Lifetime of the 1314-keV Level in ^{144}Nd†

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The lifetime of the 1314-keV 4+ level in ^{144}Nd was measured by delayed coincidence using a self-comparison method. The decay of ^{144}Pm to ^{144}Nd proceeds mainly via a 476-keV (4+)–618-keV (2+)–696-keV (0+) γ-ray triple cascade. By means of suitable triple-coincidence requirements, the time spectra of delayed coincidences through the 4+ and the 2+ states were obtained simultaneously but stored separately. These spectra were analyzed for their relative centroid shift, which when combined with the known lifetime of the 696-keV level, yielded a half-life for the 1314-keV level of 20.5± 3.7 psec.

INTRODUCTION

In 1966, Johansson *et al.*[1] reported a value for the g factor of the 1314-keV (4+) level in ^{144}Nd which they determined by a perturbed-angular-correlation measurement. These authors noted that their results of $g = 0.05 \pm 0.05$ was not of the magnitude expected for a level characterized as a 4+ member of a vibrational band. They further pointed out that while the $B(E2)$ associated with the first excited 2+ level is enhanced by about a factor of 20, the lifetime measurement of Ofer[2] shows the $4+ \rightarrow 2+$ transition probability to be close to single-particle estimate. It would therefore appear that both the lifetime and the g-factor measurements disagree with a collective characterization of the 4+ state. The observation can be made, however, that these are not two independent pieces of evidence. In determining a g factor from perturbed angular correlations, the lifetime of the state enters as a multiplicative factor. If the lifetime is incorrect, the g factor must necessarily also be incorrect. The mean life here in question of $\tau_m = (13 \pm 4) \times 10^{-11}$ sec was measured in 1959, when such subnanosecond determinations were still tasks of formidable difficulty. Since this quantity is crucial to the correct interpretation of the structure of ^{144}Nd, we undertook to remeasure the lifetime.

EXPERIMENTAL PROCEDURE

The 1314-keV state whose lifetime is to be determined is populated in the decay of ^{144}Pm. Figure 1 shows a partial decay scheme[3] containing the levels and transitions relevant to the present experiment. The ^{144}Pm source was produced at the Yale heavy-ion accelerator by the reaction ^{141}Pr$(\alpha, n)^{144}$Pm. Examination of the radioactive product using a Ge(Li) detector showed that very little ^{143}Pm was present. It will also become evident from the description below that because of its decay characteristics, ^{143}Pm cannot possibly disturb the present experiment.

In the measurement of short lifetime by delayed coincidence, the "centroid-shift" method of data analysis is in principle capable of greater sensitivity than the "slope" method of observing the exponential decay. In practice, however, the determination of centroid shifts can be subject to many complications. Since both the method and the problems have been described in numerous review articles,[4] a thorough discussion of techniques will not be undertaken in this paper. We will present only those details which are relevant to our measurement of the lifetime of the 1314-keV state by centroid-shift analysis.

The method here employed is essentially that described by Simms, Benczer-Koller, and Wu.[5] According to these authors, two conditions must be met in order for their technique to be applicable: (1) A source must exhibit a delayed-coincidence event to be measured and a prompt-coincidence event to be used for comparison; (2) the two events must not be distinguishable in a time-to-amplitude converter (TAC); yet must be distinguishable by an auxiliary coincidence system. Evidently, both these conditions can be satisfied in the present case. The 2+ level of ^{144}Nd at 696 keV has a known mean life of 4.9 psec.[6] The 618-696-keV coincidence can be considered the "prompt" event of our experiment, which is identifiable by a triple coincidence with the 476-keV

γ ray. The "delayed" event is the 476-618-keV coincidence through the 1314-keV 4+ state; this cascade is identified by a triple coincidence with the 696-keV transition. To fulfill the condition that the events be indistinguishable in the timing channels, the 618-keV γ ray common to both cascades was detected by a small NaI detector, while the 476- and 696-keV γ rays were detected with a plastic scintillator. An energy selection channel set below the Compton edge of the 476-keV γ ray will include both transitions without distinguishing between them.

