Measurements of some gamma-ray relative intensities and internal conversion coefficients using a bent-crystal monochromator

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by

Gerald Clifford Nelson

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ABSTRACT

The X-ray and gamma-ray relative intensities were measured from the decay of Tm$^{170}$, Ho$^{166}$, Hf$^{180m}$, Eu$^{155}$ and Sm$^{155}$ with a bent-crystal monochromator and a linear least-squares computer program. The K-shell internal conversion coefficients were determined for the E2 transitions in Yb$^{170}$ and Er$^{166}$. The K-shell conversion coefficient, $\alpha_K$, for the 84.3-keV transition in Yb$^{170}$ was determined to be $1.43\pm0.04$ while the K-shell internal conversion coefficient for the 80.6-keV transition in Er$^{166}$ was determined to be $1.72\pm0.06$. The results for these $2^+\to0^+$ transitions are five percent higher than the theoretical values for these transitions. From the relative intensities of the transitions in Hf$^{180}$ it was possible to deduce a value for the total internal conversion coefficient for the 93.3-keV transition of $\alpha_1^{93} = 4.91\pm0.23$. Using the previous measurements of conversion electron intensities of Edwards and Boehm and the present measured gamma-ray relative intensities, internal conversion coefficients for all the other transitions were obtained. The present measurements of $\alpha_K$ for the 215.3-, 332.5- and 443.8-keV E2 transitions are 11 percent lower than the theoretical values, while $\alpha_K$ for the 93.3-keV E2 transition agrees closely with the theoretical value. These results are in close agreement with the previous measurements of Edwards and Boehm. The present value for $\alpha_K$ for the 501-keV transition agrees closely with the theoretical $\alpha_K$ for an E3 multipolarity. From the X-ray and gamma-ray relative intensities of the transitions in Gd$^{155}$ and the previous measurement of the ratio of K conversion electrons for the 86- and 105-keV transitions of Subba Rao, it was possible to determine the K
conversion coefficients for the 86- and 105-keV transitions of \( \alpha_K = 0.43 \pm 0.06 \) and \( \alpha_K = 0.23 \pm 0.03 \). These results are in agreement with the theoretical values for pure E1 transitions. The relative intensities of the 246-, 142- and 104-keV gamma rays following the decay of 22 minute Sm\(^{155}\) were determined with improved precision in order that they might be used to determine accurately the conversion coefficients for these transitions.
I. INTRODUCTION

A. Definition of the Internal Conversion Process

Below 1-MeV the principal processes by which an excited nucleus can make a transition to a lower energy level are gamma-ray emission and internal conversion. In the first process the nucleus emits a gamma ray with energy equal to the transition energy,

\[
N^* \rightarrow N + \gamma
\]

where \( N^* \) is the nucleus in the excited state, \( N \) is the nucleus in the lower energy state and \( \gamma \) is the emitted gamma ray which has an energy equal to the transition energy. In internal conversion the nuclear transition energy is transferred to one of the orbital electrons by a direct interaction between the electron and the charged nucleons. The electron is then ejected from the atom with an energy equal to the nuclear transition energy minus the binding energy of the electron.

\[
N^* + Ze \rightarrow N + (Z-1)e + e_{\text{continuum}}
\]

\[
E_e = E_{N^*-N} - E_{\text{binding}}
\]

where \( N^* + Ze \) is the excited nucleus with \( Z \) electrons, \( N + (Z-1)e \) is the nucleus in the lower energy state with \( Z-1 \) electrons, \( e_{\text{continuum}} \) is the ejected electron in the continuum, \( E_e \) is the energy of the ejected electron, \( E_{N^*-N} \) is the transition energy and \( E_{\text{binding}} \) is the binding energy of the ejected electron.
Following the ejection of an internal conversion electron, the atomic electrons will readjust, and an outer electron will fill the vacancy. The energy difference is carried off by one of two processes. The first and predominant process is the emission of an X-ray which will have an energy equal to the difference between the binding energy of the shell in which the vacancy occurred and the binding energy of the shell from which the outer electron came. The other process by which energy is carried off following internal conversion is the emission of a second electron called an Auger electron. The resulting atom is ionized in two shells. The energy of the emitted electron is approximately given by

$$E(KXY) = E(K) - E(X) - E^X(Y) = E(K) - E^Y(X) - E(Y),$$

where K, X, and Y are respectively the shell from which the internal conversion electron is ejected, the shell from which the K shell is filled, and the shell from which the Auger electron is emitted. $E^X(Y)$ is the electron binding energy of the Y shell in an atom with charge Z ionized in the X shell.

For a given transition the internal conversion coefficient, $\alpha$, is defined as the ratio of $N_e$, the number of internal conversion electrons emitted per unit time, to $N_\gamma$, the number of gamma rays emitted per unit time,

$$\alpha = \frac{N_e}{N_\gamma}.$$

The internal conversion coefficient for a particular shell or subshell is defined similarly. For the K shell

$$\alpha_K = \frac{N^K_e}{N_\gamma},$$
where \( N^K_e \) is the number of internal conversion electrons emitted from the K shell per unit time. The total internal conversion coefficient is the sum of the internal conversion coefficients of the individual shells.

\[
\alpha_T = \alpha_K + \alpha_L + \alpha_M + \ldots
\]

B. Remarks About Internal Conversion Coefficients

The internal conversion coefficients depend strongly on five parameters: the shell in which the conversion occurs, the transition energy, the atomic number, the angular momentum change and the parity change between the initial and final nuclear states. Internal conversion coefficients always increase as the transition energy decreases. They normally increase with \( Z \), and always increase as the angular momentum, \( L \), increases. To a large extent, internal conversion coefficients are independent of detailed nuclear structure. This makes it possible to obtain information about the spin and parity of the nuclear transition by comparing the experimentally determined conversion coefficients with those theoretically predicted.

When the nuclear angular momenta for initial and final states are \( J_i \) and \( J_f \), the emitted gamma ray can have any angular momentum \( L \) for which

\[
\Delta J = |J_i - J_f| \leq L \leq J_i + J_f.
\]

The electromagnetic transitions are classified as electric \( 2^L \), EL, or magnetic \( 2^L \), ML, if the parity change between the initial and final nuclear states is \((-1)^L\) or \((-1)^{L+1}\), respectively.

The internal conversion coefficient is in general a mixture of
conversion coefficients of pure angular momentum $L$

$$\alpha = \sum L a_L a_L ,$$

where $\sum L a_L = 1$.

The $a_L$ represent the fraction of total gamma rays emitted with angular momentum $L$. For a given type of multipole, the relative intensity for multipoles with $L$ and $L+2$ is given by (1)

$$\frac{a_{L+2}}{a_L} \approx \left( \frac{R}{\lambda} \right)^4 \ll 1,$$

where $R$ is the nuclear radius and $\lambda$ is the wavelength of the radiation. For $A = 200$ and $E = 511$-keV, one gets $a_{L+2} / a_L = 3.2 \times 10^{-9}$. Therefore, the mixture can be restricted to two multipoles. Assuming parity conservation in electromagnetic transitions, and from the parity selection rules, if the parity changes in the transition, only electric multipoles of odd order or magnetic multipoles of even order can occur. If the parity remains the same, only electric multipoles of even order or magnetic multipoles of odd order can occur. For example, if $J_i = 1$ and $J_f = 2$, and the parity does not change

$$\alpha_K = a_1 \alpha_K(M1) + a_2 \alpha_K(E2),$$

and

$$a_1 + a_2 = 1.$$
Similarly,

\[ \alpha_L = \alpha_1 \alpha_L(M1) + \alpha_2 \alpha_L(E2), \]

where \( L \) now denotes the \( L \) shell. These two equations can then be solved for the mixing ratio

\[ \delta^2 = \frac{\alpha_2}{\alpha_1} \left( \frac{\alpha_K(M1) - \alpha_K/\alpha_L \alpha_L(M1)}{\alpha_K(E2) - \alpha_K/\alpha_L \alpha_L(E2)} \right), \]

where \( \alpha_K/\alpha_L \) is the measured value, and \( \alpha_K(M1), \alpha_L(M1), \alpha_K(E2) \) and \( \alpha_L(E2) \) are theoretical values. If either \( J_i \) or \( J_f = 0 \), then \( L = \Delta J \), and the transition consists of only one multipole. Therefore, direct comparison can be made in this case between the experimental conversion coefficient and the theoretical conversion coefficient.

