

## Evidence for Small Deviations from the Allowed Shape in the Comparison of the Beta Spectra of $\text{Na}^{24}$ and $\text{P}^{32}$ †

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Two low- $Z$ , similar-in-energy, presumably allowed beta transitions,  $\text{Na}^{24}$  ( $E_0=1394$  kev,  $4+ \rightarrow 4+$ ,  $\log f_0 t = 6.1$ ) and  $\text{P}^{32}$  ( $E_0=1711$  kev,  $1+ \rightarrow 0+$ ,  $\log f_0 t = 7.9$ ) have been compared by means of the Argonne double-lens spectrometer at 2% resolution. Vacuum-distilled sources on 200  $\mu\text{g}/\text{cm}^2$  Al had thicknesses of 50 to 75  $\mu\text{g}/\text{cm}^2$  for  $\text{Na}^{24}$  and  $\ll 50$   $\mu\text{g}/\text{cm}^2$  for  $\text{P}^{32}$ . Considerable attention has been given the question of detector efficiency and instrumental scattering. The results show that  $\text{Na}^{24}$  has very nearly the allowed shape; its experimental shape factor,  $(N/[f(E_0-E)^2])$ , is within

0.5% that given by  $L_0$  between 110 and 1300 kev. In contrast, the experimental shape factor for  $\text{P}^{32}$  decreases about 3% from 270 to 1630 kev with a slope  $\sim 4$  times that of  $L_0$ . We conclude that there is definite evidence for small deviations from the allowed shape in one of these transitions, and further, with a lower confidence level, that the slower  $\text{P}^{32}$  transition shows the deviation. While the deviation in  $\text{P}^{32}$  is not inconsistent with previous limits set on  $T$ - $A$  interference, the large  $f_0 t$  of  $\text{P}^{32}$  suggests that second-order effects may hold the explanation.

### I. INTRODUCTION AND SUMMARY

AT the present time, there are no "standard spectra" with which the beta spectroscopist can evaluate his instrument when he is interested in precision the order of 1%. Yet, it seems clear that experimental work and theory can be profitably compared to this precision.

One such problem is the extent of small deviations from the allowed shape. We assume that even after careful scrutiny of the instrument, some small residual instrumental distortion may remain undetected. Yet if one finds differences in two presumably allowed spectra of similar atomic number and end-point energy under the same experimental conditions, it should constitute convincing evidence that such deviations exist. The present work is such a comparison between  $\text{P}^{32}$  and  $\text{Na}^{24}$ .

The most recent work on  $\text{P}^{32}$  by Pohm, Waddell, and Jensen<sup>1</sup> using volatilized sources indicated that above 250 kev (the end point of often present  $\text{P}^{33}$  contaminant) the spectrum is very close to the allowed distribution. In their intermediate-image spectrometer they found the experimental shape factor<sup>2</sup> decreased by approximately 1.5%, and in their thin-lens spectrometer it increased by approximately 1.5% from 260 to 1530 kev.

The  $\text{Na}^{24}$  beta spectrum has not been studied in as much detail as  $\text{P}^{32}$  has. In perhaps the outstanding earlier work, Siegbahn,<sup>3</sup> working with a source of average surface density  $\sim 150$   $\mu\text{g}/\text{cm}^2$  but deposited from liquid, found the Kurie plot to be straight (within a few percent) for  $E > 200$  kev.

We have re-examined these spectra and find that there is a difference in their shapes. In our instrument the  $\text{Na}^{24}$  beta transition has the allowed shape (i.e., its shape factor is, within 0.5%, the same as  $L_0$ ); in con-

trast, the  $\text{P}^{32}$  shape factor decreases gradually about 3% from 270 to 1630 kev with a slope  $\sim 4$  times that of  $L_0$ .

