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Publication Date
1963-06-01

UCRL-10792

University of California

Ernest O. Lawrence Radiation Laboratory

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Berkeley, California

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Submitted to Nuclear Physics

UCRL-10792

UNIVERSITY OF CALIFORNIA

Lawrence Radiation Laboratory
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Contract No. W.7405-eng-48

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ABSTRACT

Some features of the decay scheme of 33-hour Ce^{143} have been studied with a 50-cm radius iron-free beta-spectrometer equipped with a nanosecond "time sorter."

From the measured L-subshell conversion ratios the 57.37 ± 0.05-keV transition is found to be predominantly M1, with less than 0.3% E2 admixture. The half-life of the 57-keV state is measured as 4.17 ± 0.09 nanoseconds. Similarly, the 293.3 ± 0.1-keV M1-E2 transition, de-exciting the 350.7-keV level, is found to have a 34 ± 16% E2 admixture and half-life $\leq$ 0.3 nanosecond. The K-line of the 350.7 ± 0.1-keV crossover transition has also been observed.

Analysis of these data, in combination with angular correlation results of other experimenters, supports a 5/2+ assignment for the 57.37-keV level, and suggests a 3/2+ assignment for the 350.7-keV state. The magnetic moment of the 57-keV state is discussed in terms of the presently available data.

PROPERTIES OF THE 57- AND 351-keV EXCITED STATES IN $_{59}Pr^{143}$ *

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1. Introduction

The properties of the low-lying excited states of the odd-mass praseodymium isotopes (Z = 59) have not been clearly established. In the case of Pr^{141}_{82}, the early measurements of the spin and magnetic moment[1] supported a ground state assignment of 5/2+, and the single-particle orbital $d_{5/2}$ has usually been used in describing this state. Gamma-ray angular distribution data from aligned nuclei support a 7/2+ assignment for the first excited state at 142 keV.[2] This is the typical situation in the $40 < Z < 50$ region: close-lying 5/2+ and 7/2+ states, often crossing.

In the case of Pr^{143}_{84}, the ground-state spin has recently been measured[3] to be 7/2, rather than 5/2 as had previously been assumed.[4,5] It is thus of interest to examine the properties of the 57-keV first excited state to learn, among other things, if the frequently observed 5/2+ and 7/2+ states have again crossed between Pr^{141} and Pr^{143}. In particular, the half-life of this state and the multipole character of its de-excitation transition are important for the interpretation of low-temperature alignment and angular-correlation experiments being conducted on Ce^{143} at this laboratory.[2,6] We report here measurements of these two quantities. In addition, we have obtained a value for the M1-E2 mixing in the 293-keV transition which also complements the angular correlation and alignment data.

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2. Experimental Method

A sample of 18-hour Ce^{143} was prepared by irradiating ~ 1 mg of cerium oxide enriched† to 87% in Ce^{142} for 48 hours in a neutron flux of 2×10^{14} n-cm^{-2}-sec^{-1} at the Materials Testing Reactor, Arco, Idaho. This active material was dissolved in concentrated HCl and transferred to a boat-shaped tantalum filament in which it was dried. Sources were prepared by subliming the material in vacuuo through a 1 mm × 10 mm collimator on to a 7 mg-cm^{-2} Al foil backing. The estimated mean surface density of the source deposit was $\approx$ 30 μg-cm^{-2}.

The Berkeley 50-cm radius iron-free spectrometer[7,8] was used in this investigation for scanning the L conversion lines in detail. The electron detector was a Geiger counter having a window aperture 2 mm wide × 20 mm high. The latter was covered with a multilayer formvar film of surface density ~ 100 μg/cm^2 which is > 97% transparent to 40 keV electrons. With a baffle-aperture solid angle of ~ 0.06% of 4π steradians the expected instrumental resolution is ~ 0.1% in momentum. The spectrometer was operated automatically and the output data recorded by an electric typewriter. The current was programed to advance 4 mA after each readout. As a check on the performance of the automatic current control system the current was occasionally measured manually by observing the voltage drop across a 0.01 ohm manganin precision resistance using a Leeds and Northrup type K2 potentiometer.

The experimental arrangement for measuring nanosecond lifetimes is shown schematically in fig. 1. The spectrometer baffle aperture was opened up to a solid angle of ~ 0.6% of 4π steradians (~ 0.5% resolution) in order to maximize the electron counting rate. The electron detector consisted of a 10-mm wide × 25-mm high × 1-mm thick sheet of Naton 136 plastic phosphor optically coupled

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† This material was obtained from the Separated Isotopes Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

to a 14-stage CBS 1090 phototube. A larger plastic phosphor mounted on a 14-stage RCA 6810A was placed behind the source to detect energetic β rays. The timesorter circuit indicated schematically in the left (FAST) side of fig. 1 is similar to that used by Geiger et al.[9] in conjunction with the 1-meter radius iron-free spectrometer at Chalk River. A feature of this arrangement is that the timesorter can be calibrated absolutely in the manner described by Graham et al.[10] The electrical lengths of the cables in the delay unit were calibrated with an absolute accuracy of $\lesssim 0.05$ n-sec. Before and after each lifetime measurement the overall gain of the time converter (channel number vs. delay unit setting) was checked at 4-nanosecond intervals; and typically the drifts were $\lesssim 1\%$ of the output pulse height (channel number).