Rose (1) and Sliv and Band (2) have tabulated internal conversion coefficients as a function of atomic number and transition energy. These tables have been calculated taking into account screening effects and finite nuclear size. A uniform charge distribution is used inside the nuclear volume and a Thomas-Fermi-Dirac potential is used outside the nuclear volume.

By allowing the nucleus to have a finite nuclear size, the electron wavefunction is modified since the electron moves in the field of an extended charge distribution. This is the so-called static effect because it depends only on the nuclear density. Also, the electron spends a fraction of its time inside the nucleus where it probes the details of the nuclear charges and currents. This is the dynamic effect. If this penetration term is ignored, the conversion coefficient depends only on the electron wavefunction. Rose (1) has calculated conversion coefficients for the \( K, L \),
and L_{II} shells including screening and static effects. His L_{III} conversion coefficients include only screening, and his M coefficients are calculated for a point nucleus without screening. Sliv and Band (2) have calculated conversion coefficients for the K and L shells including screening, static effects, and dynamic effects. For the dynamic effects, they assume a uniform surface current density.

The dynamic effect is usually small since the electron spends so little time inside the nucleus. However, Church and Weneser (3) have pointed out that there are transitions for which the gamma-ray matrix element is greatly inhibited, while the nuclear penetration matrix element may have its uninhibited value. In cases where the penetration terms are not important, the errors in the tabulated values are about three percent (1).

Conversion coefficients in isotopes in the highly deformed regions are of particular interest for showing nuclear structure effects. These regions are $A = 23, 150 \leq A \leq 190$ and $A > 230$. In these regions the transitions may be highly retarded over single particle estimates, and conversion coefficients for these hindered transitions may deviate considerably from those predicted by theories which do not take into account the detailed nuclear structure.

C. Experimental Methods of Measuring Internal Conversion Coefficients

Subba Rao (4) has recently written an extensive review article on the methods for measuring internal conversion coefficients. All of these methods have areas where they are applicable. It is often necessary to pick the method most suitable for the particular internal conversion coefficients under investigation. Only those methods most widely used for high precision
will be mentioned here.

In the internal-external conversion method a beta-ray spectrometer is used to determine the internal conversion electron relative intensities. The gamma-ray relative intensities from the same source are then determined by the external conversion method, which will be described in Section D of the Introduction. The internal conversion coefficients can then be determined from the ratios of the electron intensities to the gamma-ray intensities. Internal conversion coefficients have been measured to five percent with this method (5, 6).

It is possible, in cases where there is only one gamma-ray transition, to determine the K internal conversion coefficient by measuring the ratio of $N_X^K$, the number of K X-rays emitted following internal conversion to the $N_\gamma$ number of gamma rays. The K internal conversion coefficient $\alpha_K$, can then be calculated from

$$\alpha_K = \frac{N_X^K}{\omega_K N_\gamma}, \quad \text{Equation 1}$$

where $\omega_K$ is the probability that a vacancy in the K shell is filled under emission of K X-rays, and it is called the fluorescent yield of the K shell. The values of $\omega_K$ have been determined by fitting the observed data to a semi-empirical formula. These values have been tabulated by Wapstra et al. (7).

In some cases it is more convenient to measure the total transition rate, $N_\gamma + N_e$, by observing the rate of emission of particles which uniquely feed the transition. This can be done by gating the spectrum from the transition of interest by another particle which is in coincidence with that
transition. Then, along with either \( N_\gamma \) or \( N_e \), the total conversion coefficient can be determined.

Relative internal conversion coefficients can be determined from the ratios of relative internal conversion electron intensities and relative gamma-ray intensities. If a normalization constant can be determined, absolute conversion coefficients can be calculated. The internal conversion electron relative intensities can be measured to a few percent with magnetic beta-ray spectrometers. Gamma-ray relative intensities are often known to no better than five or ten percent. Thus, to determine accurately internal conversion coefficients with this method, the gamma-ray relative intensities must be measured to five percent or less.

The present investigation is concerned with the accurate measurement of gamma-ray and X-ray relative intensities and the application of these accurately determined intensities to the determination of internal conversion coefficients.

D. Some Experimental Methods of Measuring Gamma-ray Relative Intensities

Three methods have recently been used to obtain gamma-ray relative intensities with high accuracy. They are photoelectric conversion, crystal diffraction, and least-squares analysis of scintillation spectra.

Hultberg (8) has described in detail the photoelectric conversion method. In this method gamma rays, whose intensities are to be measured, pass into a converter, a substance with a high atomic number, which is mounted in the source position of a magnetic beta-ray spectrometer. The gamma rays eject K, L, and M electrons from the atoms in the converter. The energy of the electrons is given by
If the resolution is good and the converter not too thick, the shape of the distribution of photoelectrons yielded from the K shell will approximate the shape of the transmission curve of the spectrometer. The procedure is to take a series of counts at a sufficient number of settings of the magnetic field to determine the profile of the line. If the number of counts received per unit time at the field \( B \) is \( N \), and since the momentum interval accepted by a magnetic spectrometer is proportional to \( B \rho \), it follows that

\[
N(B\rho) \, d(B\rho) = (N/B\rho) \, d(B\rho),
\]

where \( n(B) \) is the number of counts per momentum interval. A plot of \( N/B \) vs. \( B\rho \) is made. The area under the line is

\[
\int \text{line} \left( \frac{N}{B\rho} \right) \, d(B\rho) = \text{const} \, \tau_K(E_\gamma) \, f(E_\gamma) = A,
\]

where \( \tau_K(E) \) is the photoelectric cross section from the K shell and \( f(E_\gamma) \) is the fraction of all K photoelectrons at energy \( E \) detected by the spectrometer. The \( f(E_\gamma) \) depends on the particular source and the converter geometry and is very difficult to determine.

For this method, intense thin sources are needed. This method takes advantage of the high resolution of the beta-ray spectrometer. Using this method, gamma-ray relative intensities can be measured to about five percent.

The crystal diffraction method has been used by Lind et al. (9), Hatch (10), Bergvall (11), and Edwards and Boehm (12). In this method, a bent-crystal spectrometer is set at a diffraction maximum for a particular gamma ray. The intensity of the gamma ray is then proportional to the
intensity of the gamma ray is then proportional to the counting rate (13). Corrections must be made for absorption of the gamma rays in the air path between the source and detector, for the absorption of the cover of the detector, the half life of the source, the efficiency of the detector, the absorption of the gamma rays in the source itself, the absorption in the source container, the absorption in the diffraction crystal, and the energy dependence of the reflectivity of the diffraction crystal. The density of the source material is sometimes not known well and can contribute a large error. Also, unless an extensive study is made of the reflectivity of the diffraction crystal, a rather large error could be introduced by assuming an analytical expression for the energy dependence of the crystal reflectivity.

Edwards (13) has carried out an extensive study of the reflectivity of the diffraction from the (310) planes of a 2mm thick quartz crystal and has measured gamma-ray relative intensities with an uncertainty of less than five percent. For very weak gamma rays this method is often the only one available for intensity measurements. Because of the solid angle and the poor efficiency of the diffraction crystal, source strengths from 0.1 curies to several curies are needed. This method takes advantage of the high resolution of the bent-crystal spectrometer.

The least-squares scintillation method has been applied extensively to activation analysis as well as gamma-ray relative intensity measurements. This method has been developed by Reynolds (14), Trombka (15, 16), Heath (17), Ferguson (18), Salmon (19), Parr and Lucas (20) and McWilliams (21). A detailed discussion will be given of the linear least-squares method since it is basically this approach which was used in the present investigation.
The procedure used in this method is to expose a NaI(Tl) crystal to the source under investigation. The resulting light pulses are converted to electrical pulses in a photomultiplier and these electrical pulses are amplified and fed into a multichannel analyzer to obtain a counts vs. pulse-height spectrum. From a library of response functions for monoenergetic gamma rays, an interpolation is made to determine the response of the NaI(Tl) crystal for the particular energies contained in the source under investigation. A computer program is then applied to determine the gamma-ray relative intensities.

The linear least-squares method for determining gamma-ray relative intensities assumes that the complex gamma-ray pulse-height spectrum is a linear combination of response functions due to the presence of gamma rays of various energies.

