

The integral (30) requires considerably less labor. As before we make use of the generating function and write

$$\begin{aligned} & \sum \xi^r \eta^s e^{-\epsilon} L_s(\epsilon) L_r(\epsilon) d\epsilon \\ &= (1-\xi)^{-5/2} (1-\eta)^{-5/2} \\ & \quad \times \int \exp\{-[(\epsilon/1-\xi) + (\eta/1-\eta) + 1]\epsilon\} d\epsilon \\ &= (1-\xi)^{-3/2} (1-\eta)^{-3/2} (1-\xi\eta)^{-1}. \end{aligned} \quad (64)$$

By expanding this in powers of ξ and η we obtain

$$H_{rs} = Z\nu \begin{bmatrix} 1 & 3/2 & 15/8 & \cdots \\ 3/2 & 13/4 & 69/16 & \cdots \\ 15/8 & 69/16 & 433/64 & \cdots \end{bmatrix}, \quad (65)$$

where the effective nuclear charge Z is defined by

$$Z = (\sum N_i Z_i^2 / n). \quad (66)$$

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The Disintegration of Ce^{141}

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The beta- and gamma-rays from 28 day Ce^{141} have been studied in a beta-ray spectrometer. The maximum beta-particle energy is 560 kev and gamma-rays at 146 kev and 315 kev have been found. With the additional aid of beta-gamma- and gamma-gamma-coincidence studies a disintegration scheme is proposed.

The maximum beta-energy of Pr^{143} has also been determined as 920 kev.

I. INTRODUCTION

IN view of the fact that all previous measurements¹⁻⁴ of the energies of the beta- and gamma-rays of Ce^{141} have been made by absorption methods, it seemed advisable to make a more thorough study of the isotope using a magnetic spectrometer. The investigation reported herein is such a study. From it has resulted both beta- and gamma-spectra of Ce^{141} and a beta-spectrum of Pr^{143} .

As a source of additional information both beta-gamma- and gamma-gamma-coincidence studies of Ce^{141} have also been conducted.

Using the results of the investigation, a possible decay scheme has been proposed for Ce^{141} .

II. APPARATUS

The magnetic spectrometer used in these investigations is the same as used previously⁵ in an investigation of the nuclear radiations from Se^{75} . The only significant change in the instrument since the Se^{75} investigation has been the replacement of the low resistance coils used to supply the magnetic field by a set of high resistance

coils⁶ and the replacement of the battery current supply for these coils by an electronic constant current supply.⁷

Counters used with the spectrometer were of the same design and employed the same argon-ethylene filling mixture as before.⁵ The window in the current investigation was a thin zapon window with a low energy cut-off at approximately 6 kev.

The circuit used for the beta-gamma- and gamma-gamma-coincidence studies was constructed in this laboratory by Mr. W. R. Konneker. It is built such that either instantaneous or delayed coincidences may be studied. In the present situation, only instantaneous coincidences were investigated. The resolving time for instantaneous coincidences may be varied in the range from 10^{-7} to 10^{-6} second in order that the apparatus may be used in conjunction with either scintillation or G-M counters.

III. EXPERIMENTAL DETAILS AND RESULTS

In order to obtain the sample used for the current investigation, cerium oxide powder was irradiated with slow neutrons from the Oak Ridge pile. Spectroscopic analysis (as supplied by Oak Ridge) of the sample used for the bombardment indicated no impurities in the sample except for a possible small quantity of iron. Slow neutron bombardment of iron has been known⁸ to lead

¹ W. H. Burgus, Plutonium Project Report CC-680, p. 13 (May 1943) as reported by G. T. Seaborg and I. Perlman, *Rev. Mod. Phys.* **20**, 585 (1948).

² M. L. Pool and J. D. Kurbatov, *Phys. Rev.* **63**, 463 (1943).

³ M. L. Pool and N. L. Krisberg, *Phys. Rev.* **73**, 1035 (1948).

