Photofission cross sections of $^{232}$Th and $^{236}$U from threshold to 8 MeV

Michael Vincent Yester

Iowa State University
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Photofission cross sections of $^{232}$Th and $^{236}$U from threshold to 8 MeV

by

Michael Vincent Yester

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I. INTRODUCTION

Within the last ten years, new discoveries have greatly altered our concepts of the fission process. Isomeric levels with half-lives ranging from 1.5 nsec to 14.0 msec have been observed in which the only known mode of decay is fission, and subbarrier fission resonances have been observed in reaction studies. These observations have been explained by theoretical calculations that predict a potential energy versus deformation function for heavy nuclei which has two maxima, rather than the previously accepted simple potential barrier with a single maximum. It also has been predicted that the second well, the well between the two maxima, could be quite deep (3 MeV) permitting the existence of discrete nuclear states in this well. In addition, it has been postulated that only certain quantum states (fission channels) are available to the nucleus at the saddle-point of fission, and, it is predicted that these states are widely separated in energy and represent simple types of motion of the nucleus.

While the above ideas are now supported by considerable experimental evidence, little is known of the character of the states in the second well. An attempt has been made to detect gamma-ray emission that precedes isomeric fission, but such transitions in the second well have not been observed (1). The subbarrier fission resonances observed in high resolution studies with particle reactions usually are attribut-
ed to transitions from the compound nuclear states of the first well to levels in the second well. Thus, the levels in the second well can be studied in their role as intermediate states to fission from the compound nuclear states in the first well. However, the maximum transfer of angular momentum can be quite large in particle-induced fission, and it is difficult to assign spins and parities to the compound nuclear levels that are excited. Photofission experiments can provide valuable information in this respect. In photon absorption, electric-dipole excitation is dominant, although electric-quadrupole and magnetic-dipole excitations can also contribute. For even-even nuclei, however, collective M1 excitations are forbidden eliminating excitations of 1+ states near threshold. Therefore, only 1− and 2+ states will contribute to the photofission cross section near threshold. Hence, photofission experiments can provide a starting point for interpreting the reaction data and, in general, can furnish tests of fission theories.

The early photofission measurements were made using gamma rays from the $^{19}$F(p,γ)$^{16}$O reaction and bremsstrahlung as the photon sources. Hyde (2) presents a good discussion of the early experiments and a comparison of some of the data. To summarize the major results, evidence of a plateau at approximately 6.0 MeV was observed for $^{238}$U, $^{232}$Th, and $^{236}$U. In the experiments performed with bremsstrahlung, a differen-
tial analysis technique was employed to obtain the cross sections from the experimental yield data, and the overall resolution in the results was poor. The experiments using the gamma rays from the $^{19}\text{F}(p,\gamma)^{16}\text{O}$ reaction only provided data at 6.14, 6.9, and 7.0 MeV. Thus, the possibility of structure in photofission cross sections has been an open question.

Recently, monochromatic gamma rays from thermal neutron capture have been used for photofission studies. Cross sections can be obtained only at various discrete energies, however, and the results are difficult to interpret as the energy spread of the gamma rays is of the order of the spacing between the levels in the compound nucleus.

To circumvent the above problems, a Compton scattering facility was built by Knowles and Ahmed (3) to provide a variable-energy monochromatic photon beam, and photofission studies were initiated. A similar facility was constructed at the Ames Laboratory Research Reactor (4,5), and a series of photofission experiments was planned. Hall began this project (6), and Anderl (7) has made a companion set of measurements to the present work.

The choice of target nuclei for this work was based on experimental as well as theoretical concerns. The silicon surface-barrier detectors used as fission-fragment counters in this work cease to function properly after receiving a
total dose of $10^{11}$ alpha particles. Consequently, only nuclides with extremely long half-lives for alpha-particle decay were considered as suitable targets. Since many experimental studies of the photofission reaction in $^{232}$Th have been made, this isotope was selected to enable direct evaluation of our experimental technique; whereas, $^{236}$U was selected since the photofission cross section of this nuclide has been measured previously only at energies of 6.14 and 7.0 MeV (8).

From a theoretical viewpoint $^{232}$Th and $^{236}$U offer an interesting comparison. Both are even-even so the spectrum of fission channels at the threshold should be similar. On the other hand, a spontaneously fissioning isomer has been observed in $^{236}$U (9,10,11,12), but no isomers have been reported for any of the thorium isotopes (13). Theory, however, predicts that the potential energy surface as a function of deformation should have two maxima in both isotopes. It is possible that the second well is quite shallow for the thorium isotopes, such that the half-lives of the isomeric states are too short to be observed with existing experimental techniques. A subbarrier resonance in the $^{230}$Th(n,f) cross section has been observed and is attributed to an excitation of a pure vibrational level in the second well (14). Thus, one might expect to see similar structure in the $^{232}$Th($\gamma$,f) reaction.
Experiments on $^{235}\text{U}(n,f)$ and $^{235}\text{U}(d,\text{pf})$, in which the compound nucleus formed is $^{236}\text{U}$, have indicated the presence of subbarrier resonances. Thus, the $^{236}\text{U}(\gamma,f)$ cross section can be compared with studies of particle-induced fission to determine the extent to which photofission measurements can add to our present understanding of nuclear fission. A study of the $^{236}\text{U}(\gamma,f)$ reaction is interesting, also, in view of the fact that the $^{235}\text{U}$ photofission cross section has been measured under the same experimental conditions as the present studies (7). The $^{235}\text{U}$ nucleus contains an odd number of neutrons so a comparison between the photofission cross sections of $^{235}\text{U}$ and $^{236}\text{U}$ can illustrate the role of the individual nucleons in the fission process. Finally, the $^{236}\text{U}$ photofission cross section can be combined with the $^{238}\text{U}$ and $^{235}\text{U}$ results obtained by Anderl (7) to provide data for a systematic study of photofission in the uranium isotopes.

The actual presentation of this work is divided into five major sections. The theory is discussed in Chapter II, and the details of the Compton scattering facility are presented in Chapter III. The photon beam used in this work is not monochromatic, and the analysis procedure used to extract the photofission cross sections from the yield data is outlined in Chapter IV. The experimental measurements are discussed in Chapter V, and the photofission cross sections obtained from the data are presented in Chapter VI. Conclusions and interpretations are presented in Chapter VII.
II. THEORY

The liquid-drop model was the first nuclear model used to treat the mechanism of fission \((15,16)\). In this model the nucleus is represented as a uniformly charged liquid drop in which the short-range forces acting between the nucleons are considered to be analogous to the surface tension of a liquid drop. The Coulomb repulsion between the protons is used for the long-range force. The model indicates that the surface tension increases as the deformation of the nucleus increases, i.e., as excitation energy is added to the nucleus. Thus, the surface tension counteracts an increase in deformation while the Coulomb forces tend to increase the deformation. If the nucleus is excited such that the electrostatic forces are larger than the surface tension, the nucleus will split into two fragments. The above ideas are contained in the expression that is calculated for the potential energy of the nucleus as a function of deformation. A schematic representation of the potential energy of deformation is shown as the solid curve in Figure 1. The fission saddle-point (fission barrier) is reached when the nucleus is excited to an energy at which the maximum in potential energy of deformation occurs. With the liquid-drop model of fission, values of the barrier heights can be calculated, spontaneous fission can be explained, and, in general, the gross features of the fission process are predicted.
Figure 1. Potential energy function of deformation with schematic representation of transition states expected for even-even nuclei.
In 1955 Bohr suggested that the nucleus could proceed toward fission only through distinct channels (17). The fundamental idea in the theory is that most of the excitation energy is transformed into potential energy of deformation as the fission threshold is approached. The small excess in energy is available as kinetic energy of vibration and rotation. Thus, the energy levels available to the nucleus at the fission threshold are expected to resemble the low-energy spectrum of the nuclear states at the ground state deformation. For even-even nuclei these transition states are expected to occur in the sequence shown in Figure 1. It should be noted that in indicating states at the top of the barrier no claim is being made that the transition states are bound states. Rather, of the many continuum states near the threshold, there exist certain states through which fission proceeds. The quantum numbers used in Figure 1 are defined as follows: I is the total angular momentum, \( M \) is the component of I along the space-fixed z-axis, and \( K \) is the component of the total angular momentum along the symmetry axis of the deformed nucleus. Information on these transition states can be obtained from studies of the energy dependence of the fission cross section and angular distribution of the fission fragments.

Since the nucleus undergoes a change of shape during the transition from the compound nuclear state to the transition
state, it is unlikely that the value of $K$ for the compound nuclear level is related to the value of $K$ for the transition state. It is assumed, however, that once the nucleus reaches the transition state deformation, $K$ is a good quantum number during scission. Throughout the entire transition from the compound nuclear state to fission, $I$ and $M$ are good quantum numbers. Thus, assuming that the fission fragments separate along the symmetry axis of the nucleus, the angular distribution of fragments from a transition state with quantum numbers $K$, $I$, and $M$ is uniquely determined. (For example, the angular distribution for a $K = 0$, $I^m = 1^-$ transition state is $(3/4)\sin^2\theta$; whereas, the angular distribution for a $K = 1$, $I^m = 1^-$ transition state is $(3/4 - (3/8)\sin^2\theta)$ (18)). Therefore, angular distribution measurements provide information on the character of the transition states, and measurements of the energy dependence of the cross section give information on the position of the transition states relative to the ground state energy. Bohr's fission-channel theory is supported by evidence of anisotropies in the angular distribution of fission fragments observed in photofission and neutron-induced fission (17).