The response functions depend on the various ways that gamma rays interact with the detector material. Below 1-MeV there are two ways in which gamma rays interact with matter. They are photoelectric absorption and Compton scattering. Photoelectric absorption is most important at low energies (below 500-keV) and Compton scattering is most important at higher energies (above 500-keV). Photoelectric absorption occurs when a gamma ray transfers all of its energy to an electron by ejecting the electron from a K, L, or M shell. The energy of the electron is given by Equation 2. After the electron is ejected from the atom, an outer shell electron will fill the vacancy causing emission of an X-ray or Auger electron as described in Section A of the Introduction.

Compton scattering is the process in which a photon interacts with an essentially free electron by transferring part of its energy to the electron
and scattering in such a way as to conserve energy and momentum. The energy, $E'_Y$, of the scattered gamma ray will be

$$E'_Y = \frac{E}{1 + \frac{E}{Mch} (1 - \cos \theta)}$$

where $\theta$ is the angle of the scattered gamma ray makes with the original direction of the gamma-ray photon and $E_Y$ is the energy of the incoming gamma ray.

In NaI(Tl), the electrons which have gained energy by photoelectric absorption or Compton scattering give rise to light pulses. The decay time of the light pulse in the crystal is longer than the interaction time of the gamma ray. Therefore, a gamma ray may be scattered several times and photoelectrically absorbed before the light pulse decays. To a first order approximation, the intensity of the light is proportional to the energy which the gamma ray loses in the crystal. A response function of a NaI(Tl) detector to a monoenergetic gamma ray of 444-keV is shown in Figure 1. It consists of a photopeak and the Compton continuum. The photopeak corresponds to the full energy of the incoming gamma ray regardless of the manner in which it transfers energy to the electrons. The maximum energy for the Compton scattering occurs when the gamma ray scatters through 180 degrees, and it is given by

$$E_C = E_Y - \frac{E}{2E_Y}$$

where $E_C$ is the maximum energy for Compton scattering, $E_Y$ is the energy of
the incoming gamma ray and $M_e^2$ is the rest mass of the electron.

Another feature which becomes evident below 100-keV is the iodine escape peak. Figure 2 shows a photopeak and an iodine escape peak for a 57-keV gamma ray. The escape peak is due to iodine X-rays escaping undetected from the NaI(Tl) crystal following photoelectric absorption.

The number of counts in the photopeak of each monoenergetic response function is related to the intensity of the gamma ray by

$$N_i = I_i t \frac{\omega}{4\pi} e^{-\mu_i d_i} e_i P_i \quad (22),$$

where $N_i$ is the number of counts in the photopeak of gamma ray $i$ with energy $E_i$, $I_i$ is the number of gamma rays of energy $E_i$ emitted per unit time, $t$ is the time the detector is exposed to the radioactive source, $\omega$ is the solid angle subtended by the crystal, $e^{-\mu_i d_i}$ is the fraction of gamma rays not absorbed before reaching the crystal, $e_i$ is the efficiency of the crystal, and $P_i$ is the ratio of the number of counts in the photopeak to the total number of counts in the response function. Thus,

$$\frac{I_i}{I_j} = \frac{N_i}{N_j} \frac{e^{-\mu_j d_j} e_j P_j}{e^{-\mu_i d_i} e_i P_i}.$$

Equation 3

The efficiency as a function of energy has been tabulated (23) for certain geometries or it can be calculated. The absorption coefficients, $\mu$, for the various materials between the source and detector are also tabulated (24, 25). The photopeak to total ratios must be experimentally determined in a scatter free geometry for the given source to crystal distance. The photopeak area is used in the method of Trombka (15, 16) to determine the gamma-ray intensity because it is least affected by scattering. The problem has
Figure 1. NaI(Tl) response function for a 444-keV gamma ray
Figure 2. NaI(Tl) response function for a 57-keV gamma ray
been thus reduced to determining $N_i/N_j$.

It will be shown in Section II that the sum of the squares of the difference between the experimental gamma-ray pulse-height spectrum and a linear combination of the normalized gamma-ray response functions will be a minimum when the $N_i$'s are the coefficients of the linear combination. Thus, a computer program to determine the best least-squares fit can be applied to determine the $N_i$'s.

To obtain the needed monoenergetic response functions, the photpeaks of the measured monoenergetic emitters are fit with Gaussians. The full width at half maximum is then determined as a function of energy. The shape of the Compton continuum is also determined as a function of energy. From this an interpolation is made to determine the count rate for each channel for the particular energy desired. A computer program is then used to determine $N_i/N_j$. The only corrections which are necessary are the energy dependence of the efficiency of the detector and the absorption of the material between the source and the detector.

This method eliminates many of the corrections involved in the crystal diffraction method and the external conversion method. Using this method, gamma-ray relative intensities have been measured with errors of three to fifteen percent. One serious disadvantage of this method is the limited number of monoenergetic emitters. This often necessitates interpolation over a large energy range.

The method used in the present investigation was developed by Brown and Hatch (22, 26). It uses the better features of the least-squares scintillation method and of the crystal-diffraction method. Basically it consists of measuring the monoenergetic response functions of the gamma rays.
of the source under investigation with a bent-crystal monochromator. A very thin line source is then placed on the focal circle of the bent-crystal spectrometer, and the diffraction crystal is removed. The collimator and detector of the spectrometer are rotated until a maximum in counting rate is observed. The composite pulse-height spectrum is then recorded in a multichannel analyzer. A linear least-squares computer program is applied to determine the relative intensities of the gamma rays. Brown and Hatch (26) found that the total response function could be used to determine the gamma-ray intensity rather than the photopeak area. This was due to the effectiveness of the collimator in reducing background. Thus, it was not necessary to know the photopeak to total ratio. They redefined the $P_i$'s in Equation 3 to be the curve to total ratio. The curve to total ratios correct for the counts between zero energy and the energy at which the fitting procedure began. The $P_i$'s were experimentally determined.

In summary, the present method experimentally measures the monoenergetic response functions for the gamma rays contained in the source under investigation. A linear least-squares analysis is then carried out to determine the relative intensities of the gamma rays contained in the observed pulse-height spectrum. Corrections are then applied for the efficiency of the detector, the absorption between the radioactive source and the detector and the curve to total ratio.
II. THEORY OF THE LINEAR LEAST-SQUARES SCINTILLATION METHOD

In this section the equations for the least-squares procedure will be derived following the method of Trombka (15). Let the composite gamma-ray pulse-height spectrum be represented by $R_i$ ($i = 1, \ldots, C$) where $R_i$ is the total number of counts in channel $i$ due to all $Q$ gamma rays, and $C$ is the number of channels used to record the composite spectrum. Let $a_{in}$ ($n = 1, \ldots, Q$) be the number of counts in channel $i$ of a normalized monoenergetic gamma-ray response function of energy $E_n$, normalized so the area under the response function is unity, i.e. $\sum_i a_{in} = 1$. Let $B_n$ be the area in the complex spectrum due to a gamma ray of energy $E_n$. Let $x_i$ be the independent random error in channel $i$ due to statistical fluctuations in $R_i$. Then, if the $a_{in}$ are assumed to be known without error,

$$R_i = \sum_{n=1}^{Q} a_{in} B_n + x_i$$

or

$$x_i = R_i - \sum_{n=1}^{Q} a_{in} B_n . \quad \text{Equation 4}$$

Now, assuming that the error $x_i$ is random, it can be shown (27, pp. 16-20) that the probability $p_i$ that there will be an error $x_i$ which lies between $x_i$ and $x_i + dx_i$ is given by

$$p_i = \frac{1}{\sqrt{2\pi} \sigma_i(R_i)} e^{-1/2 \left( \frac{x_i}{\sigma_i(R_i)} \right)^2} dx_i , \quad \text{Equation 5}$$
where $\sigma_i(R_i)$ is the standard deviation of $R_i$. The probability $P$ that $C$ errors will be observed such that $x_1$ is between $x_1$ and $x_1 + dx_1$ and $x_2$ is between $x_2$ and $x_2 + dx_2$ ... and $x_C$ is between $x_C$ and $x_C + dx_C$ will be a product of $C$ terms like Equation 5, since the measurement in a given channel is independent of the measurements in the other channels.

