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Lifetimes of nuclear levels in the decays of some mass-separated fission products

James Albert Morman
Iowa State University

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Lifetimes of nuclear levels in the
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by

James Albert Norman

A Dissertation Submitted to the
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I. INTRODUCTION

Nuclear levels were thought to have half-lives of the order of $10^{-13}$ sec until 1921. At that time Hahn (1) discovered a substance essentially the same as $^{239}\text{Pa}$, but which had a different half-life for decay by beta-ray emission than that which was normally observed. This was the first discussed isomer, a nucleus which has the same mass and charge as another, but has a different half-life.

After artificially induced radioactivity was discovered in 1934, many other isomers were found. Confirmation of the existence of isomeric levels first came from Kurchatov et al. (2) who reported a new activity in neutron irradiated bromine. Bothe and Gentner (3,4) and Snell (5) identified this new activity as an isomeric level in $^{80}\text{Br}$.

Since that time, experimental equipment and techniques have improved to the point where half-lives of the order of $10^{-12}$ sec are being measured. Many reviews of nuclear isomerism have been published (6,7,8). Figure 1 shows just some of the many isomeric nuclei with odd mass number $A$ that have been found. The data for Fig. 1 was taken from a compilation by Kantele and Tannila (9).

It is clear from Fig. 1 that the distribution of isomers is discontinuous at values of odd proton or odd neutron numbers of 50, 82 and 126. It was this feature that added impetus to the development of the shell-model theory.
Figure 1. Distribution of isomers with odd mass number as a function of neutron or proton number.
in 1950 by Mayer (10) and Hazel, Jensen and Suess (11).

In the region just before closed shells, levels of high spin are being filled simultaneously with levels of low spin. For example, near the closed shell at 50 nucleons, these levels are $g_{9/2}$ and $P_{3/2}$. Because of the large spin difference, transitions between these levels are expected to have long half-lives.

Using the single-particle shell model, Weisskopf (12) has derived expressions for the transition probability between two states of given angular momentum. These results have been simplified by Moszkowski (13). Using these estimates, the half-lives of the four lowest multipole order transitions have been plotted in Fig. 2 as a function of energy.

The multipolarity of a transition reflects the angular momentum and the parity of the emitted photon. Angular momenta of 1, 2, etc. correspond to dipole, quadrupole, etc. radiations. If the angular momentum of the initial nuclear state is $j_i$ and that of the final state is $j_f$, the possible values of the angular momentum of the photon, $L$, are limited by the selection rule

$$|j_f - j_i| \leq L \leq |j_f + j_i|.$$  

Classification as an electric or a magnetic transition can be determined from the parity of the two levels involved. If the parity of the two levels is the same, then $\Delta \pi = 0$; if
Figure 2. Weisskopf estimates for the half-lives of E1, M1, E2 and M2 transitions
they are different, then $\Delta \pi = -1$. For electric transitions of multipole order $L$,

$$\Delta \pi = (-1)^{2L}$$

and for magnetic transitions of order $L$,

$$\Delta \pi = (-1)^{2L-1}.$$

From Fig. 2 it is clear that the half-lives are expected to be longer for transitions of lower energies. Consideration of these half-life estimates and the shell-model level scheme were two factors which determined the time region in which the search for measurable lifetimes was centered in this study.

The isotopes available from the TRISTAN isotope separator (to be discussed later) have both neutron and proton numbers just above the closed shell numbers. In the high mass-number range of the separator, the proton numbers are just beyond the magic number 50, and the neutron numbers are just beyond 82. In most cases then, one would expect low multipolarity transitions according to the shell model. Figure 1 makes it evident that little work has been done in the field of lifetime measurements for these nuclei just beyond the magic numbers. The results of this study, then, will add to the data available for study in this region.

It can be seen from Fig. 2 that the low multipolarity transitions have half-lives on the order of nanoseconds for very low energies. While NaI-NaI coincidence systems can
easily measure lifetimes in this range, the poor energy res-
olution of such detectors presents an energy peak selection
problem at low energies, especially for complex decay
spectra. However, a Ge(Li) detector with its superior ener-
gy resolution can be used in place of one of the NaI
detectors.

Thus it was decided to use a Ge(Li) detector with a
small active volume, especially suited for low-energy gamma-
ray detection, as one of the two detectors in the
coincidence timing circuit. This choice then set the lower
limit of the measurable time range to approximately a few
nanoseconds, depending on the exact energy of the transi-
tion. The Ge(Li) detector used, because of its inefficiency
at energies above a few hundred keV, also set an upper limit
to the energy range studied. The second detector was chosen
to be a plastic scintillator mounted on a fast photo-
multiplier tube. Even though the plastic scintillator has a
poorer energy resolution than NaI, it provides a faster
timing signal. Since the Ge(Li) detector is used for energy
determination, the plastic scintillator was chosen to pro-
vide a start signal for the coincidence circuit.

This study is then centered on a search for nuclear
levels in several fission products, $^{136}$Xe, $^{134}$Cs, and $^{131}$Cs,
with half-lives of a few nanoseconds or longer, and with en-
ergies usually less than about 200 keV.
II. EXPERIMENTAL ARRANGEMENT

A. TRISTAN Isotope Separator System

The nuclei examined for possible isomeric states were produced using the TRISTAN isotope separator (14,15), operating on-line to the Ames Laboratory Research Reactor. The system is shown in Fig. 3 and is described briefly below.

A sample of uranium stearate, enriched in $^{235}$U, is placed in a neutron beam external to the reactor, where the uranium nuclei fission after absorbing thermal neutrons. The asymmetric mass yield of thermal neutron induced fission of $^{235}$U has peaks near mass numbers 90 and 139 for proton numbers 36 (Kr) and 54 (Xe) respectively (Fig. 4). Since the fission yield drops rapidly on either side of these peaks, the isotopes available for study from TRISTAN are limited to the fission products near these peaks and their daughters.

The noble gas fission products, which are efficiently emanated from the stearate sample, along with small amounts of bromine and iodine, are carried through a transport tube 1.7 meters long to the ion source. A sweep gas composed mainly of helium, with small amounts of stable krypton and xenon, is introduced into the sample chamber to help decrease the transport time. This gas also helps maintain a discharge in the ion source, and is later used to assist in monitoring the focus of the isotope beam.
Figure 3. Schematic layout of the TRISTAN isotope separator facility
Figure 4. Fission yield of $^{235}U$ as a function of mass number and proton number, with mass yield and element yield profiles
Both the stable sweep gas and the fission products are ionized, then accelerated through a potential of 50 kV. The resulting ion beam is sent through two electrostatic lenses for focusing before being introduced into the 90° analyzing magnet of the separator.

The mass-separated beams then arrive at a collector box, where the stable isotopes of the sweep gas facilitate visual focusing of the ion beams and the selection of the mass to be studied. This particular mass is positioned in the center of a slit aligned with the input to a switching magnet. Mass stabilization is achieved by positioning a pair of parallel copper strips such that a mass other than the one being studied is centered between the strips. These stabilization pins are connected to the inputs of a differential amplifier/integrator system which keeps the beam centered between the pins. Holding this beam in a fixed position then maintains the position of the ion beam being studied in the slit leading to the switching magnet.

At the switching magnet, the selected ion beam is directed to the experimental station being used, as well as being further cleansed of neutral or contamination components. For the measurements reported in this study, the beam was directed to the moving tape collector (MTC).

The MTC (Fig. 5) holds a reel of aluminized mylar tape on which the beam is deposited, plus a drive mechanism to
Figure 5. The moving tape collector used for short lifetime studies at TRISTAN
position the tape relative to the detectors being used. Detectors can be positioned on either side of the MTC at upper ports (on either side of the point where the beam is deposited), or at lower ports, farther along the line of motion of the tape. The upper ports are used to study parent activities of a given mass chain, or very short-lived daughter activities. The lower port provides for the study of longer-lived daughter activities. Once a sample has been collected, the accumulation of data at the lower ports can be delayed for an appropriate length of time while a new sample is being collected at the upper port position.