As mentioned previously, the liquid-drop model gives a description of the average properties of nuclear fission. It treats the nucleus as a homogeneous distribution of nucleons in energy space and excludes shell effects, i.e., the non-
uniformity in the distribution of energy states of the individual nucleons. The first attempts at including shell effects into the theory of fission consisted of extrapolating the nuclear theories that worked for deformed nuclei of lighter masses to the heavy mass region where nuclear fission has been observed. These theories, however, failed to predict an extremum in the potential energy of deformation. A scheme was developed by Strutinsky (19-21) in which shell effects were included in the liquid-drop model. The shell effects were introduced by calculating the difference in energy at a particular deformation between the sum of the single-particle energies for a realistic potential and the sum of the single-particle energies for a uniform energy distribution of nucleons. This energy difference was added to the energy associated with the liquid-drop model. Strutinsky found that the shell corrections modulated the potential energy of deformation for nuclei in the actinide region such that two maxima appeared as shown schematically in Figure 2. The theory predicts that the inner barrier is lower in energy than the outer one in the thorium isotopes, but the energy difference between the barriers decreases as Z increases such that the inner barrier is highest in the plutonium isotopes. Many variations have been introduced into the theory and further discussions are available elsewhere (22,23,24,25). It is interesting to note that Tsang and Nilsson predict that
Figure 2. Schematic representation of double-humped fission potential
the potential energy function of deformation has two maxima for some of the lighter nuclei (23).

Prior to this new theory, spontaneously fissioning isomers had been observed in some of the americium isotopes (26,27). The value for the half-life of the isomer in $^{242}$Am was found to be 0.014 seconds, and the excitation energy of the isomer (relative to the ground state) was observed to be 2.9 MeV. Such isomers could not be explained within the framework of the theories available at that time. The existence of these isomers can be explained, however, in terms of Strutinsky's new theory. The explanation afforded is that an isomer is the lowest level of the second well, and the most favorable mode of decay for such a level is fission. Many such isomers have been observed to date (13).

In addition to the fission isomers, the occurrence of subbarrier resonances observed in $(n,f)$, $(d,pf)$, and $(p,p'f)$ reactions are best explained by the existence of a double-humped fission potential. Weigman (28) explained the resonances as follows: If the nucleus is excited to a level in the first well that is very near the energy of a level in the second well, the nucleus can undergo a transition to this intermediate state. The probability of fission is quite high for states in the second well, and a resonance is seen in the cross section. This process is illustrated in Figure 2 by the dashed line indicating such a transition for an excita-
tion of energy $E$. The nature of the subbarrier resonances is quite complicated. They can be due to the excitation of undamped vibrational levels in the second well or to vibrational levels that are strongly damped into much more complicated states of a compound nucleus type. Intermediate cases are possible, also. Bjornholm and Strutinsky (29) and Lynn (14) have presented extensive treatments of these resonances.

In Figure 2, there are states indicated at each of the barriers. These represent the fission channels as formulated by Bohr (17). Thus, it is expected that resonance structure can be observed in fission cross sections at energies below and near the threshold.
III. COMPTON SCATTERING FACILITY

A. Technical Aspects

As noted previously, photofission resonances near threshold can not be resolved well with bremsstrahlung. In such beams, the number of photons in the tip of the spectrum is small compared to the total flux. Consequently, as the end-point energy of the spectrum is increased, information about the cross section at the new energy is obscured by the large contribution from the lower energy photons. Excellent resolution, of the order of electron volts, is possible if neutron- or proton-capture gamma rays are used, but photofission cross sections are then limited to energies at which strong gamma ray lines are available.

In an attempt to combine the good features of each of the above techniques, a Compton scattering facility was constructed (4,5) at the Ames Laboratory Research Reactor (ALRR). The primary source of gamma rays is a piece of nickel located at the center of a tangential tube of the ALRR. At this location the thermal neutron flux is $3 \times 10^{13}$ n/sec/cm$^2$, and through the neutron capture process, a $2.14 \times 10^4$ Ci source of 9.0-MeV gamma rays is provided. A variable energy photon beam of moderate intensity is then obtained by scattering the primary beam from an aluminum plate.
The aluminum scatterer, nickel source, and a target are positioned on the circumference of a circle. All photons emitted from the source that scatter from the plate and strike the target have scattered through the same angle as shown in Figure 3. The energy of the scattered photons is given by the Compton formula for scattering from free electrons,

\[
\frac{1}{E'} = \frac{1}{E} + \frac{1 - \cos \theta}{m_e c^2}
\]

where \( E \) is the energy of the gamma rays emitted from the nickel source (direct or primary beam), \( E' \) is the energy of the gamma rays that scatter through an angle \( \theta \), and \( m_e c^2 \) is the rest mass of the electron. When the scatterer is positioned so that a circle with a different radius of curvature is formed, a new angle and, consequently, a new energy is defined for the scattered photons striking the target. The available end-point energy range for this facility is 2.8 - 8.2 MeV. (End-point energy in this work is used to denote the energy of the scattered 9.0-MeV nickel line for a given scattering angle.) The necessary scattering geometry is maintained by rotating the plate about a fixed point (the pivot point), bending the plate to correspond to the necessary radius of curvature, and rotating the target position through an angle \( \theta \) as shown in Figure 4. As determined from the dimensions of the source, the source to scattering plate distance, and the size of the aluminum plate, the energy res-
Figure 3. Focusing principle of the Compton scattering facility
Figure 4. Scattering geometry of the Compton scattering facility

\[ \tan \alpha = \frac{\sin \theta}{r/c + \cos \theta} \]
olution of the scattered beam is $2 - 4\%$ (4). The engineering aspects of the scattering plate assembly and target chamber positioning, as well as the dimensions of the facility, are discussed in detail elsewhere (4). The layout of the facility is shown in Figure 5. A number of Compton scattering facilities with different geometries and photon sources have been used by others (3,30-36).

Shielding and collimation are important aspects of the facility. They are designed to prevent extraneous gamma rays and neutrons from reaching the target, as well as to define the primary and scattered beams. The steel thermal shield for the reactor core, Figure 5, is a major source of contaminant neutron-capture gamma rays. The shielding and collimation shown in that figure prevent these photons from reaching the target directly; however, the primary beam scattering from the aluminum plate contains a very substantial contribution from this source. Another source of undesirable photons is the beam tube itself. It is possible for a gamma ray to scatter in the beam tube and reach the target position without undergoing any additional scattering. Such photons scatter through an angle which differs from the angle defined by the scattering plate assembly and are sufficiently intense as to degrade the resolution of the primary scattered beam. The shadow shields, Figure 5, were constructed to minimize this effect. The four individual shields can be moved in and
Figure 5. Scale drawing of the scattering facility showing a horizontal plane through the center of the gamma ray beam.
cut of the beam plane, and different ones are used depending on the end-point energy. At the target position the beam covers an area 2 in. in height and 1 in. in width.

B. Photon Beam Energy Spectrum

An accurate knowledge of the photon spectrum and flux as a function of the end-point energy was necessary for the measurement of cross sections. These energy spectra were determined experimentally with a pair spectrometer consisting of a 65 cm³ Ge(Li) detector and two 3 in. x 3 in. NaI detectors. The Ge(Li) detector was centered at the target position, and the NaI detectors were arranged facing the Ge(Li) detector and at 180° with respect to each other.

Such a detection system was required since a photon can interact with the Ge(Li) detector through the photoelectric effect, Compton scattering, pair production, or combinations of these. Consequently, even a monoenergetic photon beam will produce a complex spectrum in the Ge(Li) detector. In pair production, a positron-electron pair can be created from a photon of energy E for E greater than 1.022 MeV. The positron, upon slowing down in the detector, annihilates in the vicinity of an electron and produces two 0.511 MeV gamma rays at 180° with respect to each other. These photons can escape from the detector, and such an event in the Ge(Li) detector (known as a double-escape event) is recorded as a photon of energy E - 1.022 MeV. To take advantage of this process, the
NaI detectors were set up to detect 0.511 MeV gamma rays, and a double-escape event was stored in a multichannel pulse-height analyzer when a coincidence occurred between the three detectors. The necessary triple coincidence required timing information for each detector: ARC (Amplitude and Risetime Compensated) timing was used for the Ge(Li) detector, and crossover timing was used for the NaI detectors. The electronics diagram is shown in Figure 6. This procedure made it possible to ignore all other interactions in the Ge(Li) crystal and provided a means for determining the true energy spectrum of the scattered beam.

Gamma ray spectra were measured at scattering angles corresponding to 100 keV increments in the end-point energy over a range of 3.0 to 8.0 MeV. Data were obtained in two interleaved sets of 200 keV intervals to minimize systematic errors. (The target position can be set at 0.1° intervals in the scattering angle which corresponds to an end-point energy increment of approximately 25 keV.) The counting time for each measurement was about 8 hours and the maximum number of counts in the spectra were 400 - 600 counts/5 keV. The energy range for each spectrum was 1.3 to 10.3 MeV.

Corrections were incorporated into the pair spectrometer data to obtain the actual energy spectrum of the photons at the target position. These corrections, relative efficiency of the detection system and intensity loss in the absorbers
Figure 6. Block diagram of electronics used with the pair spectrometer.
used during the measurements, are energy dependent and can change the raw spectra significantly. Consequently, these effects were considered in great detail.

The energy dependence of the detection efficiency is contingent only on the double-escape peak efficiency of the Ge(Li) detector. This was measured from 4.0 to 9.0 MeV at the thermal column of the reactor by taking spectra of the (n,\gamma) reactions in Ni, Hg, Al, and Fe with the Ge(Li) detector. The double-escape peak efficiency from 2.0 - 3.5 MeV was obtained with a $^{56}$Co source.