It should be noted that the output pulse height from the time to amplitude converter for "prompt" lifetimes is a function of the focused-electron energy. The upper right inset of fig. 1 shows the computed relationship between transit time and electron energy. The vertical distance between the two dashed curves is the spread in transit time which limits the effective resolving time to $\gtrsim 2$ nanoseconds for the largest baffle aperture in the spectrometer.

3. Results and Analysis

3.1. CONVERSION ELECTRON SPECTRUM

The L internal conversion electron spectrum of the 57.37-keV transition is shown in fig. 2. The observed linewidth of $\sim 0.15\%$ in momentum is somewhat greater than that expected for the spectrometer settings used (i.e., $\Delta p/p \approx 0.12\%$) and is attributed to the finite source thickness. Using the background rate indicated by the horizontal dashed line and fitting with the L_I peak shape we deduce the following ratios for the relative line intensities:

$$0.0725 < L_{II}/L_I < 0.087$$

and

$$0.0241 < L_{III}/L_I < 0.0347 \quad .$$

The multipolarity of this transition is predominantly M1 with a very small admixture of E2. In an attempt to establish the magnitude of the latter we compare the intensity ratios with the theoretical predictions of Sliv[11] and of Rose[12] in fig. 3. The cross-hatched regions between the vertical arrows indicate the uncertainty in the mixing ratio due to our experimental error limits. We note that for Rose's predictions (lower part) the two ratios are consistent with each other for δ^2 = E2/M1 from 0.08% to 0.16% E2. However, the analysis in the upper part (Sliv theory) shows an inconsistency outside of the indicated experimental error limit but does indicate that the magnitude of δ^2 is small, i.e., $\lesssim$ 0.3%.

The transition energy of 57.37±0.05 keV was deduced by taking advantage of the presence of a trace amount of 47-hour Sm^{153} activity in one of the Ce^{143} sources. The K line of the 103.18±0.02-keV transition[4,5] in Eu^{153} at a current setting about 4% higher than the L_I 57.37 line provided a convenient internal calibration line.

The K-conversion lines of the cascade and crossover transitions from the second excited state were observed, and our measured values of their transition energies are 293.3±0.1 keV and 350.7±0.1 keV, respectively. From an observation of the L-conversion spectrum of the 293.3-keV transition we find the following approximate subshell ratios: L_I/L_{II} = 5.2±0.7 and L_I/L_{III} = 13±4. From these results we obtain for the E2 mixing in the 293.3-keV transition the value 34±16%. A value of this admixture has also been reported by Haag et al.[2] as a result of their studies of the temperature dependence of the linear polarization of the 293-keV photon following the decay of oriented Ce^{143} nuclei; their results are consistent with δ = - 0.8, which corresponds to an E2 admixture of 39%.

3.2. LIFETIME MEASUREMENTS

Fig. 4 shows one of the "timesorter" spectra measured in the course of this work. The detector behind the source was biased to register only energetic β (or γ) events ($\gtrsim$ 150 keV) and the spectrometer current was set to focus the L 57.37 electron line. The closed circles show the data accumulated in 5 hours after subtracting a random coincidence rate of 39 counts per 5 channels. The 4 nanosecond marks on the inset time calibration scale are the mean of the channel numbers observed with the pulser calibration method before and after this measurement. The "prompt" curve illustrates the performance of the time-sorter; the width of 3.3 nanoseconds is approximately that expected from the spread in the transit times of 50-keV electrons in the spectrometer (see inset of fig. 2). "Prompt" events were observed experimentally at a spectrometer current just above the L 57 line (at 10.8 A). As plotted the data points have been corrected for the difference in the mean transit time of the electrons (1.3 n-sec) and have been normalized to the same total intensity. Another β-L 57 time spectrum of comparable statistical accuracy was also measured on a different occasion with a different source.

The experimental results were analyzed by a least squares computer program (IBM 7090) to fit the expression $A + B \exp(-\lambda t)$ for the data in channels above 160. Taking the mean of the two λ values and including an allowance for the uncertainty in the time calibration we quote for the half life of the 57.37 state in Pr^{143}:

$$T_{1/2} = 4.17 \pm 0.09 \times 10^{-9} \text{ sec} \quad .$$

Recently El-Nesr and Bashandy of the Uppsala group have reported a value of $2.9 \pm 0.2 \times 10^{-9}$ sec for this half-life.[13] Their value is shorter than that obtained in our measurements by $\sim$ 30%, which is well outside the quoted limits of error.