The MTC can be operated in a continuous mode, to carry away daughter activities while studying a short-lived parent. It can also be stepped, to move the collected sample to the lower port after a delay determined to maximize a certain daughter activity. The selection of the times involved in beam collection, tape motion, and data accumulation determine the degree to which the detection of one member of a decay chain is enhanced over the others. A complete description of the parameters involved and a program used to calculate these for maximum isobaric separation are contained in a report by Norman, Talbert, and Roberts (16).
B. Electronics

The detectors used in the lifetime measurements were a plastic scintillator and a thin-window, small volume planar Ge(Li) detector. The scintillator was a 25 mm by 44 mm cylinder of NE102 plastic mounted on an Amperex PM2106 twelve-stage photomultiplier tube, operated at 2000 volts. The output taken at the anode provided a very fast-risetime start signal (less than 5 nsec) for the timing coincidence circuit.

The stop signal was obtained from an ORTEC Low Energy Photon Spectrometer (LEPS). This is a 1-cm$^3$ volume Ge(Li) detector with a depletion depth of 5.1 mm and operated at a bias voltage of -1020 V. The preamplifier is an ORTEC Model 117A which was modified to provide separate timing and energy signals.

Both the Ge(Li) preamplifier timing signal and the scintillator timing signal were amplified by ORTEC 454 Timing Filter Amplifiers (see Fig. 6). The outputs were then fed into ORTEC 463 Constant Fraction Discriminator timing units, which provided fast logic pulse signals.

The start pulse (plastic) and the stop pulse (Ge(Li)) were connected, with a variable delay (0 - 63 nsec) in the stop side, to the inputs of an ORTEC Model 437 Time-to-pulse-height Converter (TPHC). The time interval registered by the TPHC is a measure of the delay between the formation
TFA: Timing Filter Amplifier
CPD: Constant Fraction Discriminator
SCA: Single Channel Analyzer
TPHC: Time-to-pulse-height Converter
LEPS: Low Energy Photon Spectrometer

Figure 6. Block diagram of the circuitry used in the lifetime measurement system
of a nuclear state, as indicated by the detection of a beta or gamma ray in the plastic scintillator, and the decay of the state by emission of the gamma ray seen by the LEPS. One output of the TPHC then became the input to ADC A of the dual-parameter data analysis system.

The second output of the Ge(Li) preamplifier was fed to both a linear delay amplifier and an ORTEC 460 Delay Line Amplifier. In one branch the signal went through the linear delay amplifier, then to a spectroscopy amplifier, either a Tennelec TC203BLR or an ORTEC 452. This linear energy signal was connected to ADC B of the dual-parameter system.

The delay line amplifier signal was connected to an ORTEC 458 Pulse Shape Analyzer (PSA). This unit measures the rise time of the input pulses it receives, and gives a slow logic pulse out if the rise time is within preset adjustable limits. The PSA was used to eliminate long risetime pulses from the Ge(Li) detector which caused excessive tailing on the late side of the timing peak. A more detailed discussion of this problem will follow the description of the circuit. Figure 7(a) shows a risetime distribution of pulses from the LEPS using a $^{226}$Ra source to simulate an experimental run with various energy pulses being present. Figure 7(b) shows those risetimes which the PSA was normally set to accept, with the cutoff here being at approximately 70 nsec. The method used to determine a nu-
Figure 7. Charge collection risetime distributions for the LEPS. a) Normal distribution for a 224Ra source; b) fraction of the risetimes normally accepted during runs.
The second output of the TPHC was connected to a timing single channel analyzer, adjusted to provide a slow logic pulse for every start-stop pair in the TPHC. This logic pulse and that from the PSA were both connected to a Canberra slow coincidence module. The output of the coincidence unit was used to gate both ADC's of the dual parameter system. The two delay amplifiers shown in Fig. 6 were adjusted so that the gating signal from the slow coincidence unit and the two inputs to the ADC's had the proper time relationship to be processed.

The dual-input data analysis system consists of two ADC's (either 8192-channel Geoscience or 4096-channel TMC), a format selector, a buffer tape unit, and a 16,384-channel analyzer.

The buffer tape unit records the memory addresses of every pair of coincidence events processed by ADC's A and B. For this study, a pair of data points consisted of a channel number corresponding to the energy of the gamma ray detected by the Ge(Li) detector, and a channel number indicating the time interval registered by the TPHC. If these pairs of points are considered to form an energy-time plane, and the number of counts at each point forms a third axis, then the data stored on the buffer tape can be represented as shown in Fig. 8.
Figure 8. Energy-time coincidence data stored by the buffer tape system for the decay of $^{140}$Xe
After the actual experimental run, the buffer tape can be played back into the analyzer memory through the format selector. This is designed so that gates may be set around energy peaks in the gamma spectrum of the isotope being studied. The buffer tape data is then sorted into the delayed coincidence time spectra corresponding to the energy of the gamma ray de-exciting a given level. This procedure will be described in more detail later. With the buffer tape system, the data for measuring the lifetimes of all the levels of an isotope can be taken at once, then separated into individual timing spectra later. This system saves a great deal of time since it makes it unnecessary to use a single channel analyzer gate to study individual transitions.

Time calibration for early experimental runs in this study was taken from a set of 50 ohm coaxial cables, used to insert delay in the stop channel of the system. The delay difference between the two cables was measured at first by using a Quadrupole Nanosecond Delay Box. Later an ORTEC 462 Time Calibrator was purchased and the difference in delays was measured with this unit. Whereas the cable difference was measured to be 10.0 nsec with the delay box, the more accurate time calibration resulted in a value of 9.4 nsec. All calibrations for the later runs were taken directly from the time calibrator.
C. Ge(Li) Detector Risetime Compensation

The use of Ge(Li) detectors for lifetime studies presents a problem not encountered with other detectors. The pulse shapes obtained from a Ge(Li) detector vary in shape much more than those of scintillation detectors.

Normally, there is a certain amount of time walk associated with pulses having different amplitudes. If the triggering of a discriminator level is used to indicate the arrival of a pulse, those pulses with larger amplitudes will reach this level in a shorter time than those with smaller amplitudes. A more significant pulse shape dependent effect is due to the fact that Ge(Li) pulses also have a spread in their risetimes.

Goulding (17) and Strauss, Larsen, and Sifter (18) have shown that the Ge(Li) pulse shape depends on the position within the depletion region of the detector where ionization from the gamma ray occurs, and also on whether the gamma ray is absorbed in a single interaction or multiple interactions. Even when the timing discriminator is set as low as possible, which should minimize walk effects, timing spectra taken with Ge(Li) detectors often have noticeable tailing on the late side of the time peak. The tailing can be reduced by choosing a suitable time-pickoff device.

Of the several methods available to signal the occurrence of an event in a Ge(Li) detector (19), the ARC
(amplitude and risetime compensated) method was used. Although the ARC technique is effective for true linear risetime pulses, Bengtson and Moszynski (20) have pointed out that it is less effective in compensating for pulses which change slope in time as the ion pairs in the detector enter different parts of the depletion region.

Thus, to further reduce the effect of long risetime pulses, the Pulse Shape Analyzer was used. As mentioned above, this unit selects pulses having risetimes within given limits. Only these acceptable pulses are then processed. Figure 9 shows the effect of the PSA on a prompt transition time spectrum, where values of the slope of the late side of the timing peak (converted to half-lives), the full-width at half-maximum (FWHM), and the full-width at tenth-maximum (FWTM) are plotted as a function of the fraction of the pulses accepted by the PSA. The fraction was varied by eliminating more of the longer risetime pulses and not changing the lower limit. The coincident 511-keV annihilation photons of $^{22}$Na were used to take this data. As more of the long risetime pulses are rejected, the slope and the FWHM both improve noticeably.

The risetime distributions of events in the LEPS have very different shapes as the energy of the incident gamma ray varies. Figure 10 shows the risetime distribution for a $^{22}$Na source and for a $^{57}$Co source. These distributions were
Figure 9. FWHM, FWTM and slope of the late side of the time spectrum for a $^{22}$Na source as a function of the fraction of pulses accepted by the PSA.
Figure 10. LEPS risetime distribution for a $^{22}$Na source (top) and for a $^{67}$Co source (bottom)
calibrated in time by using a BNC Model GL-3 pulser to closely approximate the shape of the pulses from the Ge(Li) preamplifier, then feeding the pulser output into the delay line amplifier. The risetime of the pulser output was varied from .05 μsec to 2.0 μsec in five steps, giving a good calibration for the entire range of the risetime distribution. The numerical values of the risetimes obtained from this calibration technique agree well with those measured using a more elaborate risetime spectrometer (21).