### II. EXPERIMENTAL

#### A. Spectrometer

For these measurements, the Argonne double-lens<sup>4</sup> spectrometer was operated at a resolution of 2% and transmission of 1% for a source  $\frac{1}{8}$  in. in diameter. We have investigated the effects of baffle-edge design and have shown that edge penetration at high energies can affect the shape of the spectrum. Specifically, it has been shown that a sharp-edged or sharp-cornered baffle immediately in front of the detector may enhance the high-energy portion of these  $\sim 1.5$  Mev spectra by several percent, the effect depending on the perimeter-to-area ratio as expected. For baffles which must touch the envelope of trajectories and into which the trajectories come with a range of angles, (i.e., near the detector) we believe a rounded edge best makes the compromise. The radius of curvature of the edge should be as large as possible consistent with keeping the extreme angles tangent to the edge.

We have reinvestigated the local static and dynamic external magnetic fields, an important consideration for the iron-free instrument. The degaussing is accomplished to the degree that 500-volt electrons (special 1-meter-long oscilloscope tube) suffer  $0 \pm 1$  mm deflection in traversing the axis of the spectrometer. This corresponds to an average field of  $< 1.4$  milligauss over the trajectory and to a transmission constant<sup>5</sup> to  $< 0.2\%$  between 0.1 and 1.7 Mev. This same method also shows the negligible effect of the ac fields (60 cps and harmonics) from power lines in the building.

We have investigated the background at zero current

† Based on work performed under the auspices of the U. S. Atomic Energy Commission.

<sup>1</sup> Pohm, Waddell, and Jensen, *Phys. Rev.* **101**, 1315 (1956). This article has extensive references to earlier work on  $\text{P}^{32}$ .

<sup>2</sup> Experimental shape factor means the plot of  $N(p)/[f(E_0-E)^2]$  vs  $E$ , where  $N(p)$  is the experimental momentum distribution,  $f$  is the Fermi function, and  $E$  is the electron energy.

<sup>3</sup> K. Siegbahn, *Phys. Rev.* **70**, 127 (1946).

<sup>4</sup> See Porter, Freedman, Novey, and Wagner, *Phys. Rev.* **103**, 921 (1956) and Argonne National Laboratory Report ANL-5525 (unpublished) for additional details of the instrument.

<sup>5</sup> These calculations are based on the fact that the image at the ring focus is considerably broader than the ring-focus aperture—i.e.,  $\frac{1}{8}$ -in. diameter source and ring-focus aperture with 1.5-mm radial opening.

and above the spectrum with sources  $\approx 4 \times 10^8$  disintegrations/min and find no increase in the zero-current background and find counting rates above background beyond the spectrum of  $2 \times 10^{-4}$  of the peak rate—a level low enough not to affect the results of these experiments.

### B. Detector

For these measurements a flow-type, loop-anode, end-window, propane-gas proportional counter operated at a pressure of 30 cm Hg was used. Voltage plateaus 200 volts long with slope  $< 1\%$  per 100 volts are obtained with various energies in the range of interest focussed on the counter. In addition, the ratio of the counting rates at the highest and lowest energy to be measured is independent of the counter voltage over at least 100 volts at a pressure of 30 cm Hg. This good an overlap of the plateaus is not found for example at a pressure of 8 cm Hg. Extensive comparisons between this gas counter and an anthracene scintillation detector<sup>6</sup> show identical spectral shapes ( $\pm 0.5\%$ ) and lend support to the statement that the efficiency of the detector is constant over the energy range 0.1–1.7 Mev. The window of the gas counter is  $\approx 900 \mu\text{g}/\text{cm}^2$  Mylar with  $25 \mu\text{g}/\text{cm}^2$  Au volatilized on the inside; it is unsupported over the 1-cm-diameter aperture and has a cutoff of 18 kev.

### C. Sources

The  $\text{Na}^{24}$  is made by  $(n, \gamma)$  reaction on  $\text{Na}_2\text{CO}_3$  in the Argonne research reactor. The samples have the characteristic 15 hr half-life out to 10 half-lives (not followed longer). The  $\text{P}^{32}$  is obtained from Oak Ridge and has a few percent  $\text{P}^{33}$  contamination whose abundance depends on the age of the material. For the  $\text{P}^{32}$  spectrum exhibited here the  $\text{P}^{33}(E_0 \approx 250 \text{ kev})$  level was 3% (beta-spectral measurements); only points above the  $\text{P}^{33}$  end point are used in the analysis.