The β-K 293.3 timesorter spectrum has also been measured (not shown) and compared with a "prompt" spectrum observed when the spectrometer current was set slightly above the K 293.3 line. From the slope of the side of the resolution curve we can set as a limit for the 350.7-keV state:

$$T_{1/2} \leq 0.3 \times 10^{-9} \text{ sec}.$$

This is consistent with the somewhat shorter limit of 0.1 nanoseconds of Gorodetsky et al.[14]

As an independent check of the performance and calibration of our apparatus we have also measured the lifetime of the 102.18-keV state in Eu^{153} using the first of the Ce^{143} sources which had a trace of 47-hour Sm^{153} in it. The data from one of the two timesorter spectra observed is displayed in fig. 5. The data from both were again analyzed by the least squares method and the resultant mean value, $4.0 \pm 0.2 \times 10^{-9}$ sec, agrees within the quoted errors with the values obtained by Nainen (3.80±0.02 n-sec)[15], Vergnes and Marty (4.0±0.2 n-sec)[16], and Graham and Walker (4.0±0.2 n-sec)[17], although that of Reyes-Suter and Suter in Uppsala (3.3±0.2 n-sec)[18] is somewhat shorter.

4. Discussion

A firm basis for discussion of the properties of the low-lying excited states of Pr^{143} has been given by the recent measurement by Budick and Marrus[3] of the ground state spin (7/2). The magnetic dipole character of the 57.37-keV transition allows the possibilities 5/2, 7/2, or 9/2 for the first excited state. From shell-model arguments the ground state is given the assignment $g_{7/2}$ and the preferred choice for the first excited state, based on the systematics of this region, would be $d_{5/2}$. It is of interest to examine the available experimental data to see if a unique assignment can in fact be made for the first excited state.

A 293-57 keV gamma-gamma directional correlation experiment has been reported by Rao and Hans.[19] Although their interpretation, based on the assumption of 5/2+ for the Pr^{143} ground state, is invalidated by the recent spin measurement of 7/2, we shall utilize their reported value of $A_2 = + 0.132 \pm 0.019$ together with our measured E2-M1 mixing ratios of the 293- and 57-keV transitions for a reinterpretation of the excited state angular momenta.

Fig. 6 shows the results for the cascade $j_1 \xrightarrow{M1\text{-}E2} j = 5/2 \xrightarrow{M1\text{-}E2} j_2 = 7/2$, analyzed according to the graphical method of Arns and Wiedenbeck[20] based on the theoretical treatment of Biedenharn and Rose.[21] No graphical fit could be found for intermediate spin choices $j = 7/2$ or $j = 9/2$ with any of the six possible values of j_1, and the appropriate curves for those choices are not shown. Of the three choices for j_1 illustrated in fig. 6 ($j = 5/2$) it is seen that $j_1 = 3/2$ and $j_1 = 7/2$ are consistent with the angular correlation and multipolarity data. This establishes the spin of the 57.37-keV state as $j = 5/2$. This is not consistent with the conclusion of Haag et al.[2] that "the 351 and 57 keV states have the same spin." However, it might be noted that our multipolarity limits for the 294-keV transition are consistent with their value of the mixing parameter, $\delta \approx - 0.8$, i.e., 39% E2.

One might expect the magnetic moment of the 57.37-keV, $j = 5/2$, excited state of Pr^{143} to be similar to the 5/2+ ground state of Pr^{141}. The experimental measurement of the latter[4,5] recalculated by Lindgren[22] gives the result $\mu \simeq 5.1$ nuclear magnetons which is slightly larger than but probably consistent with the upper ($d_{5/2}$) Schmidt limit of $\mu = 4.79$. Levy and Shirley[6] at this laboratory have been studying the rotation of the γ 293-γ 57 correlation pattern in a uniform magnetic field of 10.8 kilogauss. Their preliminary result is

$$\theta = \omega\tau = 0.51 \pm 0.08 \text{ radians} \quad .$$

From this result and our measured lifetime of the 57.37-keV state we can calculate its magnetic moment,

$$\mu_I = \frac{(\omega_H \tau) I \hbar}{\tau \mu_o \beta \mathcal{H}}$$

where $\omega_H \tau$ = measured angle of rotation of the correlation

τ = mean life of intermediate state

I = spin of intermediate state

$\mathcal{H}$ = applied field strength

β = magnetic field enhancement factor caused by unpaired valence electrons.

Fig. 7 shows the value deduced for μ as a function of the parameter β. The initial ionic state of the Ce^{143} is thought to have been +3 in the experiments of Levy and Shirley, and one might expect Pr^{+4} or Pr^{+3} as the final ionic state after beta decay. The β values predicted for the +3, +4, and +5 ionic states[23] are indicated in the figure. It is seen that the assumption of a Pr^{+5} ionic state ($\beta = 1.0$) would provide the most satisfactory interpretation of these results although this does not seem as likely as the other two alternatives.

We might speculate briefly on the spin of Ce^{143}. The experiments of Martin et al.[24] suggest that the beta transition to the Pr^{143} ground state is highly hindered (log ft > 9), whereas those to the 57 and 341 states have log-ft values of 7.6 and 7.2, respectively. This suggests that the spin of the 341-keV state differs from that of the ground state, i.e., favors our 3/2+ alternative. From the shell model, Ce^{143} is expected to have odd parity, and 7/2, 5/2, and 3/2 are possible spin assignments. Of these, 3/2 ($p_{3/2}$) is most easily reconciled with the data. It should be noted that Nd^{145} and Sm^{147}, which also have 85 neutrons, have measured spins of 7/2. In the case of Nd^{145}, however, 3/2+ is the preferred assignment for the first excited state.[5]

5. Acknowledgments

This investigation was suggested to us by Professor D. A. Shirley, and we wish to thank R. M. Levy and Professor Shirley for permission to quote their preliminary results. We acknowledge with thanks technical assistance given by C. Butler, S. Clark, J. Harris, G. Kilian, D. McClure, and P. Salz.