These changes in the risetime distribution are reflected in the timing spectra. As the energy of the gamma rays decreases the FWHM and the slope of the late side are significantly degraded. Figure 11 shows the variation of these two quantities as a function of energy. Both lines were taken from data accumulated during experimental runs with sources of $^{140}$Xe and $^{141}$Xe, both with the PSA set to accept the same range of pulse shapes.
Figure 11. FWHM and slope of the late side of the time spectrum as a function of energy for a $^{22}$Na source
III. DATA ANALYSIS

A. Time Spectra

Each data collection run usually produced several buffer tapes, described previously. In describing the data analysis, it is best to consider each event recorded on the buffer tape as having coordinates \((E,t)\), where both energy and time are given in units of channel number.

Several factors entered in the determination of the range of energies studied in each run. First, the LEPS becomes very inefficient at detecting gamma rays with energies above approximately 300 keV. Second, consideration was given to the decay scheme of the nucleus studied; or if this was not available, a list of gamma rays emitted by the nucleus was checked. If there was a gap in the distribution of gamma rays between, for example, 200 and 300 keV, the energy range was set to stop at 200 keV.

For any given run, the third and most limiting factor was the pulse shape analyzer. The input discriminator on this unit has a dual range of 0.1 V and 10 V. Using the 0.1-V range, energy pulses of approximately 10 keV to 120 keV would be passed by the unit. On the higher range, energies of about 40 keV to 250 keV were accepted. These limits depended on the gain of the delay line amplifier, which was set as high as possible without causing excessive saturation. If there are any very low energy peaks in the
spectrum, for example the 13.8-keV gamma ray in the decay of $^{136}$Xe, a high gain is necessary so that the discriminator can be set above the noise level, yet not be set above the peak.

During all initial runs, the TPHC range was set at 200 nsec, covering a 512-channel spectrum. If a very long-lived level was seen, such as in the case of $^{136}$Xe, another run was performed using a longer time range.

The first step in analyzing the data was to plot an energy profile, an example of which is shown in Fig. 12 for the decay of $^{136}$Xe. This spectrum contains all the energy peaks that were involved in the coincidences recorded on the buffer tape. In the profile, the counts in channel 100, for example, are those events having energy coordinate 100, and any time coordinate.

Using the profile as a guide, bands are set around the energy peaks of interest as shown in Fig. 12. The channel numbers giving the upper and lower limits of the bands are programmed into the format selector. When the buffer tapes are played back, the format selector then sorts the coincidence events into time spectra and stores them in the analyzer memory. If CH1 and CH2 are the limits of one band, the resulting time spectrum would consist of events having coordinates (CH1,t) to (CH2,t) for each value of t from channel 1 to channel 512. The four time spectra resulting
Figure 12. Energy profile of the decay of $^{136}$Xe showing locations of four energy peak bands (E1 to E4) and their respective background bands (B1 to B4)
from the energy peak band settings $E_1 - E_4$ indicated in Fig. 12 are shown in Fig. 13.

The number of bands which could be run at one time using the format selector method was limited to 16, when each of the resulting spectra was 1024 channels long, because of the 16K capacity of the analyzer. For some of the later runs, these bands were run using a computer program code BUFPTAP. Using this program with an IBM 360 computer, more than 16 bands could be run at once because of the larger memory available. The resulting time spectra are recorded on magnetic tape for retrieval in the next step of the analysis.

Each of these spectra has contributions from the Compton distribution of gamma rays higher in energy and from beta rays detected in the Ge(Li) detector. To eliminate these counts, a background gate was set at an energy just above that of the peak being studied, as shown in Fig. 12. The width of a background gate was chosen so that the area of the Compton background under the peak would equal the area in the background region selected by the gate. In calculating these areas, it was assumed that the Compton background could be approximated by a straight line over the small energy ranges of the bands. If another spectrum peak interfered with the placement of the background gate, this gate was divided. Part of it was put at a higher energy
Figure 13. $^{140}$Xe delayed coincidence time spectra from the energy peak gates shown in Fig. 12.
than the peak of interest, and the rest of the band was set at an energy just below the peak. The background spectra were then subtracted channel by channel from their respective peak spectra.

B. Delayed Coincidence Spectra Analysis

1. **Slope method**

The next step is to determine exactly what the time spectra represent. If a number of nuclei are formed in a certain excited state at time \( t = 0 \), the probability that this excited level of a given nucleus has not decayed \((22)\) before time \( t \) is \( e^{-\lambda t} \). The probability that the excited level of any particular nucleus will decay at any given time is \( \lambda \). Thus the probability that some level will decay at time \( t \), having been formed at time \( t = 0 \), is

\[
f(t) = \lambda e^{-\lambda t}
\]

and the mean life \( \tau = \lambda^{-1} \).

The mean life of a transition is found by measuring the decay probability as a function of time. To do this, early experimentalists would set up a coincidence system with a variable delay in one side of the circuit. With one detector producing pulses from the event that populated the isomeric level, and another detector registering the events which de-excited the level, the pulse outputs of the coincidence unit were counted at a function of delay. Hence
the common name given to the method — delayed coincidence measurements. According to the above probability distribution, the slope of a semi-log plot of the number of counts per unit time vs. delay added to the circuit would give the transition probability or mean life.

The variable delay technique has given way to multichannel analyzer time scaling and time-to-pulse-height converters for measuring shorter lifetimes. With the shorter lifetimes comes the problem of time jitter in the measuring system, and the ways that it affects the data.

If the system had no time jitter, a measured decay curve would look like Fig. 14(a). However, because of the finite response time of the system, such a curve usually looks like that labelled $f(t)$ in Fig. 14(b). This figure is typical of a system using NaI detectors, and is also included for comparison to a curve from the system in use here. In analyzing these curves, it is important to know when the presence of time jitter or other curve degrading effects will interfere with the measurement of the slope. To decide this, one must consider, along with $f(t)$ given above, the response of the system to a prompt transition, for which there is essentially zero delay between the formation and the decay of a nuclear state. The prompt curve, $P(t)$ (see Fig. 14(b)), is then the time distribution caused by the inherent time jitter and response of the system.
Figure 14. Delayed coincidence curves. a) Ideal curve with no time jitter present; b) prompt and delayed curves typical of a NaI-NaI system.
The response of the system (23) to a delayed transition can be written as

\[ F(t) = \int f(t')P(t-t')dt' . \]

\(F(t)\) is interpreted as the probability that a transition from the excited state will be detected at time \(t\). \(P(t-t')\) must be included because a gamma ray which is emitted at time \(t'\) may be detected at time \(t\), due to the time resolution of the system. Since \(f(t')\) is zero for \(t' < 0\), the integral range is from 0 to \(\infty\). Letting \(y = t-t'\), \(F(t)\) becomes

\[ F(t) = \lambda e^{\lambda y} P(y)dy . \]

Differentiating with respect to \(t\) and simplifying gives

\[ \frac{dF}{dt} = -\lambda F(t) + \lambda P(t) \]

or

\[ \frac{1}{F} \frac{dF}{dt} = -\lambda \left[ 1 - \frac{P(t)}{F(t)} \right] . \]

Finally this becomes

\[ \frac{d}{dt}(\ln F) = -\lambda \left[ 1 - \frac{P(t)}{F(t)} \right] . \]

What this means is that the slope of the measured distribution (\(\ln F(t)\)) will give directly the inverse of the mean life of the transition, if it is measured in a region of the curve where \(P(t) \ll F(t)\). As long as the prompt response falls off rapidly with time, the slope can be measured in a region where \(F(t) \gg P(t)\). An example of a fit
using the slope method is shown in Fig. 15. This spectrum represents the decay of the 14.4-keV level of $^{57}$Fe. With a time calibration of .75 nsec/channel, the half-life according to the linear least squares fit is 99.1 nsec, agreeing very well with the previously measured value of 98 nsec (9).