$$P = \prod_{i=1}^{C} p_i = e^{-\sum_{i=1}^{C} \frac{1}{2} \left( \frac{x_i}{\sigma_i(R_i)} \right)^2} \prod_{i=1}^{C} \frac{dx_i}{\sqrt{2\pi} \sigma_i^2(R_i)}$$ \hspace{1cm} \text{Equation 6}$$

The principle of maximum probability states that the most probable values of the $B_n$ are those values which maximize $P$. $P$ is a maximum when

$$\sum_{i=1}^{C} x_i^2 \prod_{i=1}^{C} \frac{1}{2\sigma_i^2(R_i)}$$

is a minimum. Substituting Equation 4 into this expression we are led to minimize

$$U = \sum_{i=1}^{C} \frac{(R_i - \sum_{n=1}^{Q} a_{in} B_n)^2}{2\sigma_i^2(R_i)}$$ \hspace{1cm} \text{Equation 7}$$

with respect to $B_k$. Taking partial derivatives with respect to the $B_k$ and setting them equal to zero we are led to

$$\frac{\partial U}{\partial B_k} = 0 = \sum_{i=1}^{C} \frac{(-2)(R_i - \sum_{n=1}^{Q} a_{in} B_n)(a_{iK})}{2 \sigma_i^2(R_i)}$$
\[
\frac{C}{\sum_{i=1}^{Q} \frac{a_{ik}B_i}{\sigma_i^2 (R_i)}} = \frac{1}{\sigma_i^2 (R_i)} (a_{ik})^T (R_i) - \sum_{n=1}^{Q} a_{in} B_n.
\]

Equation 8

These are the normal equations. Letting \( \omega_i = \frac{1}{\sigma_i^2 (R_i)} \) and \( a_{ki} = a_{ik} \), the normal equations become

\[
\sum_{i=1}^{Q} a_{ki} \omega_i R_i - \sum_{i=1}^{Q} a_{ki} \omega_i \sum_{n=1}^{Q} a_{in} B_n = 0.
\]

Equation 9

Rewriting Equation 9 in matrix notation we have

\[
(A^T W A) B = A^T W R
\]

or

\[
B = (A^T W A)^{-1} A^T W R
\]

Equation 10

for \( A^T W A \) nonsingular. In this equation \( A \) is a \( C \times Q \) matrix, \( W \) is a \( Q \times Q \) diagonal matrix with the weights on the diagonal, \( R \) is a \( C \times 1 \) column matrix and \( B \) is a \( Q \times 1 \) column matrix. The relative intensities can then be calculated by substituting \( B_i = N_i \) in Equation 3.

One of the major advantages of the linear least-squares method is that it enables one to obtain the standard deviation in the gamma-ray relative intensities. The derivation of the standard deviations of the \( B_i \)'s will be found in Appendix C along with the derivation of the equation used as a figure of merit. Only the results of these derivations will be quoted here.
The expected value of the matrix $R$ is given by

$$E(R) = \begin{pmatrix} E(R_1) \\ . \\ . \\ . \\ E(R_C) \end{pmatrix}$$

From the definition of covariance, the $ij$ element of the covariance matrix is given by

$$\text{cov}(R_i, R_j) = E[(R_i - E(R_i))(R_j - E(R_j))] .$$

The covariance matrix, $\text{cov}(R)$, has the variance of the $R_i$ on the diagonal and zero for the off diagonal elements because the fluctuations in each channel are assumed to be independent of those in any other channel. Thus

$$\text{cov}(R) = E\left[(R - E(R))(R - E(R))^T\right] .$$

It is assumed that $A$ and $W$ are known without error and thus $(A^TWA)^{-1}A^TW$ is known without error. Then, it will be shown in Appendix C that when $B = CR$, where $C$ is known without error,

$$\text{cov}(B) = \sigma^2 \left(A^TWA\right)^{-1} .$$

$\text{cov}(B)$ is a $Q \times Q$ matrix with the variances of the $B_i$ on the diagonal. It is also shown in Appendix C that an unbiased estimate of $\sigma^2$ is $S^2$ where
\[ S^2 = \frac{\sum_{j=1}^{C} \omega_j^2 \left( R_j - \sum_{k=1}^{Q} a_{jk} B_k \right)^2}{C - Q} \]

\( S^2 \) has a \( \chi^2 \) distribution and can be used as a figure of merit. The expected value of \( S^2 \) is 1.

The gamma-ray relative intensities can be then determined from

\[ \frac{I_i}{I_j} = \frac{N_i}{N_j} \frac{e^{-\mu_j d_j} \epsilon_j p_j}{e^{-\mu_i d_i} \epsilon_i p_i} \]

by looking up the \( \epsilon \)'s, \( P \)'s and \( \mu \)'s in tables and obtaining the \( N \)'s from the elements of \( B = (A^T W A)^{-1} A^T W R \). The standard deviations in the \( N \)'s are given by

\[ \sigma(N) = \sqrt{S^2 \left( (A^T W A)^{-1} \right)} \]

The standard deviations in the gamma-ray relative intensities are obtained from the fractional deviations of the gamma-ray relative intensities which are in turn determined from the square root of the sum of the squares of the fractional deviations of \( N, \epsilon, P \) and \( e^{-\mu d} \).
III. EXPERIMENTAL EQUIPMENT AND METHODS

The two meter bent-crystal spectrometer used in the present experiment is patterned after one described by Seppi et al. (28). Figure 3 is a schematic drawing and Figure 4 is a line drawing of the bent crystal spectrometer. It consists of five basic elements. These are a radioactive source, a bent diffraction crystal, a device for measuring the rotation of the diffraction crystal, a collimator to separate the direct beam from the diffracted beam, and a detector.

In the present experimental arrangement, the radioactive source consists of a quartz capillary approximately one inch long with an inside diameter varying between 0.002 and 0.020 inches. This capillary is filled with the material to be studied. The capillary is then irradiated with neutrons. Because of the solid angle involved and the poor efficiency of the diffraction crystal, sources from 0.1 curies to several curies, depending on the particular isotope under study, are needed. This is one of the limiting factors in determining which nuclei can be studied with a bent-crystal spectrometer.

After the source material has been irradiated, it is placed in a source holder which precisely positions it on the focal circle of the bent-crystal spectrometer. During the present investigation, two source holders were used. Figures 5 and 6 are line drawings of these source holders. The first source holder consists of two cylindrical lead pigs. The outer one is permanently fixed on the focal circle of the spectrometer and has a rotating shutter to allow the beam of gamma rays to reach the diffraction crystal or
Figure 3. Schematic drawing of the bent-crystal spectrometer
Figure 4. Line drawing of the bent-crystal spectrometer
to shield the source completely so personnel can move freely between the source and the diffraction crystal. The inner lead pig served as the source holder and a container for transporting the source. The quartz capillary containing the source material is held in a V groove by two spring clips. The bottom half of the inner pig can be raised or lowered to shield or expose the source. The inner pig is positioned in the stationary pig by three positioning screws. This source holder was very effective in working with long-lived sources.

The second source holder was designed to be used with short-lived materials. Boasso (29) has described this system in detail. Basically it consists of a rabbit made from beryllium metal and lexan plastic and a receiver to position accurately the rabbit on the focal circle of the spectrometer. Beryllium was chosen because of its small cross section for neutron capture (0.009 barns) and the long half life of the resulting activity ($2.7 \times 10^6$ years). Thus, for the irradiation times of interest, very little contaminating activity would be produced from the beryllium. Lexan was chosen for its high impact strength and for its ability to retain its strength after irradiation with neutrons. The receiver can be rotated in all directions for alignment of the source, as can be seen in Figure 6. Figure 7 shows a detailed drawing of the rabbit. It has a tapered nose cone and a key slot which match a similar taper and key in the receiver. This enables the source, which is contained in a V groove in the nose cone of the rabbit, to be repositioned to less than $25 \times 10^{-6}$ inches (29). The rabbit can then be placed in a closed loop with the reactor for fast transport via a pneumatic tube to and from the reactor. Since the rabbit may come out of the reactor with any orientation of the source, a means is necessary to
Figure 5. Original source holder used with the bent-crystal spectrometer
Figure 6. Line drawing of the source holder and transfer system used with the beryllium rabbit
rotate the rabbit until the source is vertical. This transfer system is shown in Figure 6. As the rabbit returns from the reactor it is slowed down and stopped by a plunger. This action trips a micro switch which starts a motor that rotates the rabbit until a pin falls into the key in the rabbit. At this time the source is vertical. An air cylinder then moves the rabbit over to the rear of the receiver and a vacuum system pulls the rabbit into the receiver. The whole process, from withdrawal from the reactor to the seating of the rabbit in the receiver, takes about 12 seconds. To irradiate the rabbit, a button is pushed which begins a sequence of withdrawing the rabbit from the receiver and moving it to the pneumatic tube to be sent into the reactor. Figure 8 is a photograph of the transfer system and the pneumatic tubes which are connected to the reactor.