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

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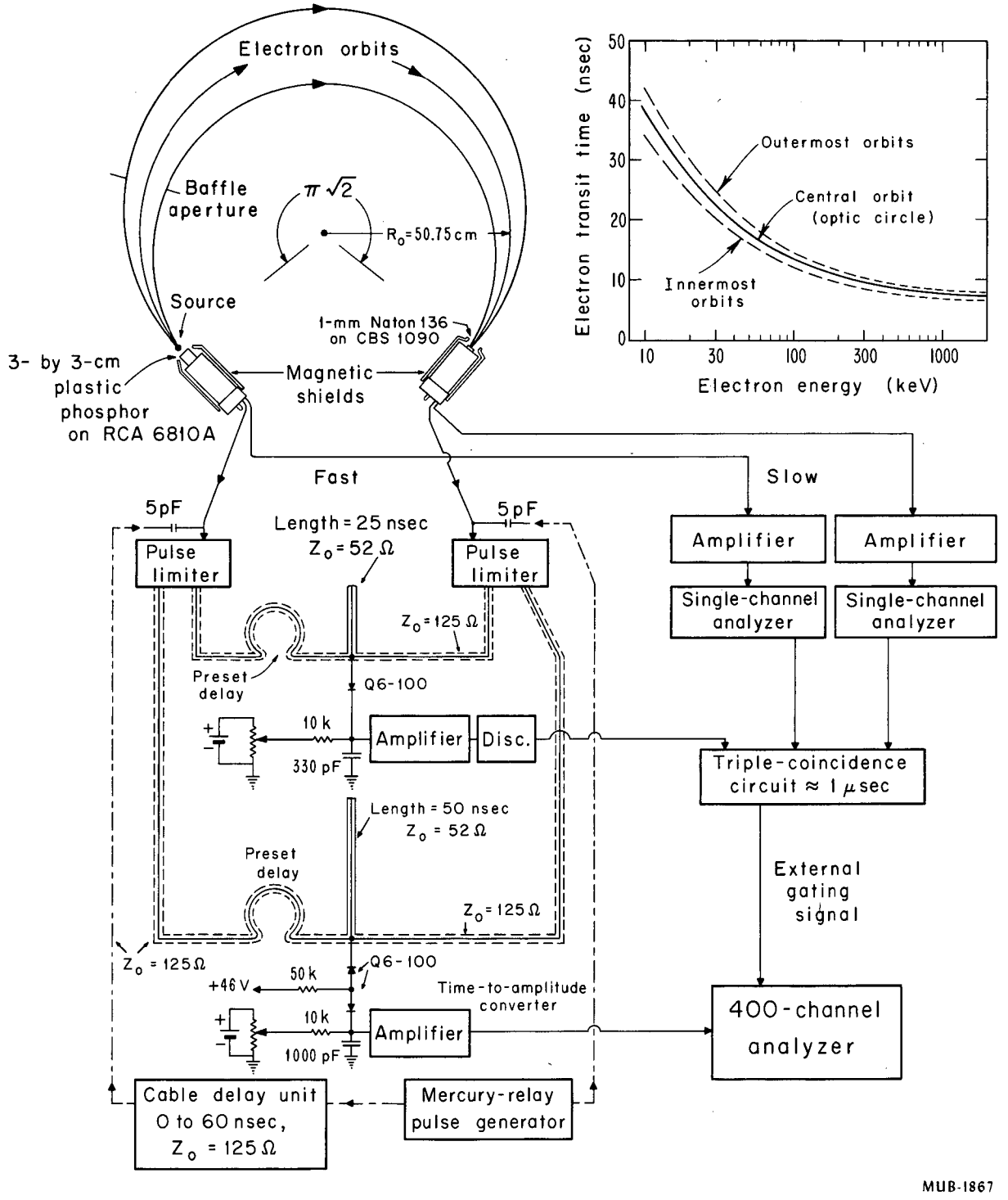

Fig. 1 Schematic diagram of the timesorter arrangement used to measure lifetimes with the 50-cm radius $\pi\sqrt{2}$ spectrometer. The inset at the upper right shows the predicted variation of electron transit time in the spectrometer as a function of electron energy.