In this study, the slope method was used only once for the case of a very long-lived isomeric level. The reason that the slope could not be measured accurately for the other cases was due to the shape of the prompt transition distribution, $P(t)$, for the Ge(Li) - plastic system. Figure 16 shows both a prompt and a delayed transition from the decay of $^{131}$Xe at an energy of 106 keV. The tailing on the late side of the time spectrum in most cases precluded the direct measurement of the mean life. The tailing was reduced by using the pulse shape analyzer, but near the lower limit of lifetimes for the system, even this was insufficient for accurate slope determination. Because of this, a fitting technique was developed to determine mean lives which took advantage of the data made available by the buffer tape system. As will be shown below, the system was used to provide the prompt spectrum, $P(t-t')$, so that the integral in the expression for $F(t)$ could be evaluated.

2. Non-linear least squares fitting method

The first problem in developing a fitting program was to find a function, $F(t)$, which would reproduce the experi-
Figure 15. Time spectrum for the decay of the 14.4-keV level of $^{57}$Fe showing the straight line fitted to the data
Figure 16. Prompt and delayed transition curves for the Ge(Li)-plastic system, taken from the decay of the 106-keV level in $^{137}$Cs.
mentally observed time spectra. The logical starting place was

\[ F(t) = C e^{-\lambda t'} P(t-t') dt' \]

where \( C \) is a normalization constant. Rather than subtract the chance coincidence background from the time spectra, this was put into \( F(t) \) as \((D*t + E)\), with both \( D \) and \( E \) as fit parameters. Also, it was found that subtracting a background time spectrum did not always remove the entire prompt contribution from the time spectrum being studied. To account for this another term, \( G*P(t) \), was added to \( F(t) \), with \( G \) to be determined in the fit.

The function \( F(t) \) now has five parameters to be fit: \( \lambda \), the inverse of the mean life; \( C \), a normalization constant; \( D \) and \( E \), the parameters describing the chance coincidence background, assumed linear in time; and \( G \), the residual contribution from the prompt transitions. Thus,

\[ F(t) = C e^{-\lambda t'} P(t-t') dt' + D*t + E + G*P(t) . \]

The problem with this function was \( P(t-t') \). Whereas the curve from a NaI system is approximately Gaussian, no functional expression could be found which described the prompt response of this system. It was also noted that this response varied significantly with energy. Since the buffer tape system stores all coincidence events that occur during a run, a prompt transition for any energy could be obtained
by setting a band on the Compton background between energy peaks. Thus the spectrum \( P(t) \) for each fit was taken to be an experimentally measured prompt spectrum.

Since the prompt distribution, \( P(t) \), and the time spectrum to be fit, \( F(t) \), were both arrays of discrete numbers, as opposed to a continuous function, the integral in the fit function was changed to the form:

\[
F(t_i) = C \sum_{j=0}^{i} e^{-\lambda t_{jp}} (t_i - t_j) + D t_i + E + G P(t_i).
\]

This effectively approximates the integration by using a sum of rectangular areas, since the data points are uniformly spaced in time. By letting \( t_i - t_j = t_k \) and relabelling, the first term becomes

\[
C \sum_{j=1}^{i} e^{-\lambda (t_i - t_j)} P(t_j)
\]

and this form was used in the fitting program. Since the measured spectrum \( P(t_j) \) is taken to start in channel 1, the sum starts at \( j=1 \).

The non-linear least squares program was set up as an iterative routine to minimize \( \chi^2 \), where

\[
\chi^2 = \sum_{i} \frac{w_i [F(t_i) - y_i]^2}{\sum_{i} w_i}
\]

In this expression, \( F(t_i) \) is calculated from the function for each point, and \( y_i \) is the corresponding data point in the time spectrum being fit. \( W_i \) is the statistical weight
of the data point $y_i$, taken to be
\[ w_i = \frac{1}{\sigma_i^2} = \frac{1}{N_d + N_b}. \]

$N_d$ is the number of counts in the measured delayed time spectrum before subtracting the background spectrum, and $N_b$ is the number of counts in this background spectrum.

Minimizing $\chi^2$ by setting
\[ \frac{\partial}{\partial q_i} \chi^2 = 0 \]
for each of the fit parameters $q_i$ gives a system of five equations in five unknowns. Because these equations are non-linear, they cannot in general be solved exactly. Therefore a variational technique has to be used, specifically, Newton's Method (24).

Let the five fit parameters be represented by $q_i$, $i=1$ to 5 and let $\delta_i$ be the corresponding small incremental changes in $q_i$. If initial values are chosen for $q_i$, the fit function can be expanded about the initial point as
\[ F(q_i + \delta_i) = F(q_i) + \sum_k \frac{\partial F}{\partial q_k} \delta_k + O(\delta^2) + \ldots \]

Using this expression in $\chi^2$, and setting the derivatives with respect to $q_i$ equal to zero, results in the five equations:
\[ w_i [y_i - F(q_i)] \frac{\partial F}{\partial q_k} = w_i \sum_k \frac{\partial F}{\partial q_k} \frac{\partial F}{\partial q_k} \delta_k \]

These equations are now linear in $\delta_i$ and can be solved for
these five parameters. However, since higher order terms have been dropped in forming these equations, the solution is only approximate, and an iterative procedure is required to find the values of $q_i$ that are needed.

At the start of the iteration process, initial values of $q_i$ are supplied. Initially, $\lambda$ is taken from a straight-line estimate of the slope of the experimental spectrum. Both $D$ and $G$ usually begin at zero. $E$ is also estimated from a graph of the data. The initial value of $C$ is calculated during the first iteration by taking a ratio of the maximum calculated point in the spectrum to the maximum point in the data. Using these values the five equations are solved by matrix inversion for the values of $\delta_i$. The five parameters $q_i$ are then changed to $(q_i + \delta_i)$ and the process of setting up the equations and solving them is repeated. This procedure continues until the fractional change in each parameter $q_i$ is less than a specified convergence limit, usually set at 0.001.

Using the time calibration determined for the run, the program then lists values of the decay constant, the mean life and the half-life. The program also produces a computer plot of the input time spectrum with the calculated fit function $F(t)$ superimposed for a visual check of the fit.

An example of the results of this fitting program can be seen in Fig. 17. Using a $^{133}$Ba source, this time
Figure 17. Delayed coincidence time spectrum and fitted function for the decay of the 81-keV level of $^{133}$Cs
spectrum represents the decay of the 81-keV level in $^{133}$Cs. The program converged to a value of 0.112/nsec for the transition probability, or a half-life of 6.2 nsec. This compares well with the accepted value of 6.3 nsec (9).

The uncertainties given with the measured lifetimes come from two sources. The statistical uncertainty of the least-squares fit is taken from the diagonal element of the error matrix calculated during the fitting procedure.

The second contribution to the uncertainty comes from the fact that the choice of the prompt spectrum has a direct effect on the value of the mean life found by the fitting program. To estimate this effect on the half-lives, time spectra were fit using prompt spectra from energies both higher and lower than the energy corresponding to the peak being fit.

For each transition that had a measurable half-life reported in this work, the time spectrum was fitted using prompt spectra taken from energies approximately 15 keV away, both above and below, from the transition energy being studied. Using the values of the half-life for a given transition obtained this way, plus the original value that was calculated, the effect of the prompt spectrum choice on the half-life value could be estimated over an energy range of about 30 keV.
Since the actual prompt spectrum for each fit reported here was taken from approximately 5 keV above the transition energy peak, a correction was then made to the half-life value on the basis of the above estimate. For the worse case, the 69-keV transition in $^{131}$Cs, this amounted to a reduction of the half-life value by 0.3 nsec. For the other reported half-lives, the corrections were less than this.

Thus the half-lives listed in the following tables are the values found by the fitting program, corrected for an apparent increase in half-life caused by the choice of the prompt spectrum. The associated uncertainties listed are the statistical uncertainties given by the error matrix.
IV. EXPERIMENTAL RESULTS

The results presented here are divided into three sections, one each for the levels of $^{131}\text{Cs}$, $^{137}\text{Cs}$ and $^{136}\text{Xe}$. In each case, the energy levels and transitions studied are listed, along with the values of the half-lives that could be measured within the range of the equipment. Upper limits for the half-lives of the other levels studied are also given, as explained below.

In those cases where the mean life of a transition could not be determined, it was because the time spectrum of the transition under consideration had the same shape as the time spectrum of a prompt transition with the same energy; i.e., the half-life was shorter than the equipment could measure. Since the response of the system to a prompt transition at any energy can be found using the procedure described in Section IIIB, these short half-life transitions must have a mean life less than or equal to the inverse of the measured slope of the time distribution.