The essence of the technique is the simultaneous acquisition of the "prompt" and "delayed" time spectra. These spectra must be put through the same analog-to-digital converter (ADC), but stored in different memory portions in a multichannel analyzer. One way to accomplish this is to operate the analyzer in a selective storage mode, using the triple coincidences as routing signals. In the present work a two-parameter analyzer was employed. A block diagram of the experimental arrangement is shown in Fig. 2. Two channels, 476 ± 20 keV and 696 ± 25 keV, were set on the spectrum of the 3×3-in.-NaI detector. The *logic* signals corresponding to these channels were put into the Y ADC through a summing amplifier used as a fan-in. One of these two sets of signals was attenuated relative to the other, so that the pulses presented to the Y ADC would be of either one of two pulse heights. Therefore with the analyzer operated in the X-Y coincidence mode, data would be stored in only two Y planes.

The "start" signal for the TAC was derived from a 1.5-in.-diam$\times$1-in.-thick Naton-136 plastic scintillator, while the "stop" signal was from a $\frac{3}{4}$-in.-diam$\times\frac{1}{2}$-in.-thick NaI detector. Use of the NaI detector was necessary for setting a "clean" channel for the 618-keV γ ray. Obviously, because of the inferior energy resolution of plastic scintillators, no energy selection channel can be set for any one γ ray which will completely exclude all other γ rays. Therefore, a plastic scintillator was used only in conjunction with the channel which was intended to include both the 476-keV and the 696-keV transition. This channel extended from the Compton edge of the 476-keV γ ray at 310 keV down to 186 keV. With a narrow window on the 618-keV γ ray, the time spectra had a full width at half maximum of ~0.6 nsec.

Following is a summary of the data-acquisition scheme. With the two-parameter analyzer operating in the X-Y coincidence mode, the time spectra A and B described below were simultaneously obtained and stored:

A. Time spectrum of 476-618-keV coincidences. This spectrum is identified by the simultaneous detection of the 696-keV γ ray in the 3×3-in. NaI detector and is stored in the Y_A plane.

B. Time spectrum of 696-618-keV coincidences. This spectrum is identified by the simultaneous detection of the 476-keV γ ray in the 3×3-in. NaI

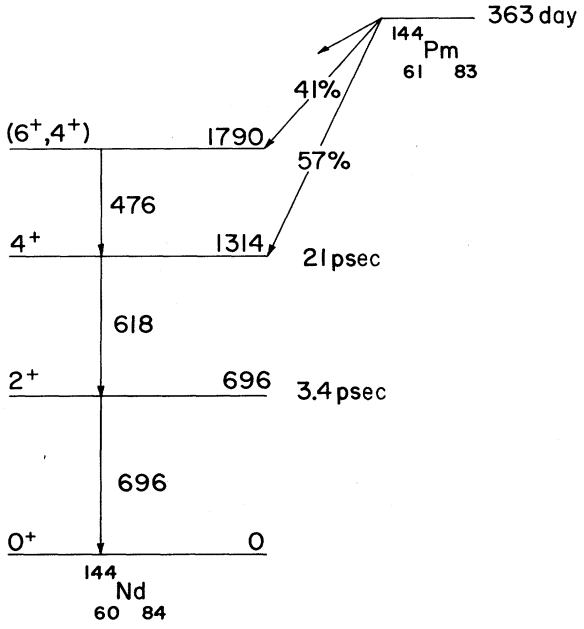

FIG. 1. Partial decay scheme of ^{144}Pm into ^{144}Nd taken from Ref. 3. The 1790-keV level is given a 6+ assignment by Raman, but is reassigned 4+ by Arya, Turk, and Arya. Lifetimes shown are half-lives of the respective states.