Using an appropriate analytical function for the peak shapes of the lines in the measured spectra (Appendix A) in conjunction with a non-linear least-squares computer routine, the double-escape peak areas were found for the strong transitions. Relative intensities were calculated from these areas and were compared to the known relative intensities for the transitions in the (n,\gamma) reactions (37) and in the decay of $^{56}$Co (38). Using the relation

$$\frac{\text{measured relative intensity}}{\text{actual relative intensity}} = \text{relative efficiency}$$

a relative efficiency curve was obtained. This is shown in Figure 7. The smooth curve in Figure 7, which is of the form

$$\ln(\text{efficiency}) = A + B\cdot E + C\cdot E^2 + D\cdot E^3$$

(1)

where $E$ is the energy, is a fit to the data.
Figure 7. Relative efficiency curve for the 65 cm$^3$ Ge(Li) detector. The smooth curve is a second-degree polynomial fit to the natural logarithm of the data points.
When the beam measurements were made, it was necessary to reduce the number of photons striking the Ge(Li) detector to prevent saturation and degradation in resolution resulting from pulse pile-up. This was accomplished by placing Pb absorbers at the entrance to the target chamber. Different thicknesses of Pb were used as the end-point energy was changed: 6.8 cm at 8.0 MeV to 2.1 cm at 3.0 MeV. It was assumed that the beam intensity decreased exponentially with the thickness of the Pb according to the relation

\[ I = I_0 e^{-\mu(E)x} \]

Here, \( x \) is the thickness of lead and \( \mu(E) \) is the absorption coefficient as a function of energy. A smooth curve, similar to equation (1), was fit to the values of the absorption coefficient taken from ref. (39). These values are appropriate only for experiments where the gamma rays are collimated to a narrow beam, but this assumption seems to be valid for our experimental arrangement.

After the beam measurements were completed, the entrance to the target chamber was filled with paraffin to reduce the neutron background in the photofission experiments. The addition of the 17.32 cm of paraffin was taken into account in a manner similar to that used for the Pb. In this case, however, the values of the absorption coefficient were fit with the equation

\[ \mu(E) = A + B \cdot E + C \cdot E^2 + D \cdot E^3 \]
The known values were obtained from ref. (39) using the relation

\[ \mu(\text{paraffin}) = (6/7) \cdot \mu(\text{carbon}) + (1/7) \cdot \mu(\text{hydrogen}). \]

Again the values for a narrow beam geometry were used.

The energy spectrum of the beam incident on the aluminum plate is given in Figure 8. This spectrum was obtained using the pair spectrometer with the target chamber positioned at 0°. The corrections mentioned above have been included, but the resolution of the Ge(Li) detector has not been unfolded from the spectrum.

Figure 9 shows some representative scattered-beam energy spectra for several end-point energies, after the energy dependent corrections have been made. Each spectrum was normalized to the same number of photons incident on the scattering plate. The normalization procedure is discussed in the next section.

In order to portray more clearly the variation of gamma ray flux with end-point energy, a plot of the total number of photons with energies from 5.0 to 8.6 MeV versus end-point energy is shown in Figure 10. (The method used to determine the absolute flux is discussed in Section D.) The sharp breaks in the curve at the higher end-point energies occur where a different moveable shadow shield was used.
Figure 8. Direct-beam photon spectrum as measured with the pair spectrometer. Spectrum is not corrected for detector resolution.
Figure 9. Representative selection of scattered beam spectra for end-point energies 8.0, 7.0, 6.0, and 5.0 MeV.
Figure 10. Plot of the total number of photons between 5.0 and 8.6 MeV per 4 x 10^6 monitor counts as a function of end-point energy.
C. Beam Monitor

A beam monitor was incorporated into the facility so that all measurements could be normalized to a constant reactor operating condition. The thermal power output of the reactor is kept at 5.0 MW, but the control rod positions are varied from time to time to maintain this power level. (The rod positions can change considerably for several hours when reactor operation commences after a shutdown, and the rods are gradually raised as the reactor fuel is used.) Such changes influence the neutron flux at the nickel source which then causes a variation in our photon flux. Thus, a normalization procedure dependent only on the time duration of a measurement, or on reactor power, will be in error.

The monitor system used consists of a Si(Li) detector that views 90° scattering of photons from the aluminum plate at the pivot post, as shown in Figure 5. The associated electronics are included in Figure 6. The discriminator threshold was set at 2.0 MeV for an enhanced sensitivity to the higher energy photons in the primary beam. All events detected with an energy above the threshold are counted on a scaler, and the total number of monitor counts for a measurement is used in the normalization procedure.

Unfortunately, the monitor count rate is a function of the scattering plate position. This functional dependence was obtained by measuring the number of monitor counts for 10
minute intervals at each scattering plate configuration used for the beam measurements. Data were taken on several days that the reactor was stable, i.e., there were no shutdowns during the measurements, and none had occurred within 12 hours. A smooth curve was drawn through the data points, and values were picked off this curve for normalizing the beam measurements and photofission experiments. The curve used in the normalization procedure is shown in Figure 11.

D. Beam Flux

The measurements described thus far, give only the relative flux of the scattered beam. The absolute flux was obtained by measuring the absolute efficiency of the pair spectrometer using a 100 microcurie $^{56}$Co source. Gamma ray spectra were taken of this source with the pair spectrometer, and with a single 3 in. x 3 in. NaI detector, using the same counting geometry for each measurement.

The spectrum obtained with the single NaI detector was used to calibrate the $^{56}$Co source by finding the areas under the photopeaks (full-energy peaks) produced by the 2.598- and 3.451-MeV transitions; two separate peaks were used for a consistency check. These areas are a measure of the number of $^{56}$Co nuclei that decayed during the counting period. After correcting these areas for the photopeak efficiency of a 3 in. by 3 in. NaI detector in a narrow-beam-geometry experiment (40), the strength of the $^{56}$Co source at a particular time
Figure 11. The variation of the monitor count rate as a function of end-point energy
and day was known.

For the pair spectrometer data, the areas of the double-escape peaks for the two transitions of interest were calculated with the same computer program used in the relative efficiency measurement. Knowing the absolute decay rate of the source from the NaI measurement, the absolute efficiency of the pair spectrometer was found to be $5.1 \pm 0.6 \times 10^{-5}$ at 2.598 MeV. The result for the 3.451-MeV transition was consistent with this, but the errors involved were large because this transition is weak and not well resolved in the NaI gamma-ray spectrum. The beam spectra were then made absolute in terms of photon flux by applying the single multiplicative factor which converted the relative efficiency of the pair spectrometer to an absolute efficiency.

For purpose of perspective, the number of 9.0-MeV photons emitted from the Ni source was compared to the number that strike the aluminum plate and to the number that reach the target position. It was estimated that at an end-point energy of 8.0 MeV, the number of 9.0-MeV scattered photons at the target position is approximately $2.0 \times 10^5$ photons/sec. The corresponding value for an end-point energy of 5.0 MeV is approximately $5.0 \times 10^4$ photons/sec. This compares to $4.0 \times 10^{10}$ 9.0-MeV photons/sec striking the aluminum plate and $2.0 \times 10^{14}$ 9.0-MeV photons/sec emitted from the Ni source. The total number of photons that strike the target
with energies ranging from 5.0 to 8.6 MeV is $5.4 \times 10^4$/sec at an end-point energy of 5.0 MeV and $9.5 \times 10^5$/sec at an end-point energy of 8.0 MeV.
IV. PHOTOFISSION MEASUREMENTS

A. Experimental Apparatus

1. **Targets**

   The targets used for the photofission measurements were in the form of metal foils prepared by the Isotope Target Laboratory of Oak Ridge National Laboratory. These foils were wrapped around hollow lucite cylinders, 2 in. high and 7/8 in. outer diameter, and tacked in place with epoxy glue. Since the range of fission fragments is approximately 14 mg/cm² for the light fragment peak and 11 mg/cm² for the heavy fragment peak in the nuclei studied, the foil thicknesses used were less than 10 mg/cm². The $^{232}$Th target consisted of 156.4 mg of material (4.4 mg/cm²) and was assayed at 100% $^{232}$Th. The $^{236}$U target consisted of 317 mg of material (8.9 mg/cm²) assayed at 89.38% $^{236}$U, 9.20% $^{235}$U, 1.306% $^{238}$U, and 0.116% $^{234}$U.

2. **Vacuum chamber**

   The range of fission fragments in air is small so all the photofission measurements were performed in a vacuum of less than 10⁻² torr. For the $^{236}$U experiment, the silicon detectors had to be cooled to minimize effects due to radiation damage caused by the high alpha particle activity from the target. The vacuum chamber used in this experiment is shown in Figure 12. Freon at a high pressure was allowed to
Figure 12. Scale drawing of vacuum chamber used in photofission measurements
expand rapidly in the expansion chamber, and this process cooled the chamber. The detectors were thermally connected to the wall of this chamber; typical temperatures involved were 18° F at the expansion chamber and 32° F at the detectors. Although the detectors were not cooled for the $^{232}$Th experiment, the same vacuum chamber base was employed in both experiments so that the detector-target configuration was kept constant.

The apparatus was designed to allow measurement of the angular distributions of the fission fragments in addition to the total photofission cross section. In angular distribution measurements, a correction for target thickness non-uniformity must be considered. This effect, however, can be averaged out by rotating the target during the experiments. Such a capability was included by coupling the target holder to a motor as shown in Figure 12. The target motor was used in the $^{232}$Th experiment, but, later, a vacuum leak occurred around the shaft whenever the target was rotated. Since uniformity in target thickness is not crucial for total cross section measurements, the target motor was not used for the $^{236}$U experiment.

3. Detection system

a. Detectors The four silicon surface-barrier detectors (each 600 mm$^2$ in surface area) used for counting fission fragments were purchased from ORTEC. The detectors were
arranged in the vacuum chamber as shown in Figure 13, and
details of the collimation and shielding at the target posi­
tion are included, also. Each detector was positioned on a
rod that was located at 45° with respect to the gamma ray
beam, and the yield points were formed from the sum of the
number of events in the four detectors.

b. Counting system A block diagram of the electron­
ics used in the counting system is shown in Figure 14. Since
both nuclides studied in this work are alpha-particle
emitters, the discriminators were set at an energy high
enough to reduce the counts due to alpha pile-up. The known
alpha spectrum from a $^{228}$Th source was used to calibrate a
mercury-switch pulser for each detection channel. Once the
pulser was calibrated in energy units, the discriminator
thresholds could be set at any energy. The above procedure
is covered in greater detail by Anderl (7). For the $^{232}$Th
experiment the threshold was set at 15 MeV; whereas, a 20 MeV
threshold was used in the $^{236}$U experiment.