⁴ W. Bothe, *Zeits. f. Naturforschung* **1**, 179 (1946).

⁵ Ter-Pogossian, Robinson, and Cook, *Phys. Rev.* **75**, 995 (1949).

⁶ We are indebted to the Moloney Electric Company, St. Louis, and especially to Mr. Wooley of that organization for their kind cooperation in making the new high resistance coils for us.

⁷ W. C. Elmore, AEC-2208-G, 29 (1948).

⁸ G. T. Seaborg and I. Perlman, *Rev. Mod. Phys.* **20**, 585 (1948).

to only one radioactive isotope of iron, Fe^{59} , which has two high energy gamma-rays at 1.1 and 1.3 Mev. Neither of these gamma-rays have been found in the current investigation. It is thus concluded that no significant amount of iron impurity is present.

Cerium has four stable isotopes at mass numbers 136, 138, 140, and 142. The isotopes at 136 and 138 are of very low abundance and have not yet been found to produce a significant quantity of radioactive material through the (n, γ) process.⁸ The resulting radioactive isotopes from such a process would have had half-lives sufficiently different from that of Ce^{141} that it is felt that, had a significant quantity of either Ce^{137} or Ce^{139} been present, it could have been detected in the gamma-ray measurements. The isotopes at 140 and 142 both produce radioactive products by means of the (n, γ) reaction and both of the products were present in significant quantities in the present investigation. Fortunately the Ce^{141} has a half-life of 28 days while the Ce^{143} has a half-life of only 33 hours. The Ce^{143} decays to Pr^{143} which further decays to Nd^{143} by beta-emission with a half-life of 13.8 days. The Ce^{141} can, however, be separated

from the Pr^{143} both chemically and through half-life measurements.

The latter of these two methods of separation was used in the study of the gamma-radiations. The cerium oxide as received from Oak Ridge was initially placed in a small brass container built for the purpose whose walls were sufficiently thick to completely absorb all possible beta-radiation from the source. A 50 mg/cm^2 uranium foil radiator was fastened at the proper location on this container and the photoelectrons thus produced were analyzed in the magnetic spectrometer. The results are shown in Figs. 1 and 2. The open and solid circles represent two sets of data taken with approximately six days intervening. In Fig. 1 the data have been corrected for decay assuming a 28 day half-life. In Fig. 2 the correcting process assumed 13.8 days as the half-life. It takes no stretch of the imagination to see that the half-life at all parts of the spectrum is much closer to 28 days than to 13.8 days.

It seems possible to associate two groups of photoelectron lines with gamma-rays. The lines marked K_1 , L_1 , and M_1 appear at the correct momenta to be asso-