If it is desired to study a nuclide which has a half-life of more than a few hours, it is necessary to irradiate the quartz capillary in one of the vertical thimbles of the reactor and then manually place the capillary in the nose cone of the rabbit. The rabbit is then placed in the transfer system which is moved behind the receiver where the vacuum system pulls the rabbit into the receiver.

In the present spectrometer, the diffraction crystal is bent to a radius of two meters as described by DuMond (30). The crystal is held between two clamping blocks which are machined to a radius of two meters. Two crystals were used in the present investigation. One was the \((400)\) planes of a single crystal of germanium 2.75 inches wide and 3 inches long and 1.4 mm thick. The other crystal was a single crystal of quartz 2.75 inches wide, 3 inches long, and 2 mm thick cut such that the \((310)\) planes were used for the diffraction. The quartz crystal was mainly used for X-ray measurements.
Figure 7. Line drawing of the beryllium rabbit
Figure 8. Photograph of the rabbit transfer system
while the germanium crystal was used for gamma-ray measurements.

The rotation of the diffraction crystal is controlled by a precision lead screw which is connected by an arm to the diffraction crystal. The screw is 15 inches long and has 40 threads per inch. The rotation of the screw is controlled by a Datex encoderdyne and control unit. The position of the screw is read out on a set of lights on the control unit to the nearest 0.001 revolution. The screw can be controlled in two ways. In the slewing mode, the encoder runs the screw at a continuous speed until a preset position is reached. This mode is useful in going quickly from one region to another. In the second mode, the encoder steps the screw in increments of 0.002, 0.005, or 0.010 revolutions until a preset position is reached. The 0.002 revolutions corresponds to a rotation of the diffraction crystal of approximately 0.4 seconds of arc. This mode is used when searching for diffraction peaks.

The collimator consists of 30 lead plates three inches high, 18 inches long, and 0.040 inches thick. The spaces between the plates are 0.040 of an inch near the diffraction crystal and are tapered such that if the center lines of the plates were extended, they would intersect at the source position. The collimator shields the detector from the intense undiffracted beam and is very effective in reducing scattering.

The detector consists of a Harshaw Integral Line Assembly Type 12S with a 3 inch x 3 inch NaI(Tl) crystal. The NaI(Tl) crystal has a resolution of 7.5 percent for 662-keV gamma rays. The detector is placed immediately behind the collimator and is shielded by two inches of lead.

The collimator and detector rest on a table which is constrained to rotate through an angle $2\theta$ as the diffraction crystal rotates through $\theta$ in
accordance with the mirror law. This rotation is accomplished by a gear reduction by a factor of two and a selsyn generator and receiver. This permits the diffraction crystal and source to be mechanically isolated from the detector. The diffraction crystal and source are isolated from vibrations in the floor by a concrete block one foot thick. This block rests on springs and rubber stoppers. The rubber stoppers damp out any oscillations of the concrete block. The collimator also is on a concrete block one foot thick but this block is rigidly attached to the floor since small oscillations do not effect the performance of the collimator. Figure 9 is a photograph of the bent-crystal spectrometer.

The electronic components consist of a RIDL Model 10-17 transistorized preamplifier, a RIDL Model 30-19 linear amplifier, and a RIDL Model 34-12B 400 channel multichannel analyzer. A RIDL Model 54-6 time base generator selected the counting interval for each screw setting while stepping over the diffraction peaks. A RIDL Model 33-10 single channel analyzer was used to select out the region of interest for the multichannel analyzer. The output of the multichannel analyzer was either an IBM typewriter or a Tally punch paper tape. The paper tape was converted to IBM cards on an SDS 910-IBM 1401 computer system. The power for the photomultiplier was supplied by a Fluke Model 405B high voltage power supply. All of the electronic components were connected to a Stabiline regulated power supply. Figure 10 is a block diagram of the experimental equipment.

To measure the monoenergetic response functions it was necessary to determine what settings of the lead screw corresponded to the diffraction maxima for the gamma rays contained in the source material. This was done in the following way. From a rough energy calibration for the particular
Figure 9. Photograph of the bent-crystal spectrometer and associated equipment
Figure 10. Block diagram of the bent-crystal spectrometer and associated equipment
diffraction crystal being used, the approximate location of the gamma rays could be determined. Searches were made in these energy regions to determine the exact location of the diffraction peaks. This was done by gating the multi-channel analyzer in the time mode with the single channel analyzer whose window was set over the desired energy region. The control unit was operated in the stepping mode and the time base generator was set at the desired time interval. All of the counts reaching the detector which fell within the window of the single channel analyzer were recorded in the first channel of the multichannel analyzer. At the end of the time interval determined by the time base generator, the screw was stepped through the chosen increment and the counts were recorded at this new setting in the second channel of the multichannel analyzer. This process was repeated until a preset position was reached on the control unit. Thus, the number of counts vs. screw setting was displayed on the oscilloscope screen of the multichannel analyzer. From this display it was possible to determine the screw setting for the diffraction peak. This procedure was repeated until the settings for all of the gamma rays had been determined. The response functions were then recorded by setting the screw at the diffraction maxima and recording the resulting pulse-height spectrum. Background was accounted for by recording the pulse-height spectrum on both sides of the diffraction peak and averaging. This was a very effective way of subtracting background since only a very small rotation of the diffraction crystal is necessary to obtain the background position. Thus, the geometry is almost identical with that of the diffraction peak position.

After the response functions had been measured, a very thin source of the same source material was placed on the focal circle of the bent-crystal
spectrometer at point V in Figure 3. The diffraction crystal was removed and the collimator and detector were rotated until a counting rate maximum was observed. The gamma-ray pulse-height spectrum from this source was then recorded. Background was determined by going off the transmission maximum and recording the pulse-height spectrum. In this way the composite gamma-ray spectrum was recorded in essentially the same geometry as the monoenergetic response functions.

The only corrections that had to be applied were the absorption in the air path between the source and detector and the absorption due to the aluminum covering of the NaI(Tl) crystal. The thickness of the material covering the NaI(Tl) crystal was obtained from the Harshaw Chemical Company at the time of purchase of the detector. The efficiency as a function of energy for the present geometry was calculated by a numerical integration computer program which is described in Appendix B.

One of the major difficulties with the least-squares method which is also true of the present method is the necessity for stability of the electronic components while the data is being taken. Various analytical schemes (19, 20, 31) have been devised for correcting for gain shifts which might occur between the recording of response functions, but no analytical method has been devised to correct for gain shifts during the recording of a response function. Several companies manufacture pulse-height stabilizers which electronically correct for gain shifts both during and between the recording of the response functions. However, all of those depend on a peak which is always present in the gamma-ray spectrum. In the present case it is not practical to place a weak gamma-ray source near the crystal to supply this peak because of the large amount of Compton distribution which would be
present. One method which has been devised is to put an Am$^{241}$ alpha emitter in the NaI(Tl) crystal. There are very few counts below the alpha peak. However, this peak occurs at approximately 2.5-MeV in the gamma-ray spectrum and is much too high in energy for measurements where the maximum energy being studied is 500-keV as in the present case. Since in the present investigation data were recorded over a period of 5 to 10 hours, only short term stability was needed. Thus, it was practical to rely on the stability of the system during the recording of the data.

The effect of assuming that the monoenergetic response functions are known without error has been investigated by Parr and Lucas (20). In their test cases the response functions and the complex spectra had equal statistical errors. They found that the intensities of the components changed very little by including the statistical fluctuations in the response functions, but the goodness of fit did tend to decrease. They point out that this effect is normally even less important than it was in the test cases since in most practical applications, the response functions have smaller statistical errors than the complex spectra.
IV. MEASUREMENTS AND RESULTS

A. Internal Conversion Coefficients of the E2 Transitions in Yb$^{170}$ and Er$^{166}$

The K internal conversion coefficients of the E2 transitions in Yb$^{170}$ and Er$^{166}$ were measured by determining the ratio of the K X-rays to gamma rays.