All events above the threshold in each detector were
counted on a scaler and also stored in a multichannel ana­
lyzer. This latter step was included as an auxiliary system
to the scalers and as a means for correcting the data if the
reactor shut down during a run. A summing amplifier was used
to route the signals from each fission-fragment detector and
the monitor to a particular section of the memory of the ana­
Figure 13. Top view of target chamber and final beam collimators
Figure 14. Block diagram of electronics used in the photofission studies
lyzer. The information stored in the memory was
destructively read out onto magnetic tape every 2000 seconds.
In this manner, the number of events in the fission detec-
tors and the monitor that occurred in any 2000 second inter-
val was known for the entire counting period. The reactor-
operation log was consulted to determine whether the reactor
went subcritical during a counting period. When this situa-
tion arose, the number of events detected while the reactor
was in operation was obtained from a listing of the data on
the magnetic tapes.

c. Detection efficiency The efficiency of the
detection system was measured by Anderl (7), and the basic
considerations involved are outlined below.

The efficiency of the detection system can be represent-
ed as a product of two factors. The first factor relates the
number of fission fragments that strike the detectors to the
total number that are produced in the target. The second
factor is a ratio between the number of fission fragments
counted (i.e., the number with an energy greater than the
discriminator thresholds) and the number that strike the de-
tector. The latter factor is considerably less than one be-
cause many fragments that reach the detector travel through
the target at some angle with respect to a perpendicular to
the target surface. The length of such a path can be large
compared to the range of the fragments. Consequently, the
energy spectrum of fission fragments striking the detectors is smeared out over an energy range of 0 to 100 MeV.

An estimate of the number of fragments that strike the detectors to the total number produced in the target was obtained from measurements with alpha particles. The ratio between the number of alpha particles detected to the expected total emitted from the $^{232}$Th target was experimentally determined. This ratio was corrected for the difference in range of alpha particles and fission fragments in the $^{232}$Th target for use in the efficiency calculation.

The number of fission fragments that strike the detectors with an energy below the discriminator threshold was determined by measuring the actual energy spectrum of the fragments for each detector and both targets. The direct beam was used for this measurement, and the lead plug in the front shielding wall was removed so that the direct beam passed from the reactor to the target unimpeded. With the intense photon flux provided, good statistical data on the energy spectrum of the fission fragments were collected in a multichannel analyzer in eight hours. The energy range of the accumulated spectrum was 1 to 100 MeV. The lower energy part of the spectrum was dominated by alpha particles, and eight hour runs were made with the beam port closed in order to account for the alpha-particle background. From the data taken for the $^{236}$U target, it was found that approximately 54% of
the fission fragments striking the detectors had energies below 20 MeV. The corresponding value for the $^{232}$Th target is 27%. The total detection efficiency for the photofission measurements was 3.6% for the $^{232}$Th experiment and 2.8% for the $^{236}$U experiment.

B. Experimental Procedure

Photofission data on $^{232}$Th and $^{236}$U were obtained in 100 keV increments of the beam end-point energy over a range of 5.0 to 8.0 MeV. To guard against systematic errors, the data were taken in two sets of 200 keV intervals. Fission fragments emitted from the targets were counted with the silicon surface-barrier detectors, and each yield point was corrected for the variation in monitor count rate with end-point energy (see Chapter III and Figure 11). Poisson statistics were used to calculate the experimental errors. A periodic check on the stability of the counting system, in particular the discriminator settings, was performed with the aid of the mercury-switch pulser.

As there was a possible contribution to the yield from neutron-induced fission, background data were obtained with the lead-filled beam gates (see Figure 5) lowered. The 6 in. of Pb effectively reduces the photon beam intensity by a factor of $10^4$ but the neutron flux at the target is not changed significantly. These measurements were made at intervals of 500 keV or less and normalized in the same manner.
as the yield data; a smooth curve was fit to the background data for use in subtracting this contribution from the yield. An error, $s$, was assigned to each point given by the variance of the fit,

$$s^2 = \frac{1}{N-n-1} \sum \left( \frac{1}{\sigma_i^2} \right) \left( \frac{1}{\sigma_i^2} \right) \left( y_i - y(x_i) \right)^2$$

where $N$ is the number of data points, $n$ is the number of parameters in the fit, $y_i$ is the data, $y(x_i)$ is the fitting function used, and $\sigma_i$ is the uncertainty in $y_i$.

C. Photofission Yields

1. $^{232}$Th yield

Each yield point was obtained in a total time of approximately 36 hours. A plot of the yield data, corrected for the monitor count rate, is shown in Figure 15; the statistical errors for the data range from 5% to 17%. Included also in Figure 15 is the measured background, and the straight line is a linear least-squares fit to the data. Comparing the yield and the background, it can be seen that the cross section below 5.0 MeV contains little or no strength so the observed threshold for the $^{232}$Th($\gamma$,f) reaction is 5.0 MeV. Duplicate data points were taken at several end-point energies (6.6, 7.4, and 7.9 MeV), and these were consistent with the original data within the statistical errors.
Figure 15. Measured photofission yield and background for $^{232}\text{Th}$
Ample time was available for precise measurement of natural background during reactor shutdowns. When the measured rate was compared to the fission background rate (reactor in operation, beam port open, but the gamma-ray beam blocked) the natural background accounted for the measured background except at the higher end-point energies. Even though the discriminators in the counting system were set at 15 MeV, some alpha pile-up must have occurred. The remainder of the background was attributed to neutron-induced fission.

The structure in the yield at 6.5 MeV is indicative of structure in the cross section. At the higher energies, however, the structure in the yield results from the fact that the beam flux is a discontinuous function of end-point energy (Figure 10).

2. $^{236}$U yield

The problem of alpha-particle pile-up was expected to be quite significant for the $^{236}$U target as a consequence of the greater amount of material and comparatively shorter half-life for alpha-particle emission. Therefore, the discriminators were set at 20 MeV, eliminating any significant contribution to the data from alpha pile-up. The high alpha rate, $7.4 \times 10^5$/sec, from the target can cause considerable radiation damage in surface barrier detectors at room temperatures, and the detectors were cooled for this measurement as explained previously. The run time required in the $^{236}$U ex-
The measured yield is shown in Figure 16, and the statistical errors range from 4% to 18%. Ten duplicate yield points were taken to obtain a good check on the reproducibility of the measurement. The relatively short counting time made such an extensive test feasible. These points were taken at end-point energies of 5.0, 5.3, 5.6, 6.3, 6.4, 6.5, 6.9, 7.2, 7.7, and 8.0 MeV. All but two of the duplicate points overlapped the originals within estimated errors, and the other two were within two standard deviations of the originals. The yield curve shown was formed by averaging the duplicate and original data points.

The results of the background measurements are included in Figure 16. The background was due to neutron-induced fission from the $^{235}$U contamination. Since the background was sizeable, many background points were taken for this measurement, and the smooth curve through the background data is a linear least-squares fit. As in the $^{232}$Th measurement, the yield at an end-point energy of 5.0 MeV was at background, and the $^{236}$U($\gamma$,f) threshold is assumed to be 5.0 MeV.
Figure 16. Measured photofission yield and background for $^{236}\text{U}$
V. DATA ANALYSIS

A. Least Structure Analysis

The Compton scattering facility used in this work provides a photon beam with a continuous energy distribution, rather than one discrete energy. Hence, the photofission cross sections could not be obtained directly, but were extracted from a measurement of fission yield as a function of end-point energy. The yield and cross section are related by the integral equation

\[ Y(E) = \frac{A}{N(E,E)} \sigma(E') \, dE' \, . \]

\( Y_T(E) \) is the total yield of photofission events per monitor count at an end-point energy \( E \). \( N_T(E,E') \) is the total number of photons per monitor count striking the target with energy between \( E' \) and \( E' + dE' \) at an end-point energy \( E \). The cross section per nucleus in units of cm\(^2\) at an energy \( E' \) is represented by \( \sigma(E') \), and \( A \) is the number of target nuclei per cm\(^2\). The area involved is the cross sectional area, 1 in. \( \times \) 2 in., of the beam at the target position as defined by the final collimator. In the integration limits, \( E_{\text{th}} \) is the observed threshold energy of the photofission reaction, and \( E_m \) is the maximum energy of photons in the beam.

A more convenient equation is the reduced-yield equation
\[ y(E) = \frac{1}{N(E,E')} \int_{E_{th}}^{E} s(E') \, dE' \]  (2)

Here, \( y(E) \) is the sum total of fission events detected at an end-point energy \( E \), normalized to \( M_f \) monitor counts, and corrected for the monitor response. The measured yield, \( y(E) \), is related to the total yield, \( Y_T(E) \), by

\[ Y_T(E) = \frac{y(E)}{M_f \epsilon_f} \]

where \( \epsilon_f \) is the total efficiency of the fission-fragment detection system. \( N(E,E') \) represents the beam spectra measured for \( M_b \) monitor counts after the energy dependent corrections (relative efficiency of the pair spectrometer and absorption due to lead and paraffin) have been included. Thus,

\[ N_T(E,E') = \frac{N(E,E')}{F M_b} \]

where \( F \) is the factor that converts the relative efficiency of the pair spectrometer to an absolute efficiency. Using the above relations, the correspondence between \( s(E') \), the reduced cross section, and \( \sigma(E') \), the true cross section, becomes

\[ \sigma(E') = \frac{s(E') M_f F}{\epsilon_f M_f A} \]  (3)

An analytical function for \( N(E,E') \) could not be determined, and no assumptions about the energy dependence of \( s(E') \) could be made, so equation (2) was solved numerically. Equation (2), however, is inherently unstable (41). That is, a small change in \( y(E) \) can produce large changes in \( s(E') \),...
and the degree of instability depends on the shape of the kernel $N(E, E')$. Also, $\gamma(E)$ is a measured quantity; therefore, statistical errors are associated with each yield point. Thus, any technique used for solving the reduced yield equation must involve some criterion by which $s(E')$ is judged to be an acceptable solution.