Both  $\text{NaNO}_3$  and  $\text{H}_3\text{PO}_4$  volatilized very easily (at  $< 900^\circ\text{C}$ ) from platinum filaments onto  $200\text{-}\mu\text{g}/\text{cm}^2$  Al source backings. The thickness of the Na samples is obtained from the known weight of material on the filament and previously calibrated volatilization efficiency. They are  $50\text{--}75 \mu\text{g}/\text{cm}^2$  and visible. Upper limits on the thickness of the  $\text{P}^{32}$  samples can be computed from the Oak Ridge estimate of total solids in the stock solutions. This estimate,  $< 50 \mu\text{g}/\text{cm}^2$ , is not realistic because the  $\text{P}^{32}$  fractionates away from much of the solid material in the low-temperature volatilization. The spectrometer samples are usually invisible or barely visible and probably  $\lesssim 10 \mu\text{g}/\text{cm}^2$ . In view of the results it is important to state that contaminant  $\beta$  activity can be ruled out as the source of small deviation of the  $\text{P}^{32}$  spectrum.<sup>7</sup>

<sup>6</sup> For description, operation, and efficiency correction, see reference 4.

<sup>7</sup> Such an impurity would have to have very low  $\gamma$  intensity, a beta spectrum with  $E_0$  in the range 1 to 1.4 Mev, a half-life not too different from 14 days, and be volatile at relatively low tempera-

### D. Procedures and Treatment of Data

Every other point on the spectrum is counted with steps of increasing current and the alternate points counted with decreasing current. Such procedures have failed to show up any hysteresis effect. The spectrum is surveyed in this way a number of times; this constitutes a "run." Interspersed throughout the run are background counts above the spectrum and at zero current (no difference); an average of these is used (approximately 8 counts/min).

Half-life corrections are small for  $\text{P}^{32}$ , never exceeding 2% for a given run. For  $\text{Na}^{24}$ , half-life corrections are  $\leq 15\%$ ; we use a half-life for  $\text{Na}^{24}$  of 14.93 hr.<sup>8</sup> The procedure for taking the spectrum insures that all points are counted both early and late in the run, thus minimizing uncertainties introduced by half-life corrections.

Maximum counting rates are less than 4000/min; no resolving-time loss corrections are necessary (dead time of counter and electronics  $\approx 7 \mu\text{sec}$ ).

Spectrometer resolution corrections are negligible over most of the spectrum; only within 95% of the end-point energy do they exceed 0.1% and are they made.<sup>9</sup>

Kurie plots are constructed<sup>10</sup> and the points with  $E > \frac{2}{3}E_0$  are used to determine  $E_0$  for that run.<sup>11</sup> Calibration of the instrument is accomplished with conversion lines of  $\text{Au}^{198}$  or  $\text{Cs}^{137}$  (similarly volatilized  $\frac{1}{8}$  in. diam sources). With the  $E_0$  value the experimental shape factor can be constructed. It should be emphasized that it is not so important that the absolute value<sup>12</sup> of  $E_0$  be obtained as that the value obtained in a particular run be used in computing the experimental shape factor for that run. The calibration constant for a spectrometer may be changed a percent without influencing appreciably the experimental shape factor if  $E_0$  is derived from the data in each case.

tures. The  $\text{P}^{32}$  is made by  $(n, p)$  on sulfur, which also limits the possibilities. We searched for contaminant gamma radiation in  $\text{P}^{32}$  with a  $2\frac{1}{2}$  in.  $\times$   $2\frac{1}{2}$  in. NaI crystal in the higher energy region and with a  $\frac{1}{2}$  in.  $\times$  1 in. diam NaI crystal in the low-energy region and can set a limit  $< 10^{-5}$  for the  $\gamma/\beta$  ratio from 0.03 to 2 Mev. Further, the  $\text{P}^{32}$  samples have included Oak Ridge shipments over a period of time since 1954 so that a good sampling of batches has failed to indicate any identifiable contaminants except  $\text{P}^{33}$ .