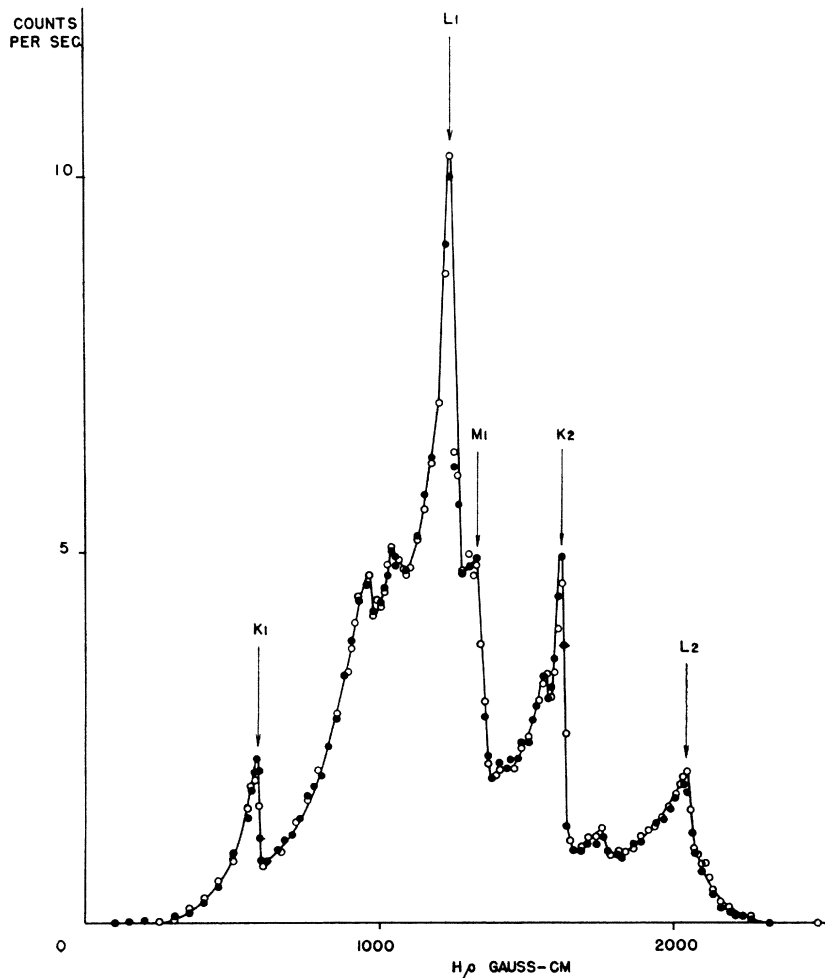
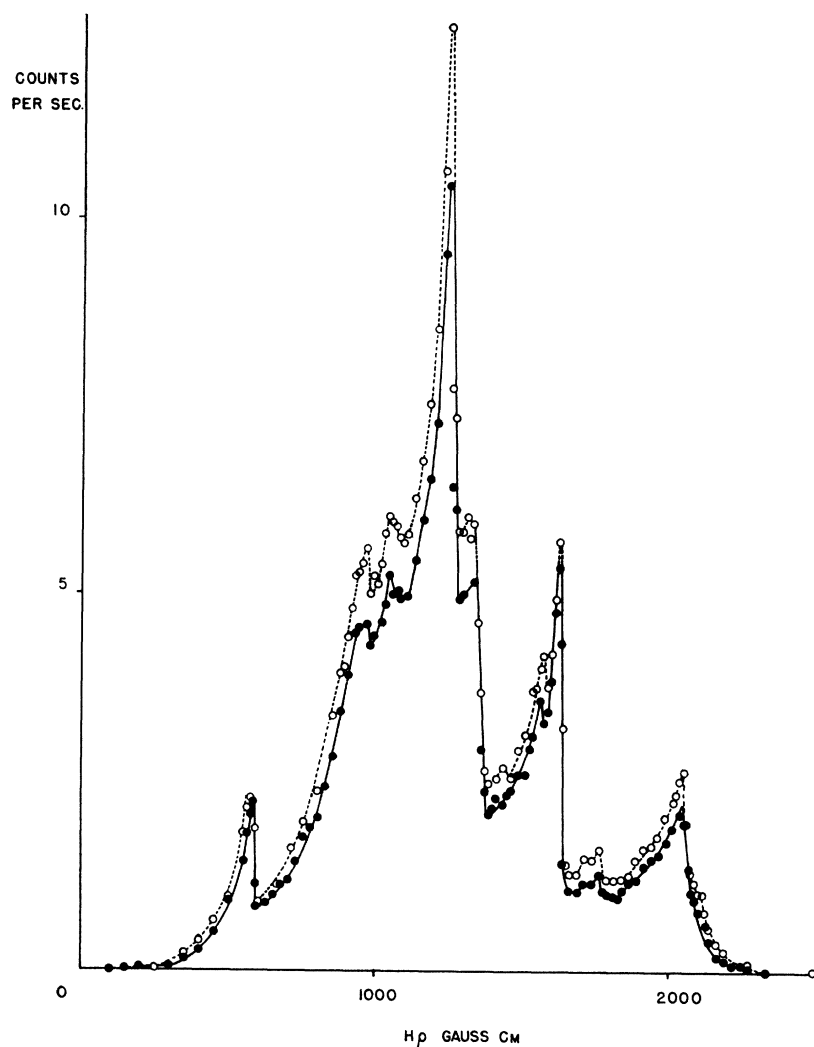