The method used in this work to obtain $s(E')$ is based on Cook's Least Structure Analysis (42). A similar treatment of this technique has been reported by Phillips (41), and Ramanis et al. (43) have compared the different methods of solving the reduced yield equation, equation (2), used in photonuclear yield curve analysis. As the name "least structure" implies, smoothing is applied to $s(E')$ during the analysis procedure such that the structure in $s(E')$ is a minimum under an appropriate constraint. Unless $\gamma(E)$ is measured with extreme accuracy, smoothing must be applied to either $\gamma(E)$ or $s(E')$ to prevent large oscillations from appearing in $s(E')$. Since $\gamma(E)$ is the measured quantity, it is more reasonable to perform smoothing on $s(E')$.

Equation (2) can be written as

$$\gamma(E_i) = \sum_j N(E_i, E_j) \bar{s}(E_j)$$

by assuming that $N(E, E')$ is constant over an energy range $\Delta E$. $N(E_i, E_j)$ is defined by
The function \( \tilde{s}(E_j) \), which will be referred to as the least structure solution, represents the average of \( s(E) \) over the interval \( E_j - \Delta E/2 \) to \( E_j + \Delta E/2 \). Mathematically, this can be written as

\[
\tilde{s}(E_j) = \frac{1}{\Delta E} \int_{E_j - \Delta E/2}^{E_j + \Delta E/2} s(E') \, dE'.
\]

The energies \( E_i \) are determined from the relation

\[
E_i = E_{th} + (i-1)\Delta E \quad i = 1, 2, \ldots, n
\]

where

\[
\Delta E = \frac{E_m - E_{th}}{n-1}.
\]

For convenience, let a subscript refer to a particular energy. Thus,

\[
y_i = y(E_i)
\]

and, therefore,

\[
y_i = \sum_j N_{ij} \tilde{s}_j.
\]

In further discussions \( N_{ij} \) will be referred to as the beam matrix. The constraint used by Cook to determine how much smoothing should be applied to \( s \) is that

\[
\chi^2 = \sum_{i=1}^{n} \left( \frac{\sum_j N_{ij} \tilde{s}_j - y_i}{dy_i} \right)^2 = n
\]

where \( dy_i \) is the error associated with \( y_i \).
B. Present Application Of Least Structure Analysis

In Cook's original treatment (42), n yield points were considered and $E_m$, the maximum energy of photons in the beam, corresponded to the end-point energy $E_n$. Therefore, the beam matrix was $n \times n$. In our case the "end-point energy" refers to the centroid of the scattered 9.0-MeV gamma rays at a particular scattering angle. Since there is an energy spread associated with the scattered gamma rays, there will be photons in the beam with energies greater than the nominal end-point energy (Figure 9). To account for these photons, the particular value chosen for $E_m$ was 8.6 MeV. The number of photons with energies ranging from 8.0 to 8.6 MeV represents a substantial contribution to the total flux at the higher end-point energies. For example, at an end-point energy of 8.0 MeV, 14% of the photons with energies between 5.0 MeV and 8.6 MeV are in the energy range of 8.0 to 8.6 MeV.

The equation derived by Cook for the least structure solution involves taking the inverse of the beam matrix (42). Since the beam matrix used in this work is $n \times m$ with $m > n$, the result quoted in ref. (42) was not immediately applicable to our situation. In the derivation of the least structure solution, however, the more general result

$$\mathbf{N^T \cdot W \cdot \hat{y}} = (\mathbf{N^T \cdot W \cdot N + \lambda S}) \cdot \hat{s}$$

appears as derived in Appendix B. This solution was previ-
ously considered by Twomey (44), but statistical weighting was not included. In the above matrix equation, $N^T$ is $m \times n$ and represents the transpose of $N$, the beam matrix. $W$ is an $n \times n$ diagonal matrix with elements $(1/dy_i)^2$. $S$ is an $m \times m$ smoothing matrix and the particular $S$ used was that labelled $S_1$ by Cook in ref. (42). The particular form of $S_1$ in ref. (42) is in error, however, and the correct form is given in Appendix B. The factor $\lambda$ is a scale factor and determines how much smoothing is to be applied such that $\chi^2 = n$, where $n$ is the number of data points. If $N$ is a square matrix the above equation reduces to the solution quoted by Cook (42).

The beam matrix was obtained by forming each beam spectrum for a particular end-point energy into 100 keV bins. Each element of the beam matrix, $N_{ij}$, is simply the sum of photons with energies between $E_j - \Delta E/2$ to $E_j + \Delta E/2$ for an end-point energy $E_j$. This step-wise approximation is suitable for our photon spectrum since the structure in the beam itself is wider than 100 keV. In using a rectangular beam matrix as described, more values for the cross section are obtained than yield points measured. The values of the least structure solution from 8.0 to 8.6 MeV are determined from insufficient information to be used as a reliable measure of the cross section in that energy range. In fact, the errors associated with these values are consistently larger than the errors for the solution at energies of 5.0 to 8.0
MeV. Thus, only the values up to and including 8.0 MeV will be presented.

C. Test and Particular Aspects of Analysis Procedure

As explained in the previous sections, the solution, $s$, that we obtain for the reduced-yield equation is a smoothed representation of the true solution. Thus, $s$ is related to the true solution, $s'$, by the relation

$$ s_i = \sum_j R_{ij} s'_j $$

where $R$ is called the resolution function. An expression for $R$ can be obtained from the equation used to find $s$

$$ N^T \cdot W \cdot \hat{y} = (N^T \cdot W \cdot N + \lambda S) \cdot \hat{s} $$

and the equation for $s'$

$$ \hat{y} = N \cdot \hat{s}' $$

Combining the above relations we find that

$$ R = (N^T \cdot W \cdot N + \lambda S)^{-1} \cdot N^T \cdot W $$

$R$ is called the resolution function because it is a measure of the minimum separation in energy of structure in a cross section that can be seen as separate peaks in the solution to the yield equation. Ideally, one would like the resolution function, $R(E_i, E_j)$, to be a delta function of unit amplitude that peaks at an energy $E_j = E_i$. In actuality, $R$ is Gaussian in shape and is characterized by some full-width-at-half-maximum. The value of this full-width-at-half-
maximum will be quoted as a measure of the overall resolution for the cross sections obtained.

A test case illustrating the meaning of the resolution function and its dependence on the statistical quality of the data was investigated. A cross section, with a functional form of two Gaussians added together, was constructed. From this assumed cross section, the values of the average cross section, $s'$, were calculated at 100 keV intervals. The yield, $Y_i'$, for the above cross section was obtained by multiplying the beam matrix and $s'$ together. A yield, $\hat{Y}_i$, with statistical fluctuations was then obtained from the relation

$$\hat{Y}_i = Y_i' + r\sqrt{Y_i'^2} .$$

The random number $r$ was generated from a Gaussian distribution of unit amplitude, zero mean, and unit standard deviation. A second test yield was formed from the same average cross section, but the statistical errors used were smaller by a factor of $\sqrt{10}$. That is,

$$Y_i = Y_i' + r\sqrt{\frac{Y_i'^2}{10}}$$

for the second case.

The solutions to the test yields are shown in Figure 17 along with the assumed cross section. Since the least structure solutions are smoothed and averaged, the solutions are represented as smooth curves even though information is
Figure 17. Test cases illustrating the dependence of the resolution of the analysis procedure on the statistical quality of the yield data.
obtained at 100 keV intervals. The length of the horizontal error bars represents the full-width-at-half-maximum of the resolution function at that energy. In test case #1 the widths of the resolution function at the energies of the two peaks overlap, and the solution appears as one large broad peak with only a hint that the peak is a doublet. In test case #2, however, the resolution is better, and two peaks are observed in the solution. The vertical error bars represent the propagation of the errors in the yield data through the defining equation (equation 4) for $s$. Thus, although the overall resolution is limited by the intrinsic resolution of the photon beam, this value can be approached by minimizing the statistical errors in the yield data.

From the above test and many others, it was found that good solutions to the yield equation could be obtained.
VI. PHOTOFISSION CROSS SECTIONS

A. $^{232}$Th Photofission Cross Section

The $^{232}$Th yield curve was analyzed according to the procedure outlined in Chapter V, and the result is given in Figure 18 and Table 1. The error bars do not include the ±30% error in the scale factor relating the reduced cross section and the absolute cross section (equation (3)). A prominent resonance is visible at an energy of approximately 6.3 MeV, and another resonance is indicated at an energy of about 7.6 MeV. The experimental resolution for this measurement varies from 450 keV at 5.0 MeV to 700 keV at 8.0 MeV.