<sup>8</sup> J. Tobailem, *J. phys. radium* **16**, 48 (1955),  $14.90 \pm 0.05$  hr; E. E. Lockett and R. H. Thomas, *Nucleonics* **11**, No. 3, 14 (1953),  $14.97 \pm 0.02$  hr.

<sup>9</sup> G. E. Owen and H. Primakoff, *Phys. Rev.* **74**, 1406 (1948); *Rev. Sci. Instr.* **21**, 447 (1950). George W. Hinman, Carnegie Institute of Technology Report NYO-91, 1951 (unpublished). See also reference 4.

<sup>10</sup> Fermi function  $f$  and screening corrections ( $< 0.25\%$  for these low- $Z$   $\beta^-$  emitters) are taken from National Bureau of Standards Applied Mathematics Series 13, 1952 (unpublished). Energy values from J. L. Wolfson and H. S. Gellman, Chalk River Report PD-255 (unpublished).

<sup>11</sup> Estimates of the standard deviation of  $E_0$  obtained from a weighted least-squares fitting of a straight line to the data for individual runs yield values  $< 1$  kev for each run.

<sup>12</sup> Incidentally, we find for  $\text{P}^{32}$ , average of 11 runs:  $E_0 = 1711 \pm 2$  kev; for  $\text{Na}^{24}$ , average of 3 runs:  $E_0 = 1394 \pm 4$  kev.

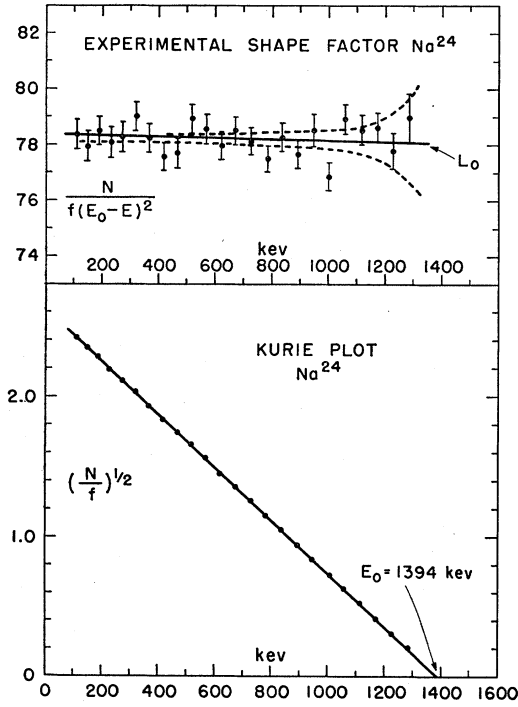


FIG. 1. Experimental shape factor and Kurie plot for Na<sup>24</sup>. The error flags are due to counting statistics only. The solid line is the function  $L_0$  (see text) arbitrarily normalized. The dotted lines follow the average of the points if  $E_0$  is changed by  $\pm 1$  keV.

### III. RESULTS AND DISCUSSION

Figure 1 shows the Kurie plot and experimental shape factor for Na<sup>24</sup> and Fig. 2 the same for P<sup>32</sup>. These data represent the weighted (by total count) averages of two runs each on the two spectra, in the sequence P<sup>32</sup>, Na<sup>24</sup>, Na<sup>24</sup>, P<sup>32</sup>. Error flags include counting statistics only. Individual runs show the same trends as the averages. The Na<sup>24</sup> shape is clearly, within the errors, the allowed one, while the P<sup>32</sup> shape factor is not constant by several probable errors. It should be emphasized here again that the P<sup>32</sup> sources are certainly thinner than the Na<sup>24</sup> sources, yet the P<sup>32</sup> deviates in the direction characteristic of thick sources.