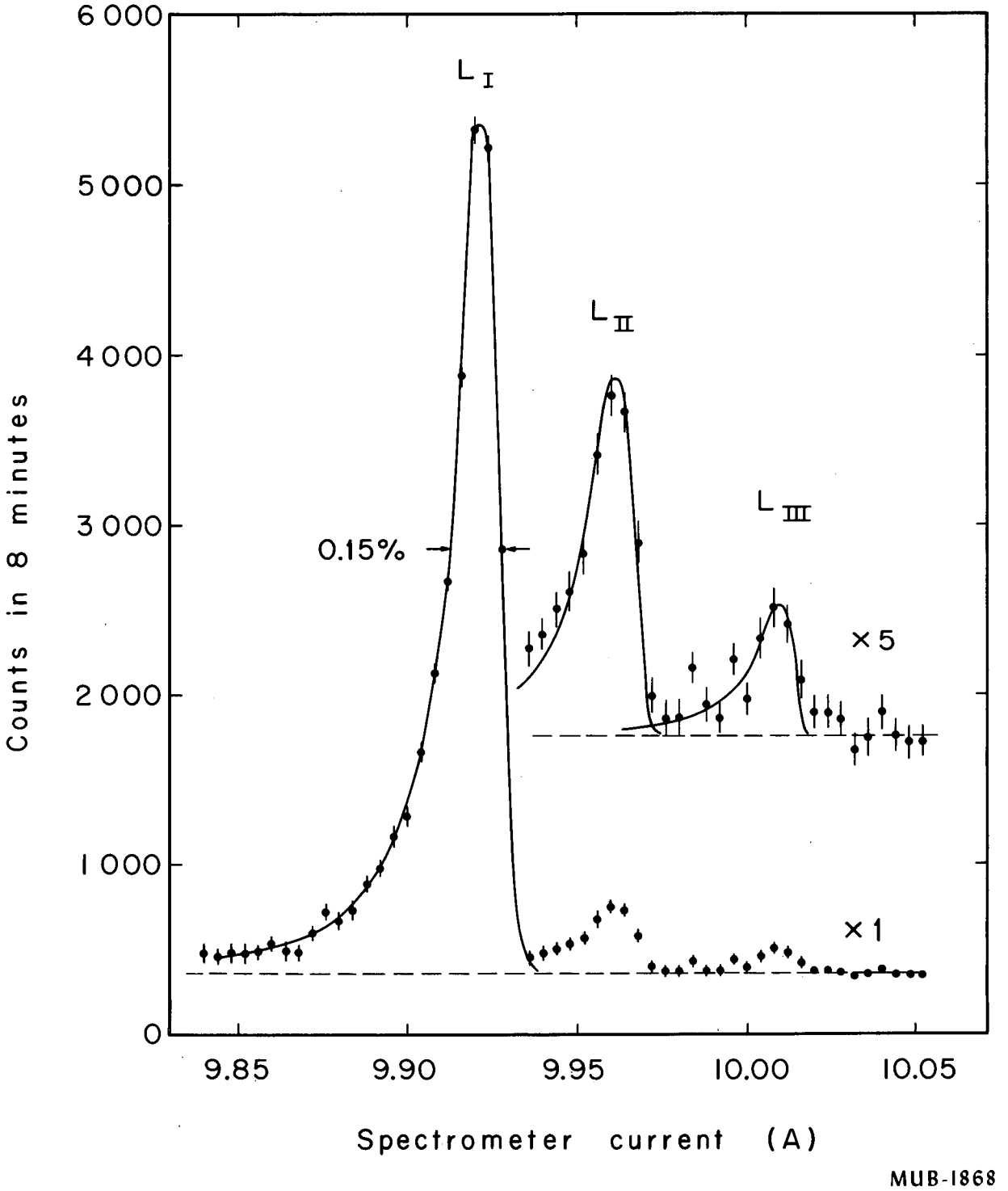

Fig. 2 L internal conversion electron spectrum of the 57.37-keV transition in Pr^{143}.

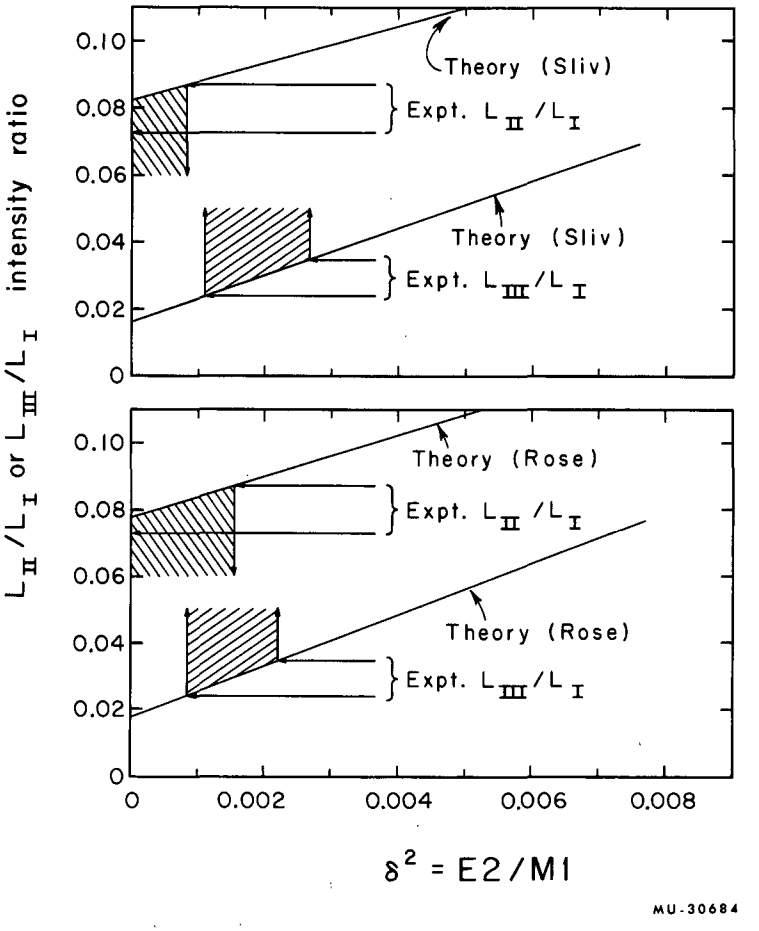

Fig. 3 Deduction of the mixing ratio, δ^2 = E2/M1 for the 57.37-keV transition from the observed L subshell intensity ratios. The theoretical curves in the upper part were deduced from the tables of Sliv[11] and those in the lower part from the tables of Rose.[12]