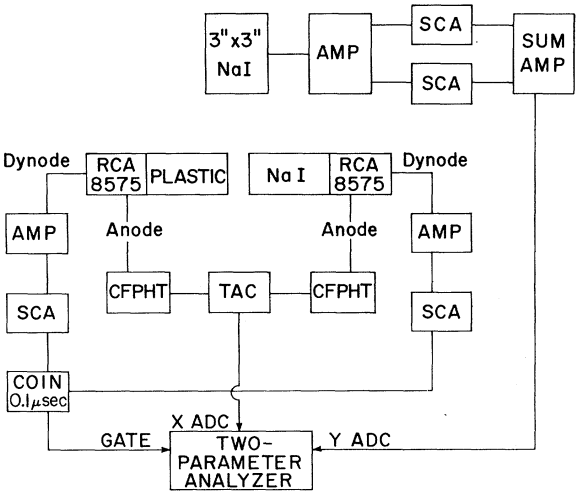

FIG. 2. Schematic diagram of the experimental arrangement for determination of the 1314-keV-state lifetime. CFPHT is the constant fraction of pulse-height trigger timing discriminator; SCA is a single-channel analyzer.

detector and is stored in the Y_B plane.
The measured centroid shift between A and B is $\Delta\tau = \tau_A + \tau_B$, where in principle, τ_A and τ_B are the respective mean lives of the 1314- and 696-keV levels. The lifetimes are added because the centroids of the two spectra shift to opposite sides of the zero of time.

In Table I are given the centroid shifts measured in eight separate runs. The errors indicated therein are purely statistical. With respect to the overall experimental uncertainty, we found that statistics constituted the single largest contribution to the possible error. The particular advantage of this type of self-comparison method is that such factors as gain shifts tend to affect both time spectra in the same way and therefore to a large extent leave the relative shift between them undisturbed. The strength of the radioactive source used was such that there were essentially no accidental triple coincidences. In the discussion below, we will point out several potential sources of error, and show that some are excluded by our experimental arrangement, while others are tractable and appropriate correction can be made for them.

(1) Misidentification of the start and stop pulses in the TAC. The 618-keV channel set for the small timing NaI detector should not include any 476-keV γ rays. Although ~5% of the accepted pulses are due to 696-keV γ rays, this should not disturb spectrum A, since storage in A would require the additional acceptance of the 618-keV γ ray in the 696-keV routing channel. The effect on spectrum B is somewhat more noteworthy. If the roles of the 696-keV and the 618-keV γ rays become reversed, an inversion in the sense of time takes place. A 5% inversion has the effect of making the intermediate lifetime appear 10% shorter. In this instance, the mean life of the 696-keV state would appear to be 4.4 psec instead of the reported[6] value of 4.9 psec.

(2) Misidentification of one set of triple coincidences for the other. This indeed must occur to some extent, since the energy selection window for the 476-keV γ ray necessarily includes some portion of the Compton spectrum of the 696-keV transition. From the singles spectrum taken with the large NaI detector and from the known decay scheme, this portion was estimated to be 18% of the counts in the 476-keV window. For this a correction must be made to the centroid-shift determination.

(3) Counter-to-counter scattering. Potentially this can be a most serious source of error. A typical scattering path length is of the order of several centimeters, which corresponds to traversal times of hundreds of picoseconds. If any of the signals used for fast timing were derived from such scattered quanta, a very large distortion of the centroid shift could result. One way of dealing with this problem is to shield the counters very well from each other. This proved impractical for us because of the low decay rate of our source. We chose to go to the opposite extreme of using a very close-packed geometry, and actually made the scattering work to our advantage.

For purposes of discussion, let the plastic scintillator, the small timing NaI detector, and the 3×3-in. NaI detector be designated as detectors 1, 2, and 3, respectively. We placed detectors 1 and 2 at 180° to each other and detector 3 at 90° to the axis along 1 and 2. The source itself was wrapped with layers of Sn and Cu so that the x rays were attenuated by about an order of magnitude. This has no particular bearing on the present experiment aside from reducing the accidentals.