If for the moment we assume scalar and tensor interactions only, the allowed-shape correction factor can be written<sup>13</sup>

$$C_0 = [G_s^2 |\int \beta|^2 + G_T^2 |\int \beta \sigma|^2] L_0. \quad (1)$$

The combination of electron radial-wave functions,  $L_0$ , is available in tables.<sup>14</sup> It is a very gentle function of energy at low  $Z$  and is plotted (arbitrary normalization) as the solid line for comparison with the experimental points.

<sup>13</sup> E. Greuling, Phys. Rev. **61**, 568 (1942); D. L. Pursey, Phil. Mag. **42**, 1193 (1951); we use the matrix element notation of E. J. Konopinski, Revs. Modern Phys. **15**, 209 (1943).

<sup>14</sup> Rose, Perry, and Dismuke, Oak Ridge National Laboratory Report ORNL-1459 (unpublished).

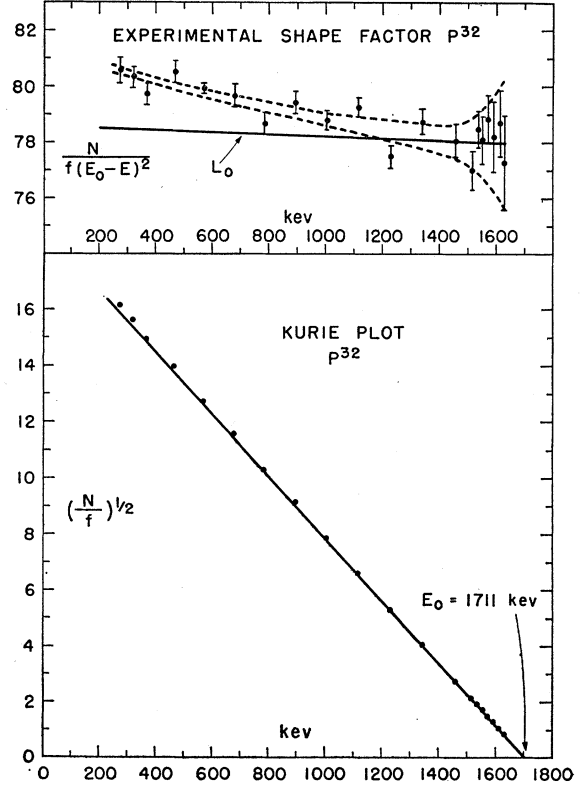


FIG. 2. Experimental shape factor and Kurie plot for P<sup>32</sup>. The error flags are due to counting statistics only. The solid line is the function  $L_0$  (see text) arbitrarily normalized. The dotted lines follow the local average of the points if  $E_0$  is changed by  $\pm 1$  keV.

If pseudoscalar-type interaction is neglected, the more general allowed-shape factor including Fierz<sup>15</sup> interference can be written<sup>13</sup> for  $\beta^-$  emission (assuming that  $k$  and  $l$ , defined below, are real):

$$C_0 = (G_s^2 + k^2 G_V^2) [1 + 2\phi_F (P_0/L_0)] L_0 \left| \int \beta \right|^2 + (G_T^2 + l^2 G_A^2) [1 + 2\phi_{GT} (P_0/L_0)] L_0 \left| \int \beta \sigma \right|^2,$$

where  $P_0$  is another combination<sup>14</sup> like  $L_0$ ,

$$k = - \int 1 / \int \beta, \quad l = - \int \sigma / \int \beta \sigma, \quad (2)$$

$$\phi_F = \frac{k G_S G_V}{G_s^2 + k^2 G_T^2}, \quad \phi_{GT} = \frac{l G_T G_A}{G_T^2 + l^2 G_A^2}.$$

Note that  $P_0/L_0 \rightarrow 1/W$  in the approximation  $\alpha Z \ll 1$ , leading to the  $\sim 1/W$  energy dependence for the allowed shape with Fierz interference. The spin of P<sup>32</sup> has not been measured, but using the shell-model predictions the transition is  $1+ \rightarrow 0+ (|\int \beta|^2 = 0)$ . The present results from P<sup>32</sup> then yield (taking  $l=1$ )  $0.025 \leq G_A/G_T$

<sup>15</sup> M. Fierz, Z. Physik **104**, 553 (1937).