Since the tailing of the time spectra discussed previously makes the apparent slopes less than they would be with no tailing effects, the mean-life limits have been taken to be less than the inverse of the slopes, not equal to them. The slopes calculated for the upper limits were found using the method described in Section III, part B1. The results are reported as half-lives in the following tables.
A. Half-lives of $^{134}$Cs Levels

The energy profile of the decay of $^{134}$Xe is shown in Fig. 18. For this run the MTC was operated in a mode to enhance the decay of $^{134}$Xe, but two strong peaks from the Cs decay and two from the Ba decay are also present in the energy spectrum. For reference to the energy levels involved in the transitions, the low-energy part of the $^{134}$Cs decay scheme is presented in Fig. 19. The gamma-ray energies and the decay scheme are from a study by Cook and Talbert (25), and a compilation by Auble (26).

Of the transitions studied, the 68.99-keV level was found to have a half-life of $23.3 \pm 0.7$ nsec. The delayed coincidence time spectrum for the 68.99-keV ground-state transition, along with the fitted function, is shown in Fig. 20. The time calibration for all $^{134}$Cs transition curves is $0.954$ nsec/channel.

The 105.87-keV ground-state transition half-life was measured as $8.7 \pm 0.2$ nsec. The time distribution for this transition and the fitted function are shown in Fig. 21.

Table 1 summarizes these values along with the upper limits of the half-lives for the other transitions studied. Note that two transitions from the decay of $^{134}$Ba levels are also included in the table. The corresponding levels for these two Ba peaks are not known at this time. Comments on the values of the half-lives of these levels will be made in
Figure 18. Energy profile for the decay of $^{141}$Ce

SQ. RT. COUNTS

Cesium and Barium X-rays

- Cs, Ba X-rays
- 48.72 (Cs)
- 55.42 (Cs)
- 68.99 Pb X-rays
- 81.75
- 89.74
- 100.68
- 112.94 (Ba)
- 105.87
- 118.65
- 137.60
- 187.58
- 190.22 (Ba)
Figure 19. Low energy decay scheme for $^{141}$Cs

$^{141}$Cs  $^{55}$Cs  $^{86}$

68.99  105.87  137.60  187.58  118.65  81.75

00  06  09  18  207

492.84  423.51  386.87  304.88  285.92  467.93  398.90  361.98

459.27  89.74  556.82  369.40  492.84  386.87  304.88  285.92

557.  468.  493.  467.93  398.90  361.98  285.92  187.58

00  06  09  18  207
Figure 20. Delayed coincidence time spectrum and fitted function for the decay of the 69-keV level of $^{131}$Cs
Figure 21. Delayed coincidence time spectrum and fitted function for the decay of the 106-keV level of $^{137}$Cs
Table 1. Half-lives of levels in $^{141}$Cs and $^{141}$Ba

<table>
<thead>
<tr>
<th>Transition Energy (keV)</th>
<th>Initial Level (keV)</th>
<th>Half-life (nsec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>48.72$^a$</td>
<td></td>
<td>&lt; 3.4</td>
</tr>
<tr>
<td>55.42$^a$</td>
<td></td>
<td>&lt; 3.2</td>
</tr>
<tr>
<td>68.99</td>
<td>69</td>
<td>23.3 ± 0.7</td>
</tr>
<tr>
<td>81.75</td>
<td>188</td>
<td>&lt; 2.4</td>
</tr>
<tr>
<td>89.74</td>
<td>647</td>
<td>&lt; 2.4</td>
</tr>
<tr>
<td>100.68</td>
<td>207</td>
<td>&lt; 2.3</td>
</tr>
<tr>
<td>105.87</td>
<td>106</td>
<td>8.7 ± 0.2</td>
</tr>
<tr>
<td>118.65</td>
<td>188</td>
<td>&lt; 2.1</td>
</tr>
<tr>
<td>137.60</td>
<td>207</td>
<td>&lt; 2.1</td>
</tr>
<tr>
<td>187.58</td>
<td>188</td>
<td>&lt; 1.9</td>
</tr>
</tbody>
</table>

$^a$ Transitions in $^{141}$Ba

In the following section, where the possible multipolarities of the transitions will be considered.
B. Half-lives of $^{134}\text{Cs}$ Levels

Twelve transitions in $^{134}\text{Cs}$ were intense enough to use for lifetime measurements. The energy profile for the decay of $^{134}\text{Xe}$ is shown in Fig. 22. Again the low-energy part of the decay scheme for $^{134}\text{Cs}$, taken from a study by Schick and Talbert (27), is reproduced in Fig. 23 for reference.

The level with the longest half-life measured in this work is the 14-keV level of $^{134}\text{Cs}$. The slope method was used here to obtain a value for the half-life of $471 \pm 51$ nsec. Figure 24 shows the time spectrum for this transition along with the straight line fit to the data. This spectrum and the others for the $^{134}\text{Cs}$ transitions have a time calibration of 0.468 nsec/channel.

The decay of the 65-keV level via the 50.6-keV gamma ray has a measured half-life of $3.7 \pm 0.3$ nsec. The time spectrum for this transition and the fitted function are shown in Fig. 25.

The 89.1-keV transition from the 103-keV level has a half-life which was measured as $7.0 \pm 0.4$ nsec. The time distribution for this transition and the fitted function are shown in Fig. 26. A second transition from the 103-keV level makes it possible to compare two measured values of the half-life for the decay of this level. The delayed coincidence time spectrum for the 103.0-keV ground-state transition and the fitted function are shown in Fig. 27.
Figure 22. Energy profile for the decay of $^{130}\text{Xe}$
Figure 23. Low energy decay scheme for 140Cs

140Cs
55Cs 85

13.8
50.6
80.1
103.0
89.1
112.5
118.5
104.4
84.5
212.0
198.1
147.3
109.0
99.5
93.6
158.7
218.3
167.3
128.7
119.0

00
14
65
80
112
149
222
1252
Figure 24. Delayed coincidence time spectrum and straight line fit for the decay of the 14-keV level of $^{140}$Cs
Figure 25. Delayed coincidence time spectrum and fitted function for the decay of the 51-keV level of $^{137}\text{Cs}$
Figure 26. Delayed coincidence time spectrum and fitted function for the decay of the 89-keV level of $^{137}$Cs
Figure 27. Delayed coincidence time spectrum and fitted function for the decay of the 103-keV level of $^{140}$Cs.
The half-life of this transition is $7.5 \pm 0.3$ nsec.

These results along with upper limits for eight other transitions are listed in Table 2. A discussion of these results will follow in the next section.

Table 2. Half-lives of levels in $^{140}\text{Cs}$

<table>
<thead>
<tr>
<th>Transition Energy (keV)</th>
<th>Initial Level (keV)</th>
<th>Half-life (nsec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>13.8</td>
<td>14</td>
<td>471 ± 51</td>
</tr>
<tr>
<td>50.6</td>
<td>65</td>
<td>3.6 ± 0.3</td>
</tr>
<tr>
<td>80.11</td>
<td>80</td>
<td>&lt; 2.7</td>
</tr>
<tr>
<td>84.5</td>
<td>149</td>
<td>&lt; 2.7</td>
</tr>
<tr>
<td>89.1</td>
<td>103</td>
<td>7.0 ± 0.4</td>
</tr>
<tr>
<td>93.6</td>
<td>212</td>
<td>&lt; 2.6</td>
</tr>
<tr>
<td>103.0</td>
<td>103</td>
<td>7.5 ± 0.3</td>
</tr>
<tr>
<td>104.4</td>
<td>118</td>
<td>&lt; 2.4</td>
</tr>
<tr>
<td>112.52</td>
<td>112</td>
<td>&lt; 2.3</td>
</tr>
<tr>
<td>118.46</td>
<td>118</td>
<td>&lt; 2.3</td>
</tr>
<tr>
<td>167.26</td>
<td>232</td>
<td>&lt; 2.0</td>
</tr>
<tr>
<td>212.00</td>
<td>212</td>
<td>&lt; 1.9</td>
</tr>
</tbody>
</table>
C. Half-lives of $^{136}$Xe Levels

As mentioned previously, the amount of iodine available from TRISTAN is much less than the amounts of the noble gas fission products. Hence the energy profile for the decay of $^{136}$I (Fig. 28) has noticeably fewer counts than the other profiles. Following the profile, bands were set on eight peaks. Of these, the 164.5-keV and the 291.6-keV transitions may not belong to the $^{136}$Xe level scheme, the low energy part of which is reproduced in Fig. 29. The transition energies and the preliminary decay scheme are taken from a study now in progress by Western (28). Although there are two distinct activity half-lives in the decay of $^{136}$I, all transitions in $^{136}$Xe have been put into the same level scheme here.