Recalling the earlier description for time spectra A and B, it is evident that counter-to-counter scattering is not a problem with A. For spectrum A, both the 618- and the 696-keV γ rays are detected as photopeaks in NaI counters. A signal in the third detector can only arise from the 476-keV transition. Since this detector is a plastic scintillator for which only the Compton effect is important, the relevant question which remains is whether the 476-keV γ ray entered detector 1 directly or via scattering. In the event of the latter, the 476-keV γ ray must undergo almost backward scattering. Even for an unlike scattering angle of 120°, the photon entering detector 1 would have an energy of less than 200 keV. This γ ray with its Compton edge at 85 keV would certainly be excluded by the energy selection window set for detector 1. Therefore we conclude that spectrum A was undisturbed by scattering.

Similar arguments are applicable to spectrum B,

TABLE I. Tabulation of centroid shifts observed in the measurement of the lifetime of the 1314-keV state of ^{144}Nd.

Run number (Ref. a)	Centroid shift (psec)
1	39.8 ± 11.7
2	16.4 ± 10.3
3	40.2 ± 10.8
4	31.8 ± 8.4
5	25.3 ± 9.8
6	24.8 ± 7.0
7	35.6 ± 9.4
8	41.6 ± 9.4
Weighted average:	31.0 ± 3.3

[a] Durations of individual runs ranged between 15 to 30 h.

despite the fact that the γ ray detected in 1 is now the higher-energy transition of 696 keV. The scattered γ ray corresponding to 120° has an energy of 229 keV, but this too would be excluded by the energy selection window. Therefore, again it is seen that events satisfying the requirements for spectrum B cannot be due to scattering into detector 1. In this instance, however, the possibility of scattering out of detector 1 into detector 3 is an interesting one. For 50° scattering, the Compton electron has an energy of 228 keV, and the scattered photon 468 keV. If the scattering event occurs in detector 1 and the scattered quantum deposits all its energy in detector 3, then the energy selection criteria for spectrum B are satisfied and the event would be accepted. Such events are not only possible but quite probable, since they require the true coincidence between only the 618- and 696-keV transitions. Yet these events are completely legitimate in the sense that they contain the timing information appropriate to spectrum B. The original purpose of detecting the 476-keV γ ray in detector 3 was precisely to sort out the 618-696-keV cascade for the timing detectors. In having the 696-keV γ ray scatter off detector 1 at ~50° into detector 3, the same purpose is accomplished. By means of auxiliary experiments, we ascertained that 40% of the events stored in spectrum B were due to the type of scattering just described.

A final comment might be made on the question of summing. Although some pulses in the 618-keV channel in detector 2 may be due to the summing of 476-keV γ rays with γ rays backscattered from 1, coincidences with these false signals are excluded by the energy-selection requirement for detector 1. A window of 186 to 310 keV corresponds to scattering of 618-keV γ rays only to the forward hemisphere.

(4) Effect of amplitude walk. If the pulse-height distributions within an energy selection window are not exactly alike for two γ rays, the phenomenon of apparent time shift with pulse amplitude may give rise to a systematic error. Within the channel used for the present experiment, the spectra in the plastic scintillator for the 476- and 696-keV γ rays were indeed not identical. Furthermore, the effect of this difference could not have been deduced from the respective singles components because the counter-to-counter scattering described above favors one energy range over another. To investigate the true pulse-height distributions which gave rise to the time spectra, the following measurement was carried out. With the triple-coincidence requirements unchanged from the lifetime measurements, linear signals from the plastic scintillator were presented to the X ADC, gated by double coincidences and routed to two Y planes as before. The stored spectra were then analyzed for the centroids of their respective distributions. The difference in the centroids was found to be 8.2 keV.

To correct for the effect of this energy difference requires a knowledge of the equivalent time shift. Since the timing discriminator [ORTEC 453, used in the constant fraction of pulse-height trigger (CFPHT) mode] was initially set for what appeared to be optimal performance, the remanent walk was small and difficult to determine. According to the manufacturer's specifications, the walk over the full dynamic range of the CFPHT is typically within ±120 psec. In the absence of an experimentally verified number, however, the residual walk was assumed to be $\leq$200 psec/MeV. An energy difference of 8.2 keV could then produce a systematic error of 1.6 psec.