(or  $G_T/G_A \leq 0.045$ ). This spread would overlap the spread of Pohm *et al.*<sup>1</sup> for their intermediate-image spectrometer but not for their thin-lens results on  $P^{32}$ . It also falls just within the upper limit of the spread from the  $He^6$  experiments of Schwarzschild *et al.*,<sup>16</sup> who conclude that  $-0.15 < G_A/G_T < 0.04$ ; in contrast our result does not include zero for  $G_A/G_T$ .

Even though the presence of Fermi terms might dilute (or cancel) the effects of  $T$ - $A$  interference, one might still expect to see some smaller deviation in the spectrum of  $Na^{24}(4+ \rightarrow 4+)$ . The limits for the deviation from the allowed shape in this case are best stated by specifying  $r$ , where  $C_0 = [1 + (r/W)]L_0$ . For  $Na^{24}$  we find  $-0.026 < r < 0.020$ , a result consistent with no Fierz interference. (For comparison,  $P^{32}$  yields  $0.05 < r < 0.093$ .) It must be remembered of course that the results viewed as absolute measurements of either shape individually must be regarded at a lower confidence level than the result that there is a difference in the shapes of the two spectra.

In view of the  $He^6$  spectrum and the  $Na^{24}$  spectrum, we set aside the analysis in terms of  $T$ - $A$  interference and look for other explanations. One such possibility is that through some peculiarity of the nuclear wave functions the  $|\int \beta \sigma|^2$  is suppressed to the extent that the usually neglected higher order matrix elements<sup>17</sup> make their energy dependence felt. The high  $\log f_0 t = 7.9$  lends support to this notion, but no quantitative estimate of this possibility has been made for  $P^{32}$ .<sup>18</sup>

<sup>16</sup> Schwarzschild, Rustad, and Wu, Bull. Am. Phys. Soc. Ser. II, **1**, 336 (1956).

<sup>17</sup> E. J. Konopinski in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers Inc., New York, 1955), p. 296; P. F. Zweifel, Phys. Rev. **95**, 112 (1954).

<sup>18</sup> B. C. Carlson in Iowa State College Report ISC-758 (unpublished), reports some preliminary estimates for the case of  $C^{14}$

There is the possibility that the parity<sup>19</sup> of the ground state of  $P^{32}$  is odd rather than even in which case the transition would be classed as first-forbidden non-unique; much larger deviations from the allowed shape have been reported for a few of these transitions.<sup>4</sup> This would be the first such failure of the shell-model predictions of parity in this region of the shell structure, and seems an unlikely possibility.

Finally it should be mentioned that finite-nuclear-size corrections to  $L_0$  are very small<sup>20</sup> at this  $Z$ , and, of course, have very nearly the same consequence for both spectra.

In summary, these results show first that there are differences in these allowed spectra; second, and less strongly, that the slower  $P^{32}$  transition is the one showing the deviations from the allowed shape. Further we prefer at present to suppose that the explanation lies with the suppression of the usually dominant matrix elements rather than with  $T$ - $A$  interference.

#### ACKNOWLEDGMENTS

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in which the interference between  $\int \beta \sigma$  and the retarded matrix elements, particularly  $\int \beta \alpha \times r$ , makes remarkably large contributions compared to  $|\int \beta \sigma|^2$  and introduces weak energy dependence in the shape factor of the order of 1-2%.

<sup>19</sup> It is the parity rather than the spin predictions which are subject to question here since (assuming  $0+$  for even-even  $S^{32}$ ) even parity and a spin  $> 1$  for  $P^{32}$  would make the transition second forbidden; both shape and  $f_0 t$  value rule this out.

<sup>20</sup> M. E. Rose and D. K. Holmes, Phys. Rev. **83**, 190 (1951).