None of the eight transitions had a half-life long enough to be measured. However, upper limits have been set for each. The results of the linear least squares fit to the slopes are presented in Table 3.

The 197.6-, 291.6- and 345.1-keV transition half-life limits that are listed might be due to a residual prompt component in the time spectrum. Because of the very small numbers of counts in these time spectra after background subtraction, it was implausible to determine any slope extending far from the position of the prompt peak. In one case, the 197.6-keV transition, the existence of a very long
Figure 28. Energy profile for the decay of $^{136}\text{I}$
Figure 29. Low energy decay scheme for $^{136}\text{Xe}$
Table 3. Half-lives of levels in $^{136}$Xe

<table>
<thead>
<tr>
<th>Transition Energy (keV)</th>
<th>Initial Level (keV)</th>
<th>Half-life (nsec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>164.5</td>
<td></td>
<td>&lt; 2.1</td>
</tr>
<tr>
<td>182.7</td>
<td>2444</td>
<td>&lt; 2.0</td>
</tr>
<tr>
<td>197.6$^a$</td>
<td>1892</td>
<td>&lt; 1.9</td>
</tr>
<tr>
<td>291.6$^a$</td>
<td></td>
<td>&lt; 1.7</td>
</tr>
<tr>
<td>345.1$^a$</td>
<td>2634</td>
<td>&lt; 1.6</td>
</tr>
<tr>
<td>347.2</td>
<td>2608</td>
<td>&lt; 1.6</td>
</tr>
<tr>
<td>370.0</td>
<td>2262</td>
<td>&lt; 1.4</td>
</tr>
<tr>
<td>381.2</td>
<td>1694</td>
<td>&lt; 1.4</td>
</tr>
</tbody>
</table>

$^a$Half-life limit given may be due to residual prompt spectrum as discussed in text.

Half-life has been reported by Carraz et al. (29), who measured the half-life as $2.8 \pm 0.2$ sec. Half-lives in this range are longer than would be detected with the relatively short time scales used in this study, and the values noted in Table 3 for these three transitions may not reflect the actual half-life limits.
V. DISCUSSION OF RESULTS

A. Weisskopf Estimates

The theoretical standards of reference for lifetime measurements are the single-particle Weisskopf estimates. The derivation of these estimates involves time-dependent perturbation theory, the second quantization of the electromagnetic field, and a number of simplifying assumptions which yield simple numeric expressions. A brief outline of this derivation is given below. More detailed discussions can be found elsewhere (13,30).

The transition probability per unit time for a state |i⟩ going to a state |f⟩ with the emission of a photon of energy ℏω, is given by (31):

\[ w_{fi} = \frac{2\pi}{\hbar} |\langle f|H'|i\rangle|^2 \frac{dn}{dE} \delta(\varepsilon_f + \hbar\omega - \varepsilon_i) \]

where \( H' \) is the perturbing interaction Hamiltonian, \( dn/dE \) is the number of possible final states per unit energy interval, and \( \varepsilon_i \) and \( \varepsilon_f \) are the energies of the initial and final states, respectively. This Hamiltonian is taken to be the scalar interaction between the free-photon field \( A_\mu \) and the nuclear charge four-vector, \( j_\mu \). Thus,

\[ H' = -\frac{i}{c} \overleftrightarrow{d^\tau} j_\mu A_\mu. \]

The free field is now quantized by letting

\[ \hat{A} = \sum_{k\lambda JM} \sqrt{\frac{4\pi\hbar\omega}{\mathcal{R}}} [a_{JMk\lambda} \hat{A}_{JM}(\lambda) + a^+_{JMk\lambda} \hat{A}^*_{JM}(\lambda)]. \]
where \( a \) and \( a^+ \) are the annihilation and creation operators for a single photon of energy \( \hbar \omega \), \( \hat{A}_{JM}(\lambda) \) are the solutions to Maxwell's equations for the vector potential, and \( \lambda \) is used to distinguish electric and magnetic operators.

After substitution for \( H' \) and with considerable simplification, integration over all possible photon states yields the well-known result for the total transition probability:

\[
T_{fi} = \frac{8\pi}{\hbar[(2J+1)!]^2} \frac{J+1}{J} k^{2J+1} B(\lambda J; J_f^+J_i^-)
\]

where

\[
B(\lambda J; J_f^+J_i^-) = \frac{1}{2J+1} |<J_f||O_J(\lambda)||J_i>|^2
\]

\( O_J(\lambda) \) represents the magnetic or electric multipole operator for a transition of multipolarity \( \lambda J \).

To arrive at the Weisskopf estimates, it is assumed that only one particle is involved in the transition, i.e., that \( |J_f> \) and \( |J_i> \) differ only in one single-particle orbital. Thus the reduced matrix element contains only the coordinates of one particle and the states can be represented by \( |l_j> \), representing a state with orbital angular momentum \( l \) coupled to a spin of \( 1/2 \) to give a total angular momentum \( j \). The result at this point is still very complicated, and is given in de-Shalit and Talmi (32). This result can be further simplified by requiring that the angular momentum carried away by the photon be its minimum value, \( J = |j_i-j_f| \).
The final assumption to be made is that the radial wave function is a constant throughout the interior \((r < a)\) of the nucleus, and vanishes outside \((r > a)\). This allows the radial integrals to be calculated easily.

Finally, with all of the above assumptions, we arrive at the single-particle estimates for the transition probability for electric \((EJ)\) and magnetic \((MJ)\) multipole transitions, with \(a\) in units of \(10^{-13}\) cm and \(E = \frac{\hbar \omega}{\text{MeV}}\) in MeV:

\[
T_{sp}(EJ, l_{f} j_{f}^{+} l_{i} j_{i}) = \frac{4.4(J+1)}{J(J+1)} \left(\frac{3}{J+3}\right)^2 \left(\frac{\hbar \omega}{197\text{MeV}}\right)^{2J+1} a^{2J} x S(j_{i}, J, j_{f}) \times 10^{21} \text{ sec}^{-1}
\]

\[
T_{sp}(MJ, l_{f} j_{f}^{+} l_{i} j_{i}) = \frac{0.19(J+1)}{J(J+1)} \left(\frac{3}{J+2}\right)^2 \left(\frac{g_{s}}{2 J - J + 1}\right)^{2J-2} a^{2J-2} x \left(\frac{\hbar \omega}{197\text{MeV}}\right)^{2J+1} S(j_{i}, J, j_{f}) \times 10^{21} \text{ sec}^{-1}
\]

\(S\) is a statistical factor involving the angular momenta of the initial and final states and the photon. It is usually of the order of unity, and for the results given below it is assumed that \(S = 1\).

Letting the nuclear radius \(a = 1.2 \times 10^{-13} A^{1/3}\) cm and the Lande factor \(g_{s} = 5.58\) results in the numerical values (33) of the transition half-lives, \(t_{1/2} = (\ln 2) / T_{sp}\), given in Table 4 for various multipolarities.

While used as references, the single-particle estimates for the transition probabilities do not always agree quantitatively with experimental results. However, some general
Table 4. Single-particle half-life estimates

<table>
<thead>
<tr>
<th>Multipolarity (λJ)</th>
<th>Half-life (sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>E1</td>
<td>$6.76 \times 10^{-3} E_{\gamma}^{-3a-2/3}$</td>
</tr>
<tr>
<td>M1</td>
<td>$2.20 \times 10^{-1} E_{\gamma}^{-3}$</td>
</tr>
<tr>
<td>E2</td>
<td>$9.52 \times 10^{-9} E_{\gamma}^{-5a-4/3}$</td>
</tr>
<tr>
<td>M2</td>
<td>$3.10 \times 10^{-8} E_{\gamma}^{-5a-2/3}$</td>
</tr>
<tr>
<td>E3</td>
<td>$2.05 \times 10^{-2} E_{\gamma}^{-7a-2}$</td>
</tr>
<tr>
<td>M3</td>
<td>$6.67 \times 10^{-2} E_{\gamma}^{-7a-4/3}$</td>
</tr>
<tr>
<td>E4</td>
<td>$6.50 \times 10^{4} E_{\gamma}^{-9a-8/3}$</td>
</tr>
<tr>
<td>M4</td>
<td>$2.12 \times 10^{5} E_{\gamma}^{-9a-2}$</td>
</tr>
</tbody>
</table>

trends have been noted for experimental data (34).