In summary, the measured centroid shift in the time spectra

$$\Delta\tau = \tau_A + \tau_B = 31.0 \pm 3.3 \text{ psec}$$

must be interpreted as follows: In principle, τ_A and τ_B are the respective mean lives of the 1314- and 696-keV levels. In practice, however, τ_B is a composite of τ_{696} and τ_{1314} because 18% of the time triple-coincidence events in spectrum B are due to the presence of the 696-keV γ rays in the 476-keV window used for routing. Since 40% of the stored events in B are due to counter-to-counter scattering, the portion of τ_{1314} present is 18% of the remaining 60%. Therefore

$$\tau_A = \tau_{1314}$$

and

$$\tau_B = 0.89\tau_{696} - 0.11\tau_{1314}.$$

The negative sign in the latter expression comes about because, as mentioned earlier, the two centroids shift to opposite sides of the zero of time. Because of a 5% time sense inversion in the τ_{696} portion, the apparent value of 4.4 psec should be used instead of the true value of 4.9 psec. It then follows that $\tau_{1314} = 29.4 \pm 3.7$ psec. This result, however, has not accounted for possible systematic error from amplitude walk. To include that possibility, the estimated 1.6-psec uncertainty associated with walk is added to the 3.7 psec associated with statistics to give a final result of

$$\tau_{1314} = 29.4 \pm 5.3 \text{ psec}$$

for the mean life of the 1314-keV level. This result is in serious disagreement with the previously reported[2] value of $(13 \pm 4) \times 10^{-11}$ sec.

DISCUSSION

The single-particle mean life of a 618-keV $E2$ transition is 10^{-10} sec. Compared with this value, the 618-keV $4+ \rightarrow 2+$ transition is enhanced by only a factor of 3. In this respect, although our lifetime value is in quantitative disagreement with the earlier measurement, the qualitative picture is not dramatically altered. From the present result it can be deduced that

$$B(E2, 4+ \rightarrow 2+) = (0.03 \pm 0.005) \times 10^{-48}\ e^2\,\mathrm{cm}^4 .$$

Since $B(E2, 0+ \rightarrow 2+) = (0.51 \pm 0.016) \times 10^{-48}\ e^2\,\mathrm{cm}^4$,[7] the following ratio is obtained:

$$\frac{B(E2, 4+ \rightarrow 2+)}{B(E2, 2+ \rightarrow 0+)} = 0.30 \pm 0.05 .$$

This ratio evidently disagrees with the value of 2 for vibrational nuclei. In fact, this result does not seem to be explainable by any of the several simple collective models.

A new value for the lifetime having been established, the g factor reported in Ref. 1 should be recomputed accordingly. The perturbed-angular-correlation experiments in Ref. 1 measured the product $g\tau$. Preserving the same value for this product, the g factor would now be

$$g_{4+} = 0.2 \pm 0.2 .$$

This number is no longer at obvious variance with the hydrodynamical value of $Z/A = 0.4$. Therefore, from the g factor alone, it cannot be argued that a collective picture is unacceptable. The datum not easily accomodated remains that of the relative hindrance of the $4+ \rightarrow 2+$ transition in comparison with the $2+ \rightarrow 0+$ transition. Such nonenhancement of the $4+ \rightarrow 2+$ transition does not appear to be unique to ^{144}Nd. As pointed out in Ref. 1, a similar situation exists in ^{140}Ce. One suspects that more cases are not known only because very few 4+ state lifetimes have been measured outside the regions of strong deformation. For any specific case, there is little doubt that an explanation can be found, for instance by invoking appropriate shell-model configurations. It would be very interesting, however, to see this phenomenon satisfactorily described in the framework of collective models which purport to span the near spherical, as well as strongly deformed regions.

ACKNOWLEDGMENT

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