FIG. 1. Photoelectron spectrum of a source of Ce^{141} plus Pr^{143} using a 50 mg/cm^2 uranium radiator. The open and solid circles represent two sets of data taken approximately six days apart. All parts of the spectrum have been corrected for decay to an initial time by an amount appropriate to a 28 day half-life (the reported half-life of Ce^{141}). K_1 , L_1 , and M_1 point out the K , L , and M photo lines of the 146 kev gamma-ray and K_2 and L_2 the K and L photo lines of the 315 kev gamma-ray.

FIG. 2. Same as Fig. 1 except that the spectrum has been corrected for decay to an initial time by an amount appropriate to a 13.8 day half-life (the reported half-life of Pr^{143}).



ciated with a single gamma-ray of 146 kev energy while those marked K_2 and L_2 may in like manner be associated with a gamma-ray of 315 kev energy. Other lines of much lower intensity appear at momenta of 980, 1070, 1570, and 1768 gauss-cm. Since it has thus far been impossible to assign definitely a gamma-ray energy to any of these lines, they have not been considered in the disintegration scheme. They do, however, as can be seen in Fig. 1, appear to decay with a half-life of 28 days and would thus seem to be associated with Ce^{141} . It is assumed that the apparently low intensity of K_1 is caused by its close proximity to the low energy cut-off of the counter window.

Upon completion of the photoelectron investigation, a chemical separation was made⁹ after which cerium

⁹ Chemical separations were performed by Mr. A. G. Jehle, Jr. The procedure as outlined by Mr. Jehle is described in the following two paragraphs.

The impure cerium oxide was dissolved in 10 ml HNO_3 with a small drop of HF added as catalyst. When dissolved, 3 g NH_4NO_3 was added and the solution cooled to precipitate the cerium as $(NH_4)_2Ce(NO_3)_6$. The precipitate was separated and washed once

and praseodymium were studied as beta-ray sources. The beta-spectrum of the cerium fraction is shown in Fig. 3 and that of the praseodymium (plus cerium)⁹ fraction in Fig. 4. The 146 kev gamma-ray transition appears internally converted but no trace can be ob-

by decantation with a few ml of HNO_3 containing a little NH_4NO_3 . The product was reprecipitated from 5 ml HNO_3 containing a small amount of NH_4NO_3 , separated and washed as before. The precipitate was dissolved in water, diluted to 50 ml, and the cerium precipitated as the oxalate. After cooling to room temperature, this was filtered and ignited to the carbonate.

In order to concentrate the praseodymium, the first set of filtrates from the nitrate separation were combined and concentrated further to precipitate as much of the $(NH_4)_2Ce(NO_3)_6$ as possible. After cooling, the supernatant liquid was removed by decantation and evaporated almost to dryness. A little water was added and the evaporation was repeated three more times to remove as much of the free HNO_3 as possible. The final residue was taken up in about 200 ml of water, boiled vigorously for an hour, and allowed to stand overnight to precipitate the basic ceric nitrate. The suspension and precipitate were separated by centrifuging. The praseodymium and the cerium remaining in solution was precipitated as the oxalate by adding a concentrated solution of oxalic acid to the hot filtrate. After standing overnight, the precipitate was filtered off and ignited to the carbonates.