If a measured half-life is longer than the corresponding single particle estimate, the transition is said to be hindered by a factor equal to the ratio of the measured half-life to the estimate. Also, if the measured value is shorter, the transition is said to be enhanced by a factor equal to the ratio of the single-particle estimate to the measured half-life.

Most E1 transitions are slower than the Weisskopf estimate for the transition probability. Hindrance factors
range up to $10^4$ with a few even larger.

M1 transitions generally have hindrance factors near 100, and nearly all reported M1's lie between a hindrance factor of $10^4$ and an enhancement factor of 10.

E2 transitions are in general enhanced by a factor ranging from 10 to 300, although some hindered E2's are known. This enhancement is generally viewed for most cases as a collective effect (35).

The small number of M2 transitions measured have half-lives with hindrance factors, in general, ranging from 10 to 100. There are some which reach to a factor of $10^7$.

With a few exceptions, the half-lives of M4 transitions are very close to the single-particle estimate. The few cases of E4 and E5 transitions which are known have half-lives close to the Weisskopf estimates also.

B. Internal Conversion Effects

Regardless of the model used, in order to compare values of theoretical transition probabilities with those of the experiment, internal conversion corrections must be made to the experimental values obtained from the measured half-lives (6). The single-particle estimates give a half-life for gamma-ray emission only. When competing processes such as internal conversion occur, the partial half-lives must be combined using transition probabilities according to
\[ \lambda = \lambda_\gamma + \lambda_e \]

where \( \lambda \) is the total transition probability and \( \lambda_\gamma \) and \( \lambda_e \) are the transition probabilities for gamma-ray emission and internal conversion, respectively, which are in turn proportional to \( N_\gamma \), the number of gamma rays emitted per unit time from a given source, and \( N_e \), the number of internal conversion electrons emitted per unit time. Thus

\[ N = N_\gamma + N_e = N_\gamma (1 + \alpha) \]

where \( \alpha \) is defined as the internal conversion coefficient,

\[ \alpha = \frac{N_e}{N_\gamma} \]

Similarly, if a level depopulates by the emission of more than one gamma ray, the partial transition probabilities must be added together to get the total transition probability. If the relative intensities of the transitions are known, then the partial transition probabilities can be found, and these values compared to the theoretical predictions.

Experimentally, the measured half-life for any transition is that resulting from the total transition probability \( \lambda \). The value which should be compared to the Weisskopf estimates, however, is \( \lambda_\gamma \).

\[ \tau = \frac{1}{\lambda_\gamma (1 + \alpha)} \]

or
\[ T_\gamma = T(1 + \alpha). \]

Alternatively, the predicted values can be divided by 
\((1 + \alpha)\), where \(\alpha\) is the conversion coefficient for any given 
multipolarity. The latter procedure is used for comparison 
purposes in this work.

An example of the dramatic change which the inclusion 
of the internal conversion coefficients can make is seen in 
Fig. 30. The dashed lines are the single-particle shell 
model estimates for six classes of transitions, E1, M1, E2, 
M2, E3, and M3. When the theoretical internal conversion 
coefficients (36) are included for \(Z = 55\) the solid lines 
result. This clearly indicates the necessity for correcting 
the measured half-lives for the effects of internal conver­
sion, especially at low energies.

C. Comparison of Theory with Experimental Results

To facilitate comparison of experimental results to the 
single-particle predictions, the results are usually 
expressed in Weisskopf units, i.e., given as the ratio of 
the experimental value to the Weisskopf estimate for a given 
multipolarity. The six transition half-lives from \(^{141}\)Cs and 
\(^{140}\)Cs that were measured are given in Table 5 in Weisskopf 
units for multipolarities M1 and E2. If the M1 transition 
were assumed, the hindrance factor would be numerically 
equal to the half-life in Weisskopf units. For an assumed
Figure 30. Weisskopf estimates for $A=140$ (dashed lines), and estimates corrected for internal conversion for $Z=55$ (solid lines).
### Table 5. Measured half-lives in Weisskopf units

<table>
<thead>
<tr>
<th>Transition Energy (keV)</th>
<th>Nucleus</th>
<th>$T_{1/2}/T_{1/2}^{(W)}$ (E2)</th>
<th>$T_{1/2}/T_{1/2}^{(W)}$ (M1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>13.8</td>
<td>$^{138}$Cs</td>
<td>$1.67 \times 10^{-1}$</td>
<td>$2.47 \times 10^3$</td>
</tr>
<tr>
<td>50.6</td>
<td>$^{138}$Cs</td>
<td>$2.18 \times 10^{-3}$</td>
<td>$1.64 \times 10^2$</td>
</tr>
<tr>
<td>68.99</td>
<td>$^{139}$Cs</td>
<td>$2.24 \times 10^{-2}$</td>
<td>$1.27 \times 10^3$</td>
</tr>
<tr>
<td>89.1</td>
<td>$^{138}$Cs</td>
<td>$3.89 \times 10^{-2}$</td>
<td>$1.74 \times 10^3$</td>
</tr>
<tr>
<td>103.0</td>
<td>$^{138}$Cs</td>
<td>$2.51 \times 10^{-2}$</td>
<td>$9.60 \times 10^2$</td>
</tr>
<tr>
<td>105.87</td>
<td>$^{139}$Cs</td>
<td>$2.22 \times 10^{-3}$</td>
<td>$8.29 \times 10^2$</td>
</tr>
</tbody>
</table>

E2 transition, the enhancement factor is just the inverse of the half-life in Weisskopf units.

As a guide in the assignment of transition multipolarities, the following rules, based on empirical observations, have been adopted by the Nuclear Data Group at Oak Ridge National Laboratory (37). The gamma-ray half-life is expressed in Weisskopf units for each multipolarity, and WE stands for the half-life calculated using the Weisskopf estimates.
If the half-life is: The transition is not:

<table>
<thead>
<tr>
<th>Half-life (s)</th>
<th>Designation</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt; 0.1</td>
<td>M1</td>
</tr>
<tr>
<td>&lt; 1.0</td>
<td>M2</td>
</tr>
<tr>
<td>&lt; 0.1</td>
<td>M3</td>
</tr>
<tr>
<td>&lt; 0.033</td>
<td>M4</td>
</tr>
<tr>
<td>&lt; 100</td>
<td>E1</td>
</tr>
<tr>
<td>&lt; 0.001</td>
<td>E2</td>
</tr>
<tr>
<td>&lt; 0.01</td>
<td>E3</td>
</tr>
</tbody>
</table>

The nucleus $^{141}$Cs has 86 neutrons and 55 protons. According to the shell model, the ground-state spin and parity is probably given by the last unpaired proton which is in a $g_{7/2}$ orbital. It therefore seems reasonable to assume that the ground state of $^{141}$Cs has a spin-parity of $(7/2)^+$. This assumption is supported by the fact that the odd-$A$ Cs nuclei, $^{133}$Cs to $^{139}$Cs, all have $(7/2)^+$ ground states (38). All comparisons made below concerning these odd-$A$ nuclei will be based on information from Ref. 37.

The first excited state in $^{141}$Cs is at 68.99 keV. Comparing the measured half-life to the graph of the Weisskopf estimates, and noting that the E2 enhancement factor is higher than would be expected in this region of non-deformed nuclei, this transition is assigned a multipolarity M1. This level can be accounted for by assuming that the odd proton is excited to the next single particle state available to it, the $d_{5/2}$ orbital. This would mean that the spin-parity of the 69-keV level is $(5/2)^+$. The transition to the ground state would then be an $l$-forbidden type, for which $\Delta l > \Delta j$. According to the strict shell model, such a
transition would be impossible, but in reality several M1 l- 
forgidden transitions do occur, with large hindrance 
factors. The measured hindrance factor for this M1 transi- 
tion is 1270, which is not unreasonable as elaborated below.