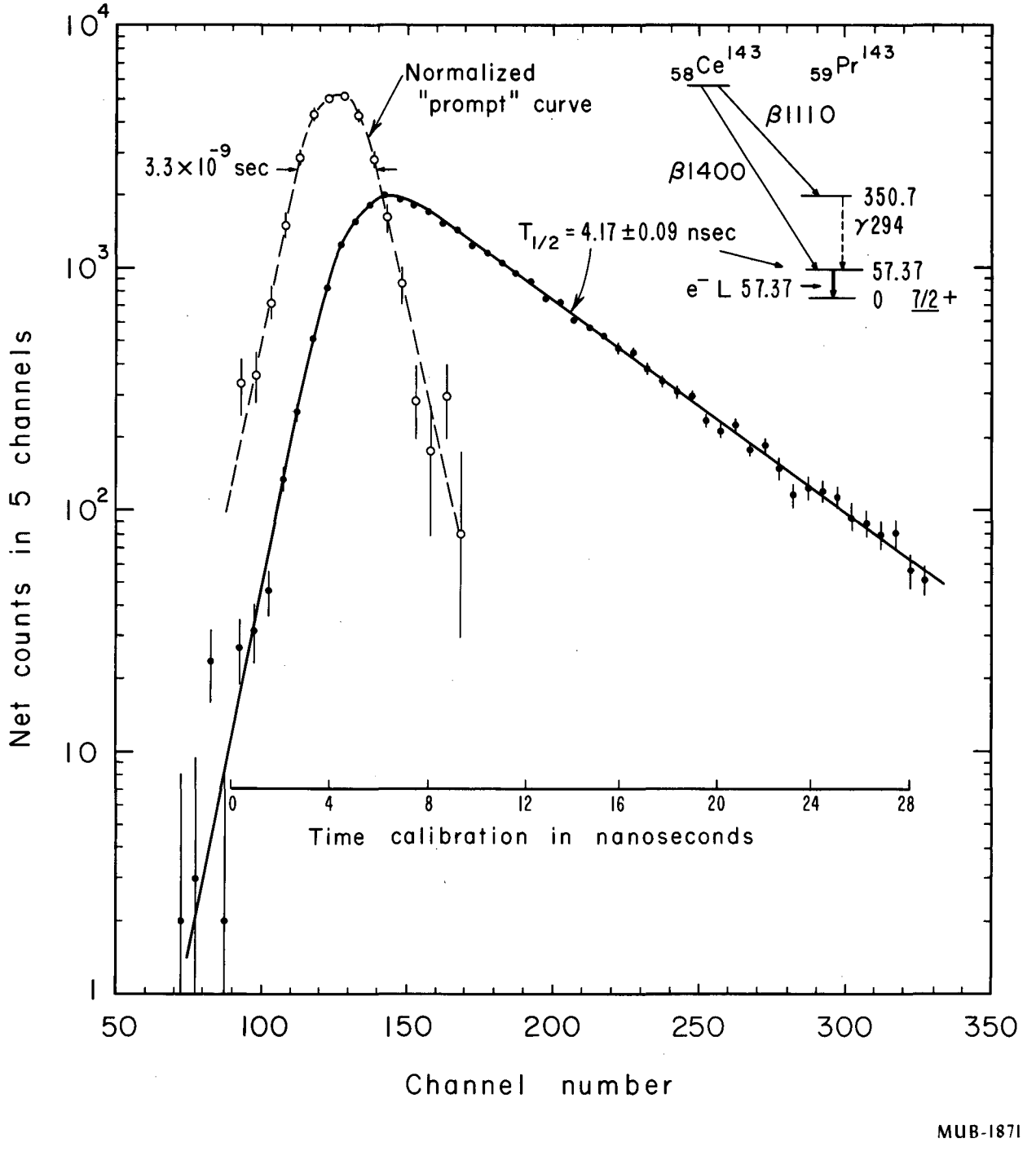

Fig. 4 One of two timesorter spectra observed with focused L 57 conversion electrons.