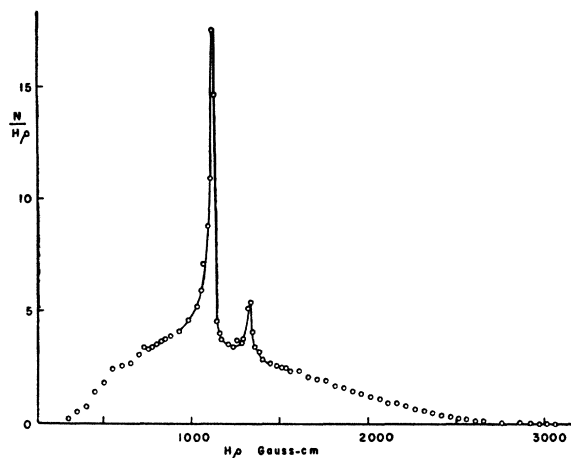


FIG. 3. Beta-spectrum of the chemically separated cerium fraction (Ce^{141}). The two internal conversion peaks correspond to the K and L peaks of the 146 kev transition in Pr^{141} . The end-point energy is 560 kev.

served of any other internally converted transition

Both beta-ray sources were mounted on a cellulose tape backing. They were made by sprinkling the source material onto the gummed side of the tape and shaking off all material which did not stick. Weighing indicated

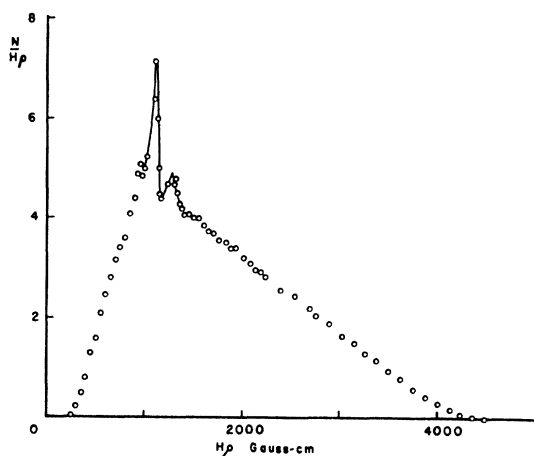


FIG. 4. Beta-spectrum of the separated praseodymium (plus cerium) fraction (see reference 9). The two internal conversion peaks are the same as those appearing in Fig. 3. The end-point energy of Pr^{143} is 920 kev.

average thicknesses of 0.8 mg/cm^2 for the Ce^{141} source and 0.2 mg/cm^2 for the Pr^{143} (plus Ce^{141})⁹ source.

End-point energies for the beta-spectra have been assigned at $560 \pm 20 \text{ kev}$ for the Ce^{141} and $920 \pm 10 \text{ kev}$ for Pr^{143} .

FK (Fermi-Kurie)¹⁰ plots were made of the spectra

¹⁰ The name FK (Fermi-Kurie) for this type of plot has been suggested by Professor E. Feenberg as a means of standardization. The name Kurie plot arose from the fact that the first use of this type of analysis of experimental data was made by Kurie, Richardson, and Paxton [Phys. Rev. 48, 167 (1935)]. Later to distinguish between the respective theories the terms Fermi plot and K-U

of Ce^{141} and Pr^{143} in the regions near their end-point energies. These are shown in Figs. 5 and 6. Interpretation of the shape of the FK plot of Ce^{141} will be discussed in connection with the tentative assignment of a disintegration scheme for that isotope.

Although coincidence measurements usually do not lead by themselves to accurate interpretations of disintegration schemes, their use as a supplementary tool in conjunction with beta-spectrometer measurement is a common practice.¹¹ In order to obtain further evidence for a possible disintegration scheme of Ce^{141} both beta-