Khan and Knowles have used Compton-scattered gamma rays to measure the photofission cross section of $^{232}$Th (45). Their result is compared with the present measurement in Figure 19. Both cross sections exhibit a large peak in the region of 6.3 MeV, but Khan and Knowles' measurement does not indicate the presence of a resonance near 7.6 MeV. Although Khan and Knowles used a similar photon source, the analysis procedure employed is quite different (45). They considered that the photon beam striking the target could be assembled from discrete lines with an energy spread associated with each line. An effective cross section was obtained from the yield by considering the presence of the discrete lines only. Then the energy spread of each line, which was experimentally measured, was unfolded from the effective cross section to
Figure 18. $^{232}$Th photofission cross section. Horizontal error bars indicate the resolution and represent the widths that would be observed for delta-function resonances.
# Table 1. Photofission Cross Sections of $^{232}$Th and $^{236}$U

<table>
<thead>
<tr>
<th>Energy (MeV)</th>
<th>$^{232}$Th Cross Section (millibarns)</th>
<th>$^{236}$U Cross Section (millibarns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.0</td>
<td>$-0.6 \pm 0.6^a$</td>
<td>$-2.6 \pm 1.3^a$</td>
</tr>
<tr>
<td>5.1</td>
<td>$-0.6 \pm 0.5$</td>
<td>$-2.0 \pm 1.1$</td>
</tr>
<tr>
<td>5.2</td>
<td>$-0.7 \pm 0.3$</td>
<td>$-1.1 \pm 0.8$</td>
</tr>
<tr>
<td>5.3</td>
<td>$-0.8 \pm 0.3$</td>
<td>$-0.1 \pm 0.7$</td>
</tr>
<tr>
<td>5.4</td>
<td>$-0.9 \pm 0.4$</td>
<td>$0.2 \pm 0.7$</td>
</tr>
<tr>
<td>5.5</td>
<td>$-0.6 \pm 0.4$</td>
<td>$0.7 \pm 0.8$</td>
</tr>
<tr>
<td>5.6</td>
<td>$0.1 \pm 0.5$</td>
<td>$1.3 \pm 0.9$</td>
</tr>
<tr>
<td>5.7</td>
<td>$0.9 \pm 0.5$</td>
<td>$2.1 \pm 1.0$</td>
</tr>
<tr>
<td>5.8</td>
<td>$1.7 \pm 0.5$</td>
<td>$3.2 \pm 1.0$</td>
</tr>
<tr>
<td>5.9</td>
<td>$2.9 \pm 0.5$</td>
<td>$4.2 \pm 1.0$</td>
</tr>
<tr>
<td>6.0</td>
<td>$4.5 \pm 0.5$</td>
<td>$4.7 \pm 1.0$</td>
</tr>
<tr>
<td>6.1</td>
<td>$6.2 \pm 0.5$</td>
<td>$4.9 \pm 1.0$</td>
</tr>
<tr>
<td>6.2</td>
<td>$7.6 \pm 0.5$</td>
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*aThese are the relative errors only. The ±30% error in the absolute scale has not been included*
Figure 19. Comparison of photofission cross section of $^{232}\text{Th}$ with that of Khan and Knowles (45)
get the actual cross section. Smoothing is not applied during the analysis. To use such an analysis technique, the relative contribution to the scattered beam from each discrete line in the direct beam must be well known. A representation of the beam could not be constructed in this manner since contaminant lines from the (n,γ) reaction in Fe and Al exist in the photon spectrum. The relative contribution of these non-localized sources to the overall intensity for each end-point energy is not known. Thus, the photon spectrum was measured and unfolded from the yield curves to obtain the cross sections.

Ignatyuk et al. have measured the $^{232}$Th photofission cross section using bremsstrahlung (46). A comparison of their result with our work is given in Figure 20. Again there is agreement on a prominent resonance at approximately 6.3 MeV. Ignatyuk et al. also see a peak at approximately 5.6 MeV.

Figure 21 shows a comparison of our measurement with that of Manfredini et al. (47) who used neutron-capture gamma rays as the photon source. Mafra et al. (48) have reported similar results. The photon spectrum consists of discrete lines several electron volts wide. Therefore, one must be careful in making a detailed comparison with these results since data are obtained at certain discrete energies only, and the excitation energy may be on or off resonance.
Figure 20. Comparison of photofission cross section of $^{232}$Th with that of Ignatyuk et al. (46)
Figure 21. Comparison of photofission cross section of $^{232}\text{Th}$ with that of Manfredini et al. (47)
B. $^{236}\text{U}$ Photofission Cross Section

The cross section obtained for the $^{236}\text{U}(\gamma,f)$ reaction is shown in Figure 22, and the numerical values are presented in Table 1. The contribution due to the presence of $^{235}\text{U}$ in the target was obtained from the $^{235}\text{U}$ photofission cross section measured by Anderl (7). Resonance structure in the $^{236}\text{U}$ photofission cross section is visible at approximately 6.0, 6.5, and 7.5 MeV, and there is also an indication of structure at approximately 5.3 MeV. The experimental resolution for this measurement varies from 500 keV at 5.0 MeV to 700 keV at 8.0 MeV. The structure at 5.3 MeV is not well resolved in the present measurement. The yield curve exhibits a significant rise from 5.1 to 5.2 MeV and a plateau from 5.3 to 5.5 MeV. The measurements at 5.0 and 5.3 MeV were repeated and the results agreed with the original data within one-half a standard deviation. There is reason to believe, therefore, that a resonance has been observed near 5.3 MeV.

As mentioned previously, ten duplicate yield points were measured in the $^{236}\text{U}$ experiment. The cross sections obtained from the original yield, the yield with the duplicate points substituted into the original yield curve, and the yield formed from the average of all the data were compared. No significant differences were observed. The only other data available on the $^{236}\text{U}$ photofission cross section were obtained by Huizenga et al. using gamma rays of energies 6.14
Figure 22. $^{235}\text{U}$ photofission cross section. Horizontal error bars indicate the resolution and represent the widths that would be observed for delta-function resonances.
and 7.0 MeV from the $^{19}\text{F}(p,\gamma)^{16}\text{O}$ reaction (8). The values obtained were $35 \pm 11$ mb at 6.14 MeV and $28 \pm 9$ mb at 7.0 MeV.
VII DISCUSSION OF RESULTS AND CONCLUSIONS

A. $^{232}$Th

1. Comparison with other measurements

Although Khan and Knowles (45) measured the photofission cross section of $^{232}$Th with a system very similar to ours, and over the same energy range, an obvious and puzzling discrepancy exists between the two sets of results at the upper end of the energy range. While a definite maximum near 7.6 MeV and a falling cross section section above this is observed in the present results, Khan and Knowles report a steady rise in the cross section in this region. Because of this discrepancy, an extensive search for possible systematic errors in the analysis procedure and beam measurement was made.

Obviously, the determination of cross sections from the yield data is dependent on an exact knowledge of the photon spectrum. In our case energy dependent corrections were included in the pair spectrometer data to obtain a representation of the photon spectrum used in the analysis procedure. The sensitivity of the measured cross sections to these corrections was determined by analyzing the $^{236}$U yield with different beam matrices in which the corrections were omitted one at a time. The $^{236}$U yield was chosen for these tests as more structure is in evidence in the cross section. No
appreciable change in the resonance structure was observed for any situation.

In addition to the above, a test was made to determine whether the maxima observed in the cross sections near 7.5 MeV could arise from an instability in the analysis technique for the last several data points. This was accomplished by unfolding the $^{236}\text{U}$ yield for different truncated sets of yield points. That is, the yield was unfolded using data only in the range of 5.0 to 6.1 MeV and separately in the range of 5.0 to 7.2 MeV. The cross sections obtained from these partial data sets matched up with the appropriate part of the cross section unfolded from the entire set of yield data.

The possibility of a systematic error in the pair spectrometer data was also considered. A plateau is observed in the measured photon spectrum over the range of 3.0 to 5.0 MeV for each end-point energy (Figure 9). Considering the direct beam spectrum (Figure 8), it is surprising that so many low energy photons are observed in the scattered beam spectra. If too many photons are included in the beam at the lower energy part of the photon spectrum, then the cross sections unfolded from the yield curves would exhibit spurious maxima at the higher energies. No systematic errors were found in the spectra taken with the pair spectrometer of the direct beam and $^{56}\text{Co}$ source. Since only discrete lines are present
in these spectra, it was easy to check for anomalous behavior in the measured spectra. It is concluded, therefore, that a true representation of the photon beam was obtained and the characteristic plateau at the lower energy part of the photon spectrum is attributed to photons that scatter in the collimators before reaching the target. As no systematic errors were found in the analysis procedure or beam measurement, we have confidence in the photofission cross sections as presented.

Ignatyuk et al. (46) report a peak at 5.6 MeV in the photofission cross section of $^{232}$Th (Figure 20); however, the resolution of our present measurement is not sufficient for us to expect to observe a peak of this strength at that energy.

Cramer and Britt have measured the fission cross section for $^{232}$Th using the $^{230}$Th($t$,pf) reaction (49). A broad maximum in the fission probability was observed at an excitation energy of 6.0 to 6.5 MeV. In a similar experiment, Back et al. have observed a resonance at 5.5 MeV (50) in addition to the broad peak reported by Cramer and Britt. A clearly defined resonance at an excitation energy of approximately 6.3 MeV in the $^{232}$Th($\alpha$,\alpha') reaction has been reported (51). There is evidence then that the resonance in the $^{232}$Th ($\gamma$,f) cross section near 6.3 MeV has been observed in particle-induced fission studies.
2. **Interpretations**

The heights of the fission barriers for $^{232}$Th have been deduced from experiments, and the values quoted in ref. (25) are $5.9 \pm 0.2$ MeV for the inner barrier and $6.1 \pm 0.2$ MeV for the outer barrier. Clearly then, the resonance reported in this work at approximately $6.3$ MeV is not a subbarrier resonance.

As noted in Figure 18, the peak at $6.3$ MeV is near the $(\gamma,n)$ threshold. If the $(\gamma,n)$ reaction begins to dominate at some energy, fluctuations can appear in the $(\gamma,f)$ cross section. Khan and Knowles (45) have estimated the photofission transmission factor for $^{232}$Th and have concluded that the structure at approximately $6.3$ MeV is a fission resonance and not a fluctuation caused by competition from the $(\gamma,n)$ reaction. Mafra et al. have measured the photofission and photoneutron cross sections for $^{232}$Th using neutron-capture gamma rays (48). The same structure visible in the $(\gamma,f)$ cross section was observed in the $(\gamma,n)$ cross section. It was suggested that the neutrons come from the deformed nucleus on the way to scission and not from a compound nuclear state.