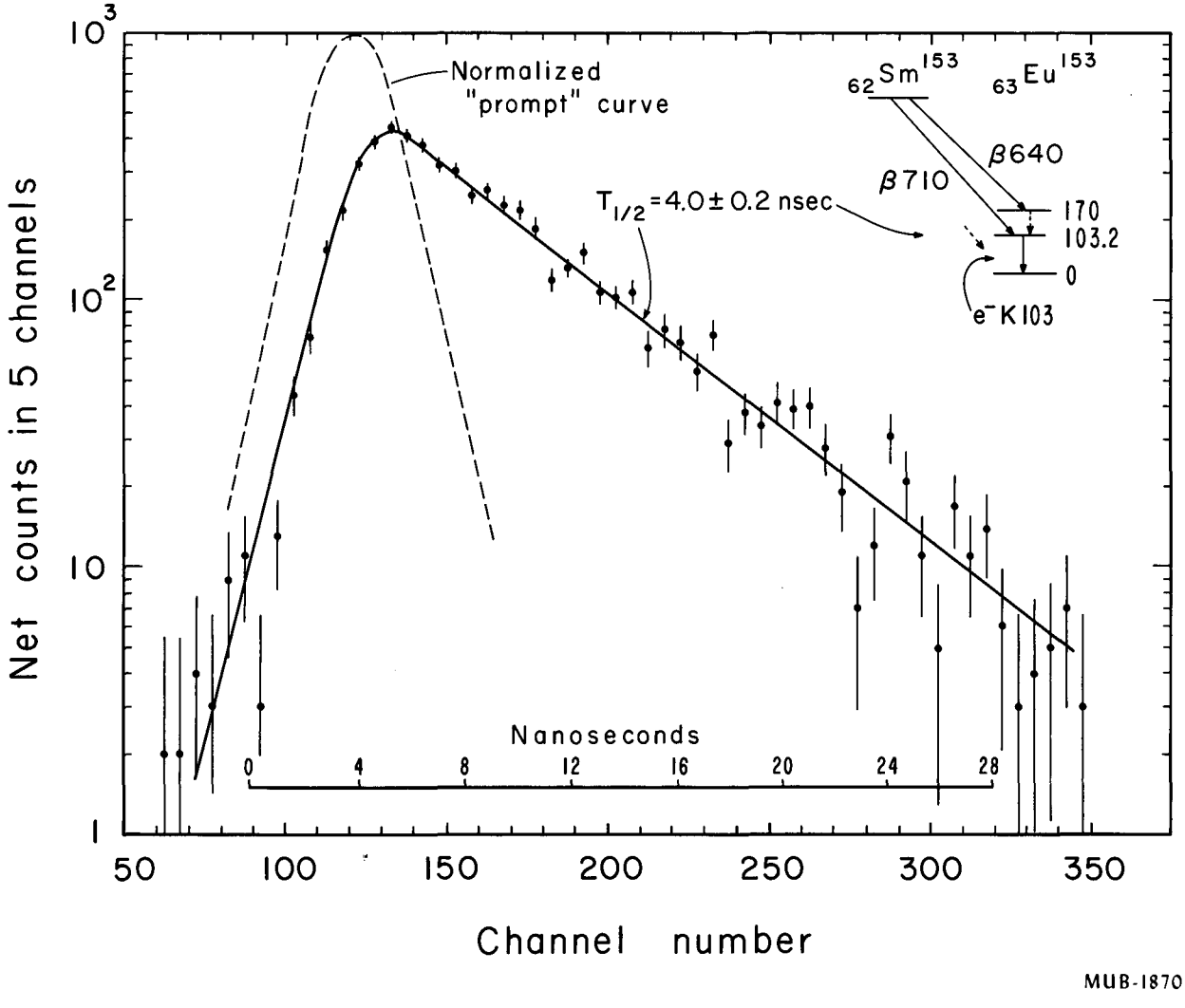

Fig. 5 One of two timesorter spectra observed with focused K 103 electrons (Eu^{153}).