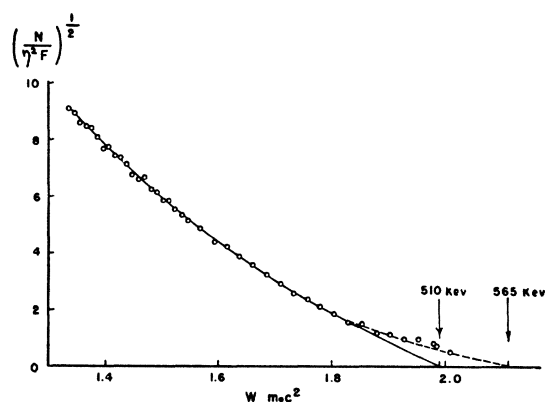


FIG. 5. Fermi-Kurie plot of the Ce^{141} beta-spectrum in the energy interval between the internal conversion lines and the spectral end point. The indicated value of 515 kev is the end point determined by a continuation of the smooth curve from the lower energy region of the plot. The end-point energy of 565 kev is the value obtained if the observed break in the curve at $W \approx 1.8 \text{ mc}^2$ is assumed correct, and, as discussed in the text, is the case here considered correct.

gamma- and gamma-gamma-coincidence measurements were made.

All attempts failed to reveal any gamma-gamma-coincidences. It would, therefore, appear that, if the

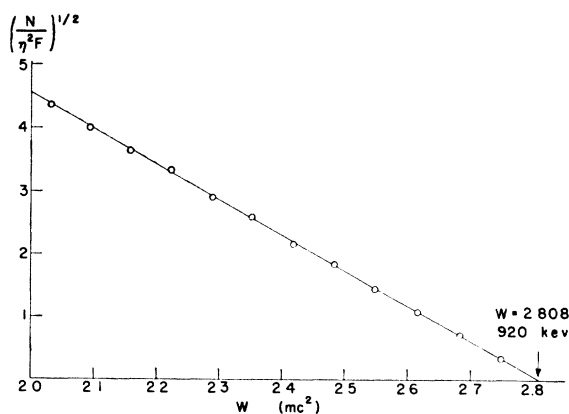


FIG. 6. Fermi-Kurie plot of the Pr^{143} beta-spectrum for energies greater than the end-point energy of the Ce^{141} spectrum.

plot came into usage. Both Kurie plot and Fermi plot have been carried over into current terminology. It is possible to eliminate this dualism through introduction of the FK plot.

¹¹ A. C. G. Mitchell, Rev. Mod. Phys. 20, 296 (1948).

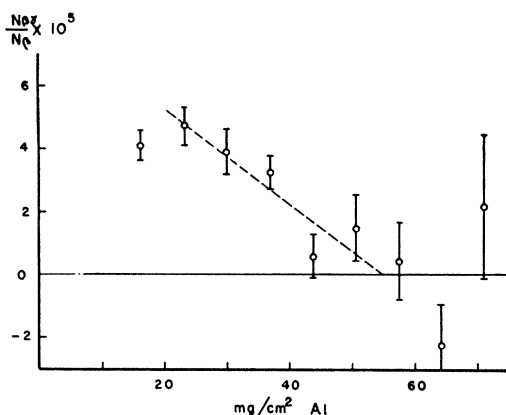


FIG. 7. Beta-gamma coincidences in Ce^{141} as a function of beta-ray energy in terms of absorber thickness.

two gamma-rays mentioned above are in coincidence, one is delayed. The results of the beta-gamma-coincidence measurements are indicated in Fig. 7. It would appear from this that no beta-particle having a kinetic energy greater than 250 keV is followed immediately by a gamma-ray whereas some of those having energy below 250 keV are in immediate coincidence with a gamma-ray.

IV. DISINTEGRATION SCHEME

The experimental evidence offered does not conclusively prove the validity of the disintegration scheme proposed in Fig. 8. The proposed scheme is, however, consistent with all experimental evidence and is the only one which to date we have been able to construct which is consistent with all such evidence.¹² The 560 keV beta-ray direct to the ground state and the 240 keV beta followed by a 315 keV gamma-ray is, in addition to spectrometer measurements, a direct result of the beta-gamma-coincidence results. The 415 keV beta-ray followed by a 145 keV gamma-transition agrees with the evidence presented only if the gamma-transition is delayed. The negative results^{13,14} for this isotope in the region 10^{-6} to 10^{-3} seconds indicates that the half-life of such a metastable state must be greater than 10^{-3} seconds. Because of the difficulties of the chemical separation of praseodymium from cerium⁹ further evidence for a metastable state in Pr^{141} will not be immediately forthcoming.