The predominant mode of decay from Tm$^{170}$ and Ho$^{166}$ is by beta decay to a low-lying $2^+$ level in the daughter nucleus as is shown in Figure 11 (32, pp. 6-4-87, 6-4-36, 1964). Nuclear structure effects are expected to be negligible in these transitions. Church and Weneser (33) have shown that for enhanced E2 transitions the static and dynamic nuclear structure effects are very small. The K conversion coefficients for these transitions have been reported to be from 5 to 20 percent higher than the theoretical values (34-46). This investigation was undertaken to determine accurately these conversion coefficients.

1. Internal conversion coefficient of the 84.3-keV transition in Yb$^{170}$

The Tm$^{170}$ sources were obtained by irradiating pure Tm$^{169}$ in the Materials Testing Reactor at Arco, Idaho, in a neutron flux of $5 \times 10^{14}$ neutrons/cm$^2$/sec for 28 days. The line source consisted of 5 mg TmCl$_3$ in a quartz capillary 1 inch long and 0.008 inches inside diameter. The material for the composite source consisted of 3 mg of TmCl$_3$ in a quartz capsule. By the time the sources were used the line source had a strength of approximately one curie. Because of the cross section for neutron capture of Tm$^{170}$ (125 barns), considerable Tm$^{171}$ will be contained in the source material.
Figure 11. Decay schemes of $^{170}$Tm and $^{166}$Ho (32, pp. 6-4-87, 6-4-36, 1964)
along with the Tm$^{170}$. Since Tm$^{171}$ has a half-life of 1.9 years as compared to 127 days for Tm$^{170}$, the percentage of Tm$^{171}$ will increase with time. Tm$^{171}$ beta decays to a 67-keV level in Yb$^{171}$. Following internal conversion of this 67-keV level, Yb X-rays will be emitted. These X-rays will have the same energy as the X-rays following internal conversion in Yb$^{170}$ and will give erroneous results for the conversion coefficient. To eliminate the Tm$^{171}$ from the Tm$^{170}$ source material, the Tm$^{170}$ was isotopically separated from the Tm$^{171}$ in the Ames Laboratory Isotope separator. This separated source was 0.2 cm x 1.5 cm and was deposited on an aluminum foil which had a thickness of 1.75 mg/cm$^2$. The estimated strength of this source was 6 millicuries. Figure 12 displays a NaI(Tl) pulse-height spectrum taken with this separated source. This pulse-height spectrum consists of the 84.3-keV photopeak, X-ray photopeak, iodine escape peak due to the Yb X-rays, and a continuous gamma-ray spectrum called the bremsstrahlung spectrum. The bremsstrahlung spectrum results from absorption of the high energy beta particles. Also present in the source but not evident in Figure 12 are Er X-rays due to the K capture branch of Tm$^{170}$ to Er$^{170}$. The energy of the Er X-rays is very close to the energy of the Yb X-rays and falls under the same photopeak as the Yb X-rays.

One gamma ray and six X-ray monochromatic response functions were measured with the quartz diffraction crystal. They were the 84.3-keV gamma ray, Yb $K_{\alpha_1}$, Yb $K_{\alpha_2}$, Yb $K_{\beta_1,3}$, and Yb $K_{\beta_2}$ X-ray response functions. In addition to these, response functions for the Er $K_{\alpha_1}$ and Er $K_{\alpha_2}$ X-rays were measured. The six X-ray response functions could not be fit to the X-ray photopeak in the composite spectrum because of the large amount of overlap. The procedure used instead was to fix the ratio of the Yb $K_{\alpha_1}$
Figure 12. Yb$^{170}$ gamma-ray spectrum taken with an isotopically separated source.
X-rays to the Yb $K_\alpha$ X-rays at the values from the tables of Wapstra et al. (7). These tables were derived by reading the values from a graph drawn smoothly through the experimental values. Thus, a response function was obtained for the Yb $K_\alpha$ X-rays. Response functions for the Yb $K_\beta$ and Er $K_\alpha$ X-rays were obtained in the same manner. The Er $K_{\beta 1,3}$ and $K_{\beta 2}$ X-rays were too weak in intensity for the recording of response functions. A response function for the Er $K_\beta$ X-rays was needed, however, to obtain an accurate measurement of the Er X-ray intensity. A response function for the Er $K_\beta$ X-rays was obtained by interpolating from the Yb $K_\beta$ response function. The ratio of Er $K_\alpha$ to Er $K_\beta$ X-rays was then fixed at the value from the tables of Wapstra et al. (7). Figure 13 shows four of the response functions fit by the linear least-squares computer program.

To account for the bremsstrahlung, a thin source was made from $^{32}P$, a pure beta emitter, which had the same dimensions as the thin source. The pulse-height spectrum from this source was then recorded in the same manner as that of the $^{170}Tm$ thin source. This pulse-height spectrum was fit to the composite spectrum along with the X-ray and gamma-ray response functions.

Four sets of data were analyzed by the linear least-squares computer program described in Appendix D. The weighted averages for the four sets of data are

$$I(84):I(YbK_\alpha):I(YbK_\beta):I(ErK_\alpha+\beta) = 944\pm18:1000\pm20:263\pm10:58\pm11 \quad (47).$$

The weights used in computing the average values were the reciprocals of the squares of the estimated errors in the relative intensities for each measurement.
Figure 13. Yb$^{170}$ response functions used in the least-squares fitting procedure
Figure 14 shows the fit obtained. The smooth curve is the computed composite spectrum, and the points are the experimental spectrum. The dashed curves are the response functions, which add up to the composite spectrum. The lower curve is the deviation of the experimental spectrum from the computed spectrum divided by the square root of the counts in the experimental spectrum. As an added check on the fit in the X-ray region, the ratio of $K_B$ X-rays to $K_\alpha$ X-rays was determined. The experimental value was 0.263±0.011 while the expected value (7) is 0.258±0.007. The fluorescent yield from Wapstra et al. (7) for Yb is 0.937±0.005. The K conversion coefficient can then be determined from

$$a_K = \frac{N_X}{\omega_K N_Y}$$

where $\omega_K$ is the fluorescent yield for the K shell. The value obtained from the four sets of data was 1.43±0.04.

The total, the L, and the M+N... conversion coefficients can be determined using $I(K):I(L):I(M+N+...)$ conversion electron intensities of Hatch et al. (34),

$$I(K):I(L):I(M+N+...) = 35.7±0.5:100:33.5±0.5.$$

The total, the L, and the M+N+... conversion coefficients can then be calculated from

$$a_L = a_K \cdot \frac{I(L)}{I(K)}.$$
Figure 14. Yb$^{170}$ composite spectrum with computed composite spectrum and monoenergetic components.
Tm$^{170}$ SEPARATED SOURCE

- **EXPERIMENTAL SPECTRUM**
- **CALCULATED SPECTRUM**
- **RESPONSE FUNCTION**

**Yb K$^\alpha$ X-RAYS**

**Yb K$^\beta$ X-RAYS**

**ESCAPE PEAK**

**Er K$^\alpha+\beta$ X-RAYS**

**BREMSSTRAHLUNG**

**CHANNEL NUMBER**

- **COUNTS x 10$^3$**
\[ \alpha_{M+N+...} = \alpha_K \cdot \frac{I(M+N+...)}{I(K)} \]

\[ \alpha_T = \alpha_K + \alpha_L + \alpha_{M+N+...} \]

and were determined to be

\[ \alpha_L = 4.01 \pm 0.12, \quad \alpha_{M+N+...} = 1.34 \pm 0.05 \quad \text{and} \quad \alpha_T = 6.78 \pm 0.14. \]

It was also possible to determine the K-capture branching ratio to \( \text{Er}^{170} \) from the Er K X-ray intensity relative to the Yb K X-ray intensity. The total number of decays to the 84.3-keV level can be determined from

\[ B_{84}^- = \frac{\alpha_T}{\alpha_T} I_{84}^\gamma + I_{84}^\gamma. \]

The relative disintegration rate can then be determined from the branching ratio to the 84.3-keV level

\[ \beta^- = \frac{B_{84}^-}{B_{84}} \]

where \( B_{84} \) is the branching ratio to the 84.3-keV level and \( \beta^- \) is the relative disintegration rate. If the K X-rays from conversion on the K-capture side are ignored, the K capture branching ratio can be determined from

\[ B_{\text{Er}} = \frac{N_{\text{Er}} I_{84}^\text{Er}}{w_K I_{84}^\text{Er} (1 + \gamma_{84})}. \]
where $\omega_{K}^{\text{Er}}$ is the K-fluorescent yield for Er and $N_{X}^{\text{Er}}$ is the relative intensity of the Er X-rays with respect to the 84-keV gamma ray. The value obtained for the K-capture branch to Er$^{170}$ from this equation is 0.19%±0.04%. This value is in agreement with the value of Day (48) of 0.15%±0.05% and of Graham et al. (35) of less than 0.3%.