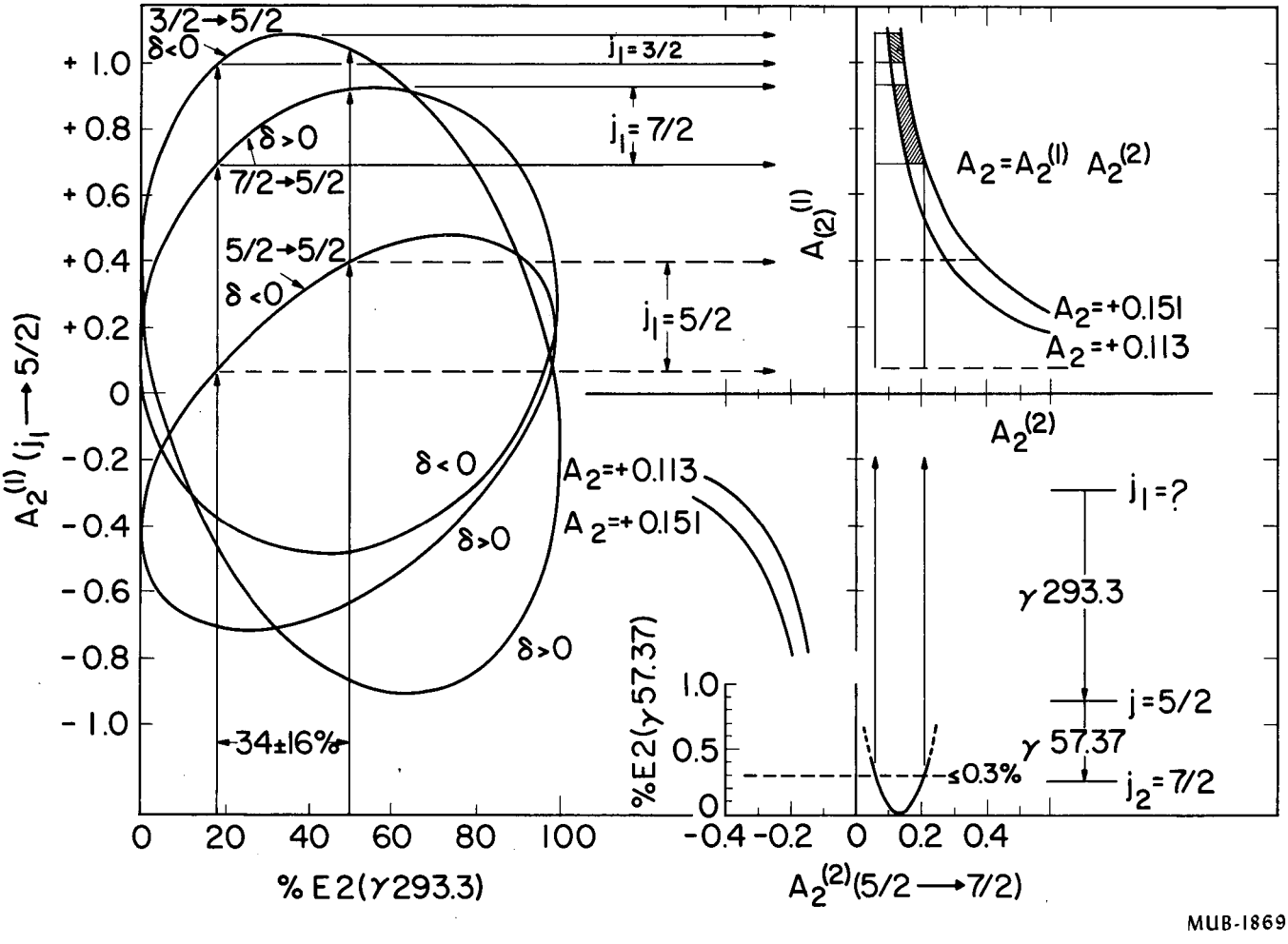

Fig. 6. Graphical analysis of the 293-57 keV gamma-gamma directional correlation according to the method of Arns and Wiedenbeck.[19] The elliptical curve for $A_2^{(2)}$ $(5/2 \rightarrow 7/2)$ was calculated directly from the tables of Biedenharn and Rose.[20] The indicated E2 admixtures were obtained in this work, and the value of the directional correlation coefficient, $A_2 = +0.132 \pm 0.019$, is that reported by Rao and Hans.[18] The cross-hatched areas show the regions of consistency for these three pieces of information.

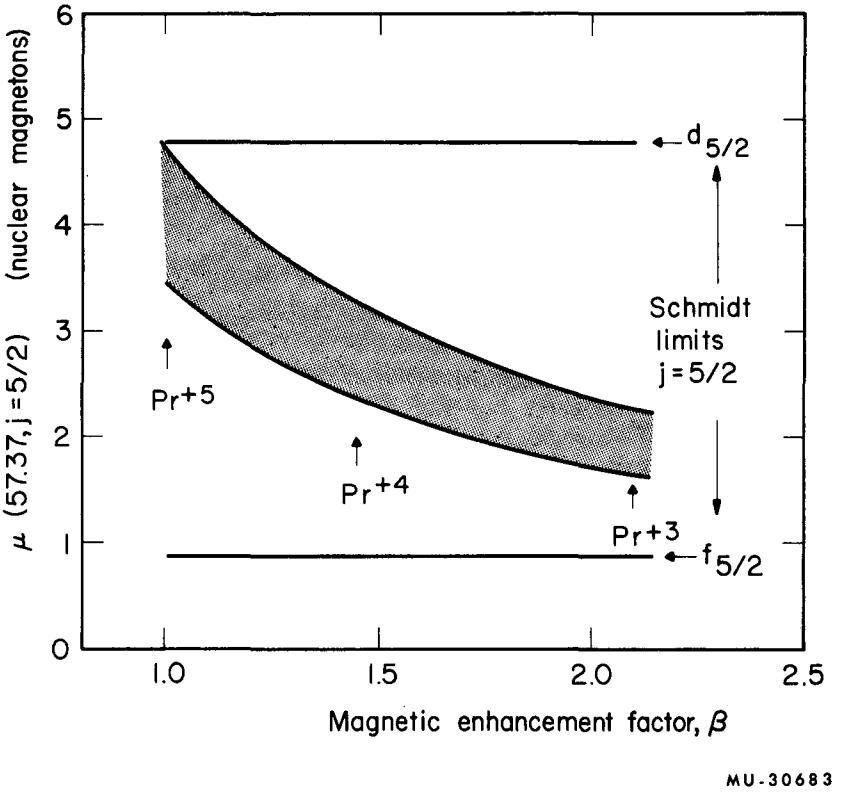

Fig. 7 Magnetic moment of the 57.37-keV state in Pr^{143}. The cross-hatched region shows how the values of the moment calculated from the experimental data depend upon the magnetic enhancement factor, β.