Experimental evidence favoring the disintegration scheme seems also to be indicated by the FK plot (Fig. 5). The experimental points in the energy region

¹² Alternate disintegration schemes require either that some of the experimental data be in error or that the calculations of internal conversion coefficients by Rose, Goertzel, Spinrad, Harr, and Strong [Phys. Rev. **76**, 184 (1949)] be in serious error for some gamma-ray transitions and not for others.

¹³ S. DeBenedetti and F. K. McGowan, Phys. Rev. **74**, 728 (1948).

¹⁴ Bunyan, Lundby, and Walker, Proc. Phys. Soc. London **62**, 253 (1949).

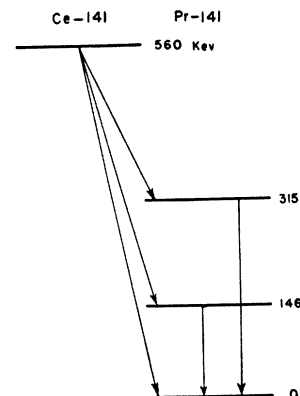


FIG. 8. Proposed disintegration scheme for the 28 day Ce^{141} . Numbers indicate the height of the energy level in keV above the ground state of the daughter Pr^{141} .

below $W = 1.8 mc^2$ may be extrapolated as indicated by the solid line to a maximum beta-particle energy of $W = 2.0 mc^2$ (510 keV). The experimental points however deviate from this at an energy slightly greater than $W = 1.8 mc^2$ and may be extrapolated as indicated by the dotted line to a maximum beta-particle energy at approximately $W = 2.1 mc^2$ (565 keV). The difference between this higher maximum energy and the energy at the apparent break in the curve near $W = 1.8 mc^2$ is, within experimental error, equal to 145 keV and therefore offers additional evidence, even though slight, in favor of the disintegration scheme of Fig. 8.

Attempts to put quantum numbers into the tentatively proposed disintegration scheme have thus far not led to any remarkable degree of success. A measure of either the half-life of the 145 keV transition or of the spin of Ce^{141} could add significantly to the knowledge of the decay of this isotope. It must thus be concluded that the proposed scheme, although both simple and in agreement with existing evidence, cannot be finally proven without further experimental data.

We are grateful to Professor E. Feenberg for many helpful discussions.

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Note added in proof: Since submission of the manuscript for this paper to the *Physical Review*, an article by C. E. Mandeville and E. Shapiro [Phys. Rev. **75**, 1834 (1949)] has appeared and an article by L. R. Shepherd [Research **1**, 671 (1948)] has come to the attention of the authors. Although the results obtained by Mandeville and Shapiro differ in many respects from those of the present work, Shepherd's measurements are in remarkably good agreement with those reported by us. For the Pr^{143} beta-spectrum end point Shepherd reports 930 ± 20 keV; our results giving 920 ± 10 keV. Shepherd's disintegration scheme for Ce^{141} gives beta-groups with maximum energies of 560 and 420 keV in excellent agreement with our two higher energy beta-transitions, the 420 keV transition subsequently going to the ground state of Pr^{141} by means of Shepherd's 141 keV (our 146 keV) gamma-transition. Since Shepherd measured only the beta-spectrum he did not observe the 315 keV gamma-transition which does not appear internally converted. Our current data neither confirm nor deny the suggestions of Mandeville and Shapiro that the 315 keV gamma-ray is an impurity with exactly the same half-life as Ce^{141} .