In Table 1 are given the measurements of the K conversion coefficient for the 84.3-keV transition in Yb$^{170}$ along with the methods used to determine these values. The theoretical values of Bhalla (49), Rose (1) and Sliv and Band (2) are given for comparison. The theoretical value of Bhalla was calculated for this transition while the values of Rose and Sliv and Band were interpolated from their tables. The present result is in agreement with most of the previous measurements but is five percent higher than the theoretical value of Bhalla and of Sliv and Band. The present result is in good agreement with the previous measurements of Hatch et al. (34) and Dingus et al. (36) which were obtained by two completely different methods. The value of Hatch et al. (34) was obtained by mixing the Tm$^{170}$ source material with Te$^{123m}$. A magnetic beta-ray spectrometer was used to measure the conversion electron relative intensities, and a bent-crystal spectrometer was used to measure the gamma-ray relative intensities. The absolute conversion coefficient for the 84.3-keV transition was then obtained by using the 159-keV transition in Te$^{123}$ for normalization. The value of Dingus et al. (36) was obtained by fitting analytical expressions with a non-linear least-squares computer program to experimental singles and coincidence spectra which were obtained with a well-type NaI(Tl) crystal. It should be noted that the value of Dingus et al. (36) was obtained with both TmCl$_3$ and pure Tm sources and that no difference in the value of the
Table 1. K conversion coefficient of the 84-keV transition in Yb$^{170}$

<table>
<thead>
<tr>
<th>Reference</th>
<th>Method</th>
<th>$\alpha_K$</th>
<th>$\alpha_{Total}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Present Result</td>
<td>$KX/\gamma$ (singles)</td>
<td>1.43±0.04</td>
<td>6.78±0.14</td>
</tr>
<tr>
<td>Hatch et al. (34)</td>
<td>Mag Spect/Bent-xtal Spect</td>
<td>1.47±0.09</td>
<td>6.96±0.24</td>
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<td>Dingus et al. (36)</td>
<td>$KX/\gamma$ (singles &amp; $\beta-\gamma$ coin)</td>
<td>1.47±0.05</td>
<td></td>
</tr>
<tr>
<td>Hooton (37)</td>
<td>$KX/\gamma$ (singles)</td>
<td>1.46±0.05</td>
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</tr>
<tr>
<td>Jansen et al. (52)</td>
<td>IEC</td>
<td>1.36±0.10$^a$</td>
<td></td>
</tr>
<tr>
<td>Jansen and Wapstra (50)</td>
<td>$KX/\gamma$ (singles &amp; $\beta-\gamma$ coin)</td>
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<td>Erman and Hultberg (53)</td>
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<td>1.37±0.07</td>
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<tr>
<td>Houtermans (51)</td>
<td>$KX/\gamma$ (singles)</td>
<td>1.34±0.08</td>
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</tr>
<tr>
<td>Thosar et al. (54)</td>
<td>$KX/\gamma$ ($\beta-\gamma$ coin)</td>
<td>1.31±0.08</td>
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<tr>
<td>Graham et al. (35)</td>
<td>$KX/\gamma$ (singles)</td>
<td>1.60±0.15</td>
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<tr>
<td>Liden and Starfelt (38)</td>
<td>$KX/\gamma$ (singles)</td>
<td>1.56±0.15</td>
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<td>Croft et al. (39)</td>
<td>$KX/\gamma$ (singles)</td>
<td>1.66±0.11</td>
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<tr>
<td></td>
<td>$KX/\gamma$ (singles with E$^-$KX coin)</td>
<td>1.52±0.07</td>
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<tr>
<td>Bisi et al. (40)</td>
<td>$KX/\gamma$ (singles)</td>
<td>1.69±0.02</td>
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<tr>
<td></td>
<td>$(KX)/\beta(\gamma)$($\beta-\gamma$ coin)</td>
<td>1.61±0.10</td>
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<tr>
<td>McGowan and Stelson (41)</td>
<td>$KX/\gamma$ (singles)</td>
<td>1.65±0.12</td>
<td></td>
</tr>
<tr>
<td>Bernstein (42)</td>
<td>coul excit-half life</td>
<td>1.41±0.11</td>
<td>6.7±0.4</td>
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<td>Fossan and Herskind (43)</td>
<td>coul excit-half life</td>
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<td>7.2±0.4</td>
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<td>Bhalla (49)</td>
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<td>1.36</td>
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<td>Sliv and Band (2)</td>
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<tr>
<td>Rose (1)</td>
<td></td>
<td>1.33</td>
<td></td>
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</table>

$^a$Originally published as 1.57±0.10$^{+0.10}_{-0.15}$, but corrected value is given in Ref. 50.
conversion coefficient was detected. Thus any effects of chemical bonding on the electron wave functions are very small. It has been suggested (50) that the higher values for the conversion coefficient were obtained with flat NaI(Tl) crystals and the lower values with well type NaI(Tl) crystals. However, the value of Dingus et al. (36), which is 10 percent higher than the theoretical value, was obtained with a well-type crystal. Several of the early singles measurements were not done with an isotopically separated source and thus the reported values for the K conversion coefficient is higher than it should be. Also the reported values of Graham et al. (35), Liden and Starfelt (38), Bisi et al. (40), McGowan and Stelson (41), Houtermans (51) and Hooton (37) have not been corrected for the presence of Er X-rays. This correction would lower these K conversion coefficients. A weighted average of the values of Dingus et al. (36), Hatch et al. (34) and the present result which were obtained by three different methods and which are in good agreement is presented here as an average experimental value for the conversion coefficient of the 84.3-keV transition in Yb\(^{170}\). This value is

\[ \alpha_K = 1.45 \pm 0.04. \]

2. Internal conversion coefficient of the 80.6-keV transition in Er\(^{166}\)

The Ho\(^{166}\) sources were made by irradiating pure Ho\(^{166}\) in a neutron flux of \(3 \times 10^{13}\) neutrons/cm\(^2\)/sec for 24 hours in the Ames Laboratory Research Reactor. The line sources consisted of 19 mg of HoO\(_2\) in a quartz capillary 1 inch long and 0.015 inches inside diameter. The material for the thin source consisted of 0.3 mg of HoO\(_2\) in a quartz capsule. The thin source for the composite spectrum was made by depositing approximately 0.1 mg Ho\(^{166}\) on
a strip of aluminized mylar 0.00025 inches thick and 0.2 cm wide and 3 cm long. Figure 15 is a NaI(Tl) pulse-height spectrum of the low energy region taken with a Ho\textsuperscript{166} source.

As can be seen from Figure 11 (32, p. 6-4-36, 1964) there is a weak beta decay to high energy states in Er\textsuperscript{166} which lead to high energy gamma-ray transitions. However, the K conversion coefficient of the 80.6-keV level in Er\textsuperscript{166} can still be measured from the ratio of K X-rays to 80.6-keV gamma rays since the high energy gamma-ray transitions are very weak in intensity and conversion coefficients for high energy transitions are small and thus produce few X-rays. However, a correction should be made under the low energy portion of the spectrum for the Compton distribution due to the high energy gamma rays.

The thermal neutron cross section of Ho\textsuperscript{165} is much larger than that of Ho\textsuperscript{166} so very little Ho\textsuperscript{167} should be produced. Any Ho\textsuperscript{167} that is produced can be allowed to decay out since its half-life is 3.7 hours compared to 27 hours for Ho\textsuperscript{166}.

As can be seen in Figure 15 the pulse-height spectrum from Ho\textsuperscript{166} is very similar to the pulse-height spectrum from Tm\textsuperscript{170}. However, there has been no observed K-capture branch to Dy\textsuperscript{166} (54). Thus, the X-ray photopeak only consists of Er X-rays.