Huizenga has calculated the dipole photofission cross section for $^{232}$Th assuming that the $K = 0$, $I^\pi = 1^-$ fission channel is at $6.3$ MeV, and that the $K = 1$, $I^\pi = 1^-$ channel occurs at $6.75$ MeV (18). Good agreement was obtained for
data furnished by Knowles in a private communication. In addition Manfredini et al. have measured the angular distribution of photofission fragments with neutron-capture gamma rays (52). They observe a peak in the ratio of dipole excitation to isotropic excitation at 6.4 MeV. Thus, it seems that the resonance observed near 6.3 MeV is due to the excitation of the $K = 0, I^\pi = 1-$ fission channel.

The resonance at 7.6 MeV will be discussed later.

B. $^{236}$U

1. Comparison with other measurements

The $^{236}$U photofission cross section reported here is in good agreement with the measurement by Huizenga et al. (8). Although the absolute scales differ, we do confirm that the cross section at 6.14 MeV is approximately equal to the value at 7.0 MeV.

Many studies of the $^{235}$U(n,f) reaction have been made. The neutron binding energy in $^{236}$U is 6.467 MeV and the ground-state spin and parity of $^{235}$U is 7/2-. Consequently, the spin and parity of the states excited in neutron-induced fission of $^{235}$U is 3- or 4- for excitation energies near the neutron binding energy. Such states will not be excited in photoexcitation of $^{236}$U.

The low-energy part of the fission cross section of $^{236}$U has been studied with other reactions. A small peak in
the fission probability of the $^{235}\text{U}(t, pf)$ reaction has been observed at approximately 5.4 MeV by Cramer and Britt (49). They also report that the fission probability reaches a maximum at 6.5 MeV. Back et al. have observed a change in slope at 5.3 MeV in the fission probability for the $^{235}\text{U}(d, pf)$ reaction (53). They attribute this to an excitation of a vibrational resonance in the second well through a $K^\pi = 0^-$ channel. Another shoulder in the fission probability is observed at 6.0 MeV and a maximum is observed at approximately 6.3 MeV.

2. Interpretations

The heights of the fission barriers for $^{236}\text{U}$ have been determined from experiments (25). The values quoted are 5.9 ± 0.2 MeV for the inner barrier and 6.0 ± 0.2 MeV for the outer one. Therefore, the small bump at approximately 5.3 MeV in the $^{236}\text{U}(\gamma, f)$ cross section is indicative of a subbarrier resonance. The energy of this peak is consistent with structure seen in the $^{234}\text{U}(t, pf)$ and $^{235}\text{U}(d, pf)$ reactions. Moreover, it is possible to excite the $I^\pi = 1^-$ member of a $K^\pi = 0^-$ band through photoexcitation. Thus, it seems that the structure in the $^{235}\text{U}(d, pf)$ reaction at 5.3 MeV (53) corresponds to the structure seen at approximately 5.3 MeV in the $^{236}\text{U}(\gamma, f)$ cross section.
The energy of the resonance observed at 6.0 MeV is below the \((\gamma, n)\) threshold (Figure 22) so the peak is a fission resonance. According to the measured barrier heights, this resonance is slightly above the second barrier. In analogy with the interpretation given for the \(^{232}\text{Th}(\gamma, f)\) cross section, it is reasonable to interpret the structure at 6.3 MeV as evidence of the \(K = 0, I^\pi = 1^-\) fission channel. This is supported by photofission angular distribution measurements made by Huizenga (18). In that measurement, a \(^{238}\text{U}\) target was irradiated with bremsstrahlung of maximum energies 5.25, 5.75, and 6.75 MeV. No quadrupole excitation was observed. The ratio of dipole excitation to isotropic excitation was found to increase from 5.25 at 5.25 MeV to 7.83 at 5.75 MeV but decrease to 2.25 at 6.75 MeV. Thus, there is evidence that a \(K = 0, I^\pi = 1^-\) level can be excited near 5.75 MeV. The bremsstrahlung spectrum is comprised of a continuous distribution of photons up to the maximum energy. Therefore, it is possible that several fission channels can be excited when the maximum energy of the bremsstrahlung spectrum is 6.75 MeV. If a \(K = 1, I^\pi = 1^-\) channel is excited in addition to a \(K = 0, I^\pi = 1^-\) channel for a maximum energy of 6.75 MeV, then the corresponding angular distribution will be a combination of two distributions. The angular distribution for a \(K = 1, I^\pi = 1^-\) level is opposite in character to the distribution of a \(K = 0, I^\pi = 1^-\) level such that the combined distribution is
nearly isotropic (see page 9). In view of the decrease in
dipole excitation observed by Huizenga (18) from 5.75 to 6.75
MeV, then, it seems possible that the resonance observed in
our photofission cross section at 6.5 MeV can be attributed
to the excitation of the $K = 1, I^\pi = 1^-$ fission channel.

The resonance observed at 7.5 MeV will be discussed
later.

C. Comparison of $^{232}$Th and $^{236}$U Results

The cross sections obtained for $^{232}$Th and $^{236}$U are shown
on the same scale in Figure 23. The cross sections are simi­
lar in that resonances are observed just above the fission
threshold and near 7.5 MeV in each nuclide. The dissimilar­i­
ties, however, are quite striking. A subbarrier resonance
seems to be present at 5.3 MeV in the $^{236}$U photofission cross
section, but no resonances of this type are observed in the
$^{232}$Th photofission cross section. Also, a third prominent
resonance is observed in the $^{236}$U($\gamma$,f) cross section, and the
strength of the resonance near 7.5 MeV in $^{236}$U is much
stronger than the corresponding resonance in the $^{232}$Th
photofission cross section. This suggests that there is a
change in the fission potential between the thorium isotopes
and the uranium isotopes.

The presence of resonances at 7.5 MeV in each nuclide is
puzzling. Similar resonances in the $^{238}$U and $^{235}$U
photofission cross sections were observed by Anderl (7) in a
Figure 23. Comparison of $^{232}\text{Th}$ and $^{236}\text{U}$ photofission cross sections
companion set of experiments to the ones reported on here. As was discussed at the beginning of the chapter, no source of systematic error could be found that might explain the presence of anomalies in the solutions to the yield equation. If one compares the strength of the resonances near 7.5 MeV for the four nuclei, it is observed that the ordering from the greatest to least strength is as follows: $^{235}$U, $^{238}$U, $^{236}$U, $^{232}$Th. It is interesting that this same trend appears in the fission channel of the giant-dipole resonance in these nuclei.

D. Comparison of $^{235}$U and $^{236}$U Results

The $^{235}$U cross section measured by Anderl (7) is compared with the $^{236}$U cross section in Figure 24. The cross sections are quite similar. The shoulder in the $^{235}$U cross section at 5.5 MeV corresponds in energy to a subbarrier fission resonance observed in the $^{234}$U(n,f) reaction at 5.5 MeV (54) so it seems that subbarrier fission resonances were observed in each nuclide. Although there is a slight energy shift between the structure in $^{235}$U and $^{236}$U photofission cross sections and the structure observed in $^{235}$U is more pronounced, the similarities suggest that the odd neutron in $^{235}$U does not affect the ($\gamma$,f) reaction greatly.
Figure 24. Comparison of $^{236}\text{U}$ and $^{235}\text{U}$ photofission cross sections. Data on $^{235}\text{U}$ was obtained by Anderl (7).
E. Suggestions For Future Work

Of primary importance, further photofission measurements are needed to clear up the discrepancies between this work and the work of Khan and Knowles concerning the resonances observed near 7.5 MeV in the $^{232}\text{Th}$ photofission cross section.

We interpret all the structure observed in the photofission cross sections as fission resonances. It is possible that only certain levels can be excited in the compound nucleus; that is, entrance channel resonances may exist. To show that the structure observed results from the nature of the exit channels (fission channels) as opposed to entrance channel phenomena, total photoabsorption cross sections should be measured for these nuclides. As neutron emission competes strongly with fission, $(\gamma,n)$ measurements with similar resolution as this work would be a great aid in understanding the structure observed in the $(\gamma,f)$ studies.

Photofission angular distributions as a function of energy could be measured with the Compton scattering facility. Such measurements could be used to calculate parameters of the fission barriers (18,46).

Although the photon beam used for these experiments is not monoenergetic, cross sections with good resolution were obtained. As discussed in Chapter V, the resolution of the solutions to the yield equation can be increased by improving
the statistical quality of the yield data. The simplest way of doing this is to construct a more efficient fission-fragment detection system. The spark chambers described by Stubbins et al. (55) might fill the need. The efficiency for such chambers was estimated to be 30%, and these detectors are not sensitive to alpha particles. Such high efficiency detectors would permit one to obtain good statistics for the yield data, and, in addition, the studies would not be limited to the nuclides with extremely long half-lives for alpha-particle decay.

F. Concluding Remarks

It has been shown that resonance structure can be observed in photofission measurements. In particular, subbarrier resonances can be excited, and the energies of the observed resonances are in agreement with structure observed in particle-induced fission cross sections.

Unfortunately, the photon beam used for the present measurements is not monochromatic, and the resolution is not as good as one would like. The Compton scattering technique is fine in principle, but a different source of photons is needed. With neutron-capture gamma-ray sources, the maximum energy is limited to less than 11 MeV, and even more serious, there is more than one energy in the direct photon spectrum so that a variable monoenergetic photon beam isn't possible. Within these limitations, however, it may be possible to
obtain a variable-energy photon beam of good resolution with a Compton scattering facility at a different reactor installation. The Ames Laboratory Research Reactor is not suited for such a facility because of the large amount of iron used in the construction of the reactor. The thermal neutron-capture cross section for iron is large and a considerable fraction of the primary beam is composed of contaminant gamma rays. A clean photon beam and a higher photon flux might be obtained at other reactors.