Again, neighboring nuclei having similar nucleon con- 
figurations support the spin-parity of (5/2)+ and the 
multipolarity assignment of M1. \(^{133}\text{Cs}\) has its first excited 
state at an energy of 81 keV, and this is a (5/2)+ level 
(38). The multipolarity of the ground-state transition from 
this level is M1 with 2.6% E2 mixing. The M1 hindrance 
factor calculated from the 6.3-nsec half-life of the 81-keV 
level is 389.

\(^{135}\text{Cs}\) has a 250-keV transition from a (5/2)+ first 
excited state to a (7/2)+ ground state. This is also an l- 
forgidden transition, and using the listed value for the 
half-life of 0.3 nsec, assuming an M1 multipolarity, results 
in a hindrance factor of 229. The multipolarity has not yet 
been confirmed.

Other nearby nuclei such as \(^{139}\text{La}\), which also has its 
unpaired proton in a \(g_{7/2}\) orbital, have l-forgidden M1 transi- 
tions from the first excited state to the ground state. 
The 166-keV transition in \(^{139}\text{La}\) has a measured half-life of 
1.7 nsec, and multipolarity M1 with less than 0.4% E2 
mixing, giving an M1 hindrance factor of 427. Based on a 
comparison with these transitions in nuclei having similar
proton configurations, it is very probable that the 69-keV transition in $^{141}\text{Cs}$ is mostly M1.

The 106-keV level in $^{141}\text{Cs}$ is the second excited state. The enhancement factor for an E2 transition here is 455. Whereas this would not be too unusual for a deformed nucleus, it is very high for this level. Because of this, the transition is probably not E2. The M1 hindrance factor of 829, however, is more reasonable, and the transition can be classified as M1, possibly with a very small admixture of E2. Among the possible spin-parities for this level are $(3/2)^+, (5/2)^+$ and $(7/2)^+$. Systematics for nearby nuclei are little help in this case, and it is not possible to say which of the spin-parity assignments is probable. All three spins occur in neighboring nuclei, and all are compatible with the measured M1 multipolarity.

The nucleus $^{140}\text{Cs}$ has 85 neutrons and 55 protons. The odd-odd nature of the nucleus makes it difficult to predict spins and parities for the energy levels. For this reason, only the multipolarities of the transitions in $^{140}\text{Cs}$ will be considered. A recent study by Adams et al. (39) gives the spins and parities for the levels of $^{140}\text{Cs}$, along with transition multipolarities based on internal conversion coefficient measurements. The 13.8-keV level has been assigned a spin-parity of 2$^-$ and the transition to the ground state, which is either 1$^-$ or 2$^-$, is designated M1. According to
their work, the 64.75-keV level has a spin-parity of 3−, and the transition to the 14-keV level is M1. The 103-keV, 104.5-keV doublet could not be resolved for conversion coefficient measurements in the study, and the combined transition is assigned a mixed multipolarity of M1 and E2.

The results of the lifetime measurements of levels in 140Cs will now be compared to the above internal conversion coefficient results. The measured half-life of 471 nsec for the 13.8-keV transition, giving an M1 hindrance factor of 2470, agrees with the above multipolarity assignment.

The 50.6-keV transition from the 64.6 keV level has a very high E2 enhancement factor of 459. Again, such high enhancement is not found except in deformed nuclei. The M1 hindrance factor of 164 is, however, very reasonable when compared to other M1 transitions. Based on this, the 51-keV transition is assigned a multipolarity of M1. This agrees with the internal conversion measurement prediction.

As mentioned above, when two gamma rays depopulate the same level, partial half-lives for each transition must be deduced from the relation for adding two transition probabilities. In the case of the 103-keV level, there are two gamma rays leaving the level. The total transition probability is

$$\lambda = \lambda(89) + \lambda(103)$$

where \(\lambda(89)\) and \(\lambda(103)\) represent the transition probabili-
ties for the two gamma rays. Since each partial transition probability is proportional to the measured intensity of the gamma ray it represents, the ratio of the intensities gives the ratio of the transition probabilities. Using the intensities given by Adams et al. (39), the partial half-life of the 89-keV transition is 23.9 nsec and that of the 103-keV transition is 10.6 nsec. These values were used to calculate the half-lives listed in Table 5 for these two gamma rays. Comparing these values to the Weisskopf estimates, with M1 hindrance factors of 1740 and 960 for the 89-keV and 103-keV transitions, respectively, both transitions are classified as M1. The E2 enhancement factors of 25.7 and 39.8 are higher than would be expected, but do not rule out the possibility of E2 mixing. These two assignments agree with the measurements of Adams et al.

D. Comparison with Previous Work

A recent study by Clark et al. (40) gives the values of the half-lives of isomeric transitions in many nuclei observed following the spontaneous fission of \(^{252}\text{Cf}\). His results for the transitions under consideration in the present work are summarized in Table 6.

The values reported by Clark et al. for the half-lives of the 13.8-keV and 68.99-keV transitions agree reasonably well with the values reported here. As will be explained below, the technique which Clark used should be reliable for
Table 6. Comparison of results to previous work

<table>
<thead>
<tr>
<th>Transition Energy (keV)</th>
<th>Nucleus</th>
<th>Clark (40) (nsec)</th>
<th>This Work (nsec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>13.8</td>
<td>(^{140}\text{Cs})</td>
<td>521 ± 10</td>
<td>471 ± 51</td>
</tr>
<tr>
<td>50.6</td>
<td>(^{140}\text{Cs})</td>
<td>8 ± 1</td>
<td>3.7 ± 0.3</td>
</tr>
<tr>
<td>68.99</td>
<td>(^{141}\text{Cs})</td>
<td>22 ± 6</td>
<td>23.3 ± 0.7</td>
</tr>
<tr>
<td>89.1</td>
<td>(^{140}\text{Cs})</td>
<td></td>
<td>7.0 ± 0.4</td>
</tr>
<tr>
<td>89.9</td>
<td>(^{141}\text{Cs})</td>
<td>12 ± 1</td>
<td>&lt; 2.4</td>
</tr>
<tr>
<td>103.0</td>
<td>(^{140}\text{Cs})</td>
<td>11 ± 1</td>
<td>7.4 ± 0.3</td>
</tr>
<tr>
<td>105.87</td>
<td>(^{141}\text{Cs})</td>
<td>14 ± 1</td>
<td>8.7 ± 0.2</td>
</tr>
</tbody>
</table>

half-lives longer than approximately 10 nsec, and the agreement for these two transitions supports this.

Clark also reports an 89.9-keV transition in \(^{140}\text{Cs}\) with a half-life of 12 nsec. This transition has probably been assigned to the wrong nucleus, and is possibly the same transition reported in this work as the 89.1-keV transition in \(^{140}\text{Cs}\) with a half-life of 7.0 nsec.

For the 50.6-, 103.0- and 105.87-keV transitions, the half-lives given by Clark et al. are all longer than those measured in this work. His values for these short half-lives are possibly overestimated because of the method used.
in his study of $^{252}\text{Cf}$ to determine half-lives. For timing purposes, the fission-gamma coincidences were sorted into successive time bins, the upper limits of the first few bins being 5, 9, 15, 29... nsec. For half-lives under 9 nsec, this time-sorting technique gives at most four data points to use in calculating the half-life.

In the present work, the time interval between successive data points was less than 1 nsec. Thus, even for short half-lives, many more data points were used to find the half-life, and the reliability of the fitted results should be much greater. It cannot be excluded, however, that the fission-product half-lives reported by Clark et al. arise from a directly populated state not fed in beta decay which de-excites through the levels measured in this work, but with a longer lifetime.

E. Conclusion

The transitions whose half-lives have been reported in this study extend the list of known isomers in an important direction. The nuclei studied have proton and neutron numbers that lie just beyond the "magic numbers" of the shell-model theory, a region where very few lifetimes have been measured previously. Theoretical calculations using many-particle configurations with various interaction potentials can be done for such nuclei. Comparison to the experimental lifetime results, as well as to the energy levels where they
are known, will provide a good test of the model used in such calculations.
VI. LITERATURE CITED


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19. ORTEC Application Note AN 31.


39. J. P. Adams, F. K. Wohn, W. L. Talbert, Jr., W. C. Schick, Jr., and J. R. McConnell, "Internal conversion coefficient measurements for transitions in 140Cs from the decay of 140Xe" (to be published).
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