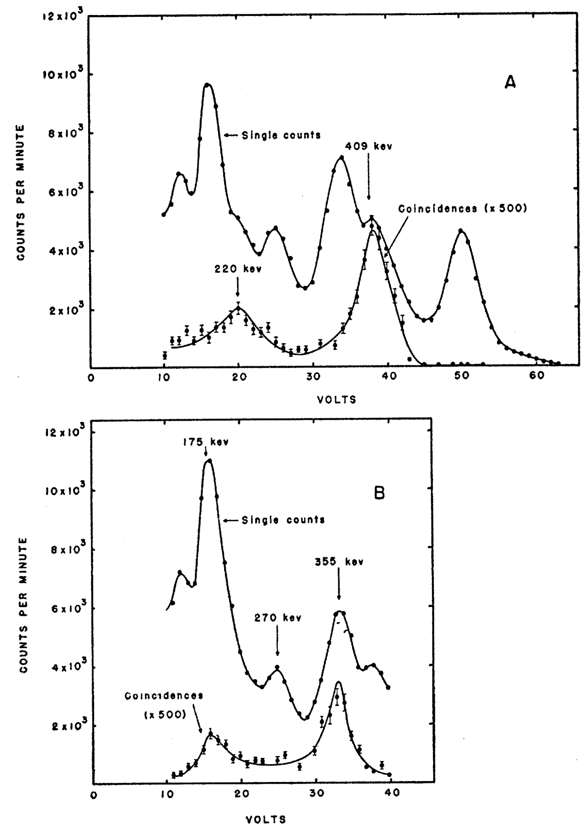

FIG. 10. Gamma-gamma coincidences in the decay of Pt^{191}; (A): with one counter fixed at 130 kev; (B): with one counter fixed at 175 kev.

[20] Swann, Portnoy, and Hill, Phys. Rev. **90**, 257 (1953); Tomlinson, Naumann, and Mihelich, Phys. Rev. **94**, 794 (1954).

[21] E. P. Tomlinson and R. A. Naumann, Phys. Rev. **100**, 955(A) (1955).