In summary, photofission measurements are interesting and can provide useful information concerning the fission process near threshold. High resolution studies cannot be made as of this time, but such studies would greatly increase our understanding of nuclear fission.
VIII. BIBLIOGRAPHY


IX. ACKNOWLEDGEMENTS

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I am also grateful to Dr. D. J. Zaffarano for his interest in this project and in my graduate career at Iowa State University.

This work would be incomplete without expressing thanks to Mr. F. A. Anderl. The many discussions with him on the different phases of the project were very beneficial and provided many new insights. It was a pleasure to work with him.

Special thanks go to my parents, Mr. and Mrs. V. B. Yester, who have done so much to help me further my education.

Finally, my wife, Carole, deserves a special note of thanks. Her patience, understanding, and encouragement have been most helpful and her support is greatly appreciated.
X. APPENDIX A: ANALYTICAL LINE SHAPES FOR GE(LI) SPECTRA

Line shapes of peaks in Ge(Li) detector gamma ray spectra are not symmetric. There tends to be a broadening of the low energy side of the peak, and the prominence of this low energy tail varies with energy and detector size. Geiger et al. (56) have reported on a particular analytical function used for fitting such peaks. They considered a peak to be composed of an infinite distribution of Gaussians starting at \( x' \) and decreasing in intensity exponentially for \( x < x' \). Mathematically this can be represented as

\[
P(x) = \int_{-\infty}^{x'} e^{-\frac{1}{2} \left( \frac{x-y}{\sigma} \right)^2} e^{-b(x'-y)} \, dy.
\]

\( P(x) \) is the line shape; \( x' \) denotes the "centroid" of the peak (\( x' \) is slightly higher than the visible peak position); \( b \) determines how fast the Gaussians decrease in intensity; \( \sigma \) is the width of the Gaussians (\( \sigma \cdot 2.345 \) is the full-width-at-half-maximum); and \( A \) is a measure of the peak height.

This function is appealing in that it involves only four variable parameters. Geiger et al., however, approximated the integral with a sum and kept only a specified number of terms. We have improved on this method and eliminated any dependence on a judgment concerning the number of terms used in the sum by obtaining an exact expression for the integral.
The calculation is straightforward but somewhat lengthy and is not reproduced in detail here. The basic steps followed include two changes of variables to get the integral into the form

\[ \int_{-\infty}^{\infty} e^{-z^2} \, dz \]

The final result is

\[ P(x) = \sqrt{\frac{\pi}{2}} A \cdot e^{(x-x')b} e^{b^2s^2/2} \left[ 1 - \text{erf} \left( \frac{1}{\sqrt{2}b} (x-x'+s^2 \cdot b) \right) \right]. \]

The error function, erf, is available as a standard function in the computer library.

In order to find relative intensities of lines in a gamma-ray spectrum, an expression for the area under the peak was needed and was calculated from

\[ \text{Area} = A \int_{-\infty}^{\infty} \left( \int_{-\infty}^{x'} e^{-\frac{1}{2} \left( \frac{x-y}{\sigma} \right)^2 - b(x'-y)} \, dy \right) \, dx. \]

This can be rewritten as

\[ \text{Area} = A \int_{-\infty}^{x'} e^{-b(x'-y)} \left( \int_{-\infty}^{\infty} e^{-\frac{1}{2} \left( \frac{x-y}{\sigma} \right)^2} \, dx \right) \, dy. \]

However

\[ \int_{-\infty}^{\infty} e^{- \frac{1}{2} \left( \frac{x-y}{\sigma} \right)^2} \, dx = \sigma \sqrt{2\pi} \]

and one finally obtains the result
Area = $\frac{A\sigma\sqrt{2\pi}}{B}$.

Two parameters for a straight line were included in the fitting function to account for any background under the peak, i.e., the complete form of the function used was

$$F(x) = B + C \cdot x + P(x).$$

All parameters are fit to a peak simultaneously in a non-linear least-squares routine. Good fits to gamma ray peaks at energies ranging from 2 MeV to 10 MeV were obtained.
XI. APPENDIX B: DERIVATION OF LEAST STRUCTURE SOLUTION

We want to find the solution to

$$V_i = \sum_j N_{ij} \bar{S}_j$$

such that the structure in $\bar{S}$ is minimized under the constraint that $\chi^2 = n$. If we let $F(\bar{S}_k)$ be the structure function, that is, $F$ is a measure of the amount of structure in $\bar{S}$, then we want to find a solution to

$$V = F(\bar{S}_k) + \frac{1}{\lambda} \chi^2 (\bar{S}_k)$$

such that $V$ is minimized for the $\bar{S}_k$. The factor $1/\lambda$ is an undetermined Lagrange multiplier. Therefore, we want to solve the equation

$$\delta F(\bar{S}_k) + \frac{1}{\lambda} \delta \chi^2 (\bar{S}_k) = 0$$

where $\delta$ represents the variation with respect to $\bar{S}_k$.

Let

$$F(\bar{S}_k) = (T \cdot \bar{S})^2$$

or

$$F(\bar{S}_k) = \sum_i \sum_j T_{ij} \bar{S}_j \bar{S}_l$$

So

$$\delta F(\bar{S}_k) = \frac{3}{\delta \bar{S}_k} \left[ \sum_i \sum_j T_{ij} \bar{S}_j \bar{S}_l \right] \delta \bar{S}_k$$

$$= \left[ \sum_i \left( \sum_j \delta_{jk} T_{ij} \bar{S}_l + \sum_j T_{ij} \delta_{ij} \right) \right] \delta \bar{S}_k$$
where $\delta_{jk}$ is the Kronecker delta

\[
\delta_{jk} = 0 \quad \text{if } j \neq k \\
\delta_{jk} = 1 \quad \text{if } j = k
\]

Thus, we can now write

\[
\delta F(\bar{S}_k) = \left[ \sum_i \left( T_{ik} \bar{T}_{il} \bar{s}_{l1} + \sum_j T_{ij} \bar{s}_{ij} T_{ik} \right) \right] \delta \bar{s}_k
\]

\[
= \left[ \sum_i \left( \sum_j (T_{ki})^T \bar{T}_{il} \bar{s}_{l1} + \sum_j (T_{ki})^T T_{ij} \bar{s}_{ij} \right) \right] \delta \bar{s}_k
\]

\[
= \left[ \sum_i \sum_j (T_{ki})^T \bar{T}_{il} \bar{s}_{l1} + \sum_j \sum_i (T_{ki})^T T_{ij} \bar{s}_{ij} \right] \delta \bar{s}_k
\]

\[
= 2 \sum_i S_{kl} \delta \bar{s}_k
\]

where the superscript $T$ refers to the transpose of the matrix, and

\[ S = (T)^T \cdot T \]

For $\chi^2(S_k)$ we have

\[ \chi^2(S_k) = \sum_i W_i \left( \sum_j (\bar{S}_{ij} - Y_i)^2 \right) \]

where $W_i = 1/dy_i^2$ and so

\[
\delta \chi^2(S_k) = \frac{\partial}{\partial \bar{s}_k} \left( \sum_i W_i \left( \sum_j (\bar{S}_{ij} - Y_i)^2 \right) \right) \delta \bar{s}_k
\]

\[ = \left( 2 \sum_i W_i N_{ik} \left( \sum_j (\bar{S}_{ij} - Y_i) \right) \right) \delta \bar{s}_k \]

We can represent $W_i$ as a matrix by using the relation

\[ W_{il} = W_i \delta_{il} \]

Therefore we can write

\[
\delta \chi^2(S_k) = \left\{ 2 \sum_i W_{il} N_{ik} \left( \sum_j (\bar{S}_{ij} - Y_i) \right) \right\} \delta \bar{s}_k
\]
since
\[ \sum_{l} W_{il} N_{lk} = \sum_{l} W_{il} \delta N_{lk} = W_{il} N_{lk} . \]

Rewriting the above, we get
\[ \delta \chi^2 (\bar{s}_k) = \left\{ 2 \sum_{l} (N_{kl})^T \sum_{i} \left( \sum_{j} N_{ij} \bar{s}_j - y_i \right) \delta \bar{s}_k \right\} \]
where we have used the fact that
\[ \bar{W}_{il} = \bar{W}_{li} . \]

Combining these results we get
\[ \left\{ \sum_{l} N_{kl} \bar{s}_l + \frac{1}{\chi_i^2} \sum_{l} (N_{kl})^T \sum_{i} \left( \sum_{j} N_{ij} \bar{s}_j - y_i \right) \right\} \delta \bar{s}_k = 0 . \]
The \( \delta \bar{s}_k \)'s are linearly independent, and assuming that \( \lambda \neq 0 \), we have,
\[ N^T \bar{W} \bar{y} = \left( N^T \bar{W} \bar{N} + \lambda S \right) \bar{s} . \]

after putting the equation in matrix form.

In our work we let \( (\bar{T} \cdot \bar{s}) \) be the first difference in the \( \bar{s}_k \)'s. That is,
\[ (\bar{T} \cdot \bar{s})_k = \bar{s}_{k+1} - \bar{s}_k . \]

From this we find that
\[
\bar{s} = \begin{bmatrix}
1 & -1 & 0 & 0 & \ldots & \ldots & \ldots & \ldots & \ldots & 0 \\
-1 & 2 & -1 & 0 & \ldots & \ldots & \ldots & \ldots & \ldots & 0 \\
0 & -1 & 2 & -1 & \ldots & \ldots & \ldots & \ldots & \ldots & 0 \\
\vdots & \ddots & \ddots & \ddots & \ddots & \ddots & \ddots & \ddots & \ddots & \ddots \\
0 & 0 & 0 & 0 & \ldots & \ldots & -1 & 2 & -1 & 0 \\
0 & 0 & 0 & 0 & \ldots & \ldots & 0 & -1 & 1 & \ddots
\end{bmatrix} .
\]
The above is what Cook (42) calls the $S_1$ smoothing matrix and the correct form is given here.