Monochromatic response functions for the 80.6-keV gamma ray, Er K\textsubscript{α1}, Er K\textsubscript{α2}, Er K\textsubscript{β1,3}, and Er K\textsubscript{β2} X-rays were measured with the quartz diffraction crystal. As in the Tm\textsuperscript{170} case, the ratios of the Er K\textsubscript{α1} to Er K\textsubscript{α2} and Er K\textsubscript{β1,3} to Er K\textsubscript{β2} were fixed at the values from the tables of Wapstra et al. (7). Three of the response functions used in the fitting procedure are displayed in Figure 16.
Figure 15. $^{166}$Er gamma-ray spectrum
Figure 16. Er$^{166}$ response functions used in the least-squares fitting procedure
The Compton distribution from the high energy gamma rays could not be determined directly because of their weak intensity and high energy. Instead, the Compton distribution from a Co$^{60}$ source was recorded with the diffraction crystal removed and the collimator set at the transmission maximum. Because this Compton distribution was small, it was subtracted out before the fitting procedure was applied. The amount to be subtracted was determined from the photopeak to total ratio for the Co$^{60}$ source and from the intensity of the high energy gamma rays in the Ho$^{166}$ source.

To check on the contribution of Ho$^{167}$ in the source material, data were taken two hours after irradiation and 24 hours after irradiation. No difference in the X-ray to gamma-ray ratio was detected.

Five sets of data were analyzed by the least-squares computer program which is described in Appendix D. The weighted averages for the five sets of data are

$$I(80):I(K_\beta):I(K_\alpha) = 781\pm14:253\pm9:1000\pm17$$

Again the weights were the reciprocals of the squares of the estimated errors in the intensities for each measurement. A least-squares fit to the data is shown in Figure 17. The experimental value of the ratio of $K_\beta$ X-rays to $K_\alpha$ X-rays was $0.253\pm0.010$, and the expected value (7) is $0.253\pm0.007$. The $K$ conversion coefficient obtained from

$$\alpha_K = \frac{N_X}{\omega_K N_Y}$$

was determined to be $\alpha_K = 1.72\pm0.06$. The fluorescent yield, $\omega_K$, from Wapstra et al. (7) was $0.932\pm0.005$. In Table 2 is given the present value along
Figure 17. $^{166}$Er composite spectrum with computed composite spectrum and monoenergetic components
Table 2. K conversion coefficient of the 80-keV transition in Er$^{166}$

<table>
<thead>
<tr>
<th>Reference</th>
<th>Method</th>
<th>$\alpha_K$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Present Result</td>
<td>$KX/\gamma$ (singles)</td>
<td>1.72±0.06</td>
</tr>
<tr>
<td>Sunyar (44)</td>
<td>$KX/\gamma$ (singles)</td>
<td>1.9±0.2</td>
</tr>
<tr>
<td>McGowan and Stelson (41)</td>
<td>$KX/\gamma$ (singles)</td>
<td>1.85±0.13</td>
</tr>
<tr>
<td>Helmer and Burson (55)</td>
<td>$KX/\gamma$ (singles)</td>
<td>1.7±0.3</td>
</tr>
<tr>
<td>Marklund et al. (45)</td>
<td>$KX/\gamma$ (singles)</td>
<td>1.76±0.15</td>
</tr>
<tr>
<td>Foglio and Bettoni (46)</td>
<td>$KX/\gamma$</td>
<td>1.75±0.07</td>
</tr>
<tr>
<td>Thosar et al. (54)</td>
<td>$KX/\gamma$ (singles and $\beta-\gamma$ coin)</td>
<td>1.67±0.07</td>
</tr>
<tr>
<td>Erman and Hultberg (53)</td>
<td>IEC</td>
<td>1.68±0.15</td>
</tr>
<tr>
<td>Theoretical</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bhalla (49)</td>
<td></td>
<td>1.62</td>
</tr>
<tr>
<td>Sliv and Band (2)</td>
<td></td>
<td>1.66</td>
</tr>
<tr>
<td>Rose (1)</td>
<td></td>
<td>1.60</td>
</tr>
</tbody>
</table>

with the previously determined values of the K conversion coefficient for the 80.6-keV transition in Er$^{166}$. The theoretical values of Bhalla (49), Rose (1) and Sliv and Band (2) are given for comparison. The value of Bhalla was calculated for this transition, while the values of Rose and Sliv and Band were interpolated from their tables. The present value is in agreement with all of the previous measurements but is six percent higher than the theoretical value of Bhalla (49).
3. Discussion of the E2 internal conversion coefficients in Yb$^{170}$ and Er$^{166}$

The present results for the K internal conversion coefficients for the 84.3-keV transition in Yb$^{170}$ and the 80.6-keV transition in Er$^{166}$ are about 5 percent higher than the theoretical values. While this is not a significant difference, it is interesting to note that they are of the same magnitude and in the same direction. The 10 to 20 percent deviations which had previously been reported were not observed. The present value for the conversion coefficient of the 84.3-keV transition in Yb$^{170}$ is in good agreement with the values reported by Dingus et al. (36) and Hatch et al. (34) which were determined by completely different methods. The present results for the 80.6-keV transition in Er$^{166}$ agrees, within the experimental error, with all of the values which have previously been reported. It is slightly higher, however, than the value reported by Thosar et al. (54) and Erman and Hultberg (53) which are in agreement with the theoretical value.

Recently Gelletly et al. (56) have reported the L subshell ratios for several E2 transitions including Yb$^{170}$ and Er$^{166}$. They report the $L_{II}/L_{III}$ subshell ratios agree with the corresponding theoretical values to less than 2 percent while the $L_{I}/L_{II}$ and $L_{I}/L_{III}$ ratios are about 5 percent higher than the theoretical values. This deviation is the same magnitude and in the same direction as the deviation of the K conversion coefficients for these transitions. A possible explanation of the difference between the experimentally determined and theoretically calculated $L_{I}/L_{II}$ and $L_{I}/L_{III}$ subshell ratios and the difference between the experimental and theoretical K conversion coefficients is that the s electron wave functions are altered.
due to the s electrons penetrating the nucleus. The p and d electron wave functions are not altered since these electrons do not penetrate the nucleus.

B. Internal Conversion Coefficients in Hf$^{180}$

The level structure in the nucleus Hf$^{180}$ following the decay of Hf$^{180m}$ is shown in Figure 18 (32, p. 6-6-121, 1965). An interesting feature of this decay is that the decay of the 641-keV level consists of three E2 transitions in cascade with no observed crossover transitions. This means that the total transition intensities of these three transitions are equal. It is possible to use this fact to determine the total internal conversion coefficient, $\alpha$, for the 93.3-keV transition from a measurement of the gamma-ray relative intensity of this transition along with that of the 332-keV transition. Measurements of the gamma-ray relative intensities and internal conversion electron relative intensities for all of the other observed transitions enable the determination of the internal conversion coefficients for the transitions using the previously determined coefficient ($\alpha_1$) for the 93.3-keV transition for normalization.

Edwards and Boehm (57) have carried out precise measurements of the gamma-ray and internal conversion line relative intensities and have obtained accurate values for the Hf$^{180}$ internal conversion coefficients through a least-squares adjustment of their data. They report that the K-shell internal conversion coefficient for the 93.3-keV transition was in agreement with theory and those for the 215.3-, 332.5-, and 443.8-keV transitions were approximately 10 percent lower than the theoretical coefficients of Rose (1). Although such a discrepancy could not be considered very significant, the fact that the deviation is of the same size and in the same
Figure 18. Decay scheme of Hf$^{180m}$ (32, p. 6-6-121, 1965)
direction for each of the latter three E2 transitions is interesting. This deviation is larger than would be expected based on most of the recent results for high precision determinations of $\alpha_K$ for pure E2 transitions.

1. Analysis of the Hf$^{180}$ X-ray and gamma-ray spectrum

Figures 19 and 20 show NaI(Tl) and Ge(Li) pulse-height spectra from the decay of 5.5 hour Hf$^{180m}$. To obtain the relative intensities of the X-rays and gamma rays, the spectrum was divided into two overlapping sections. The region from 93.3-keV to 501-keV will be called the "gamma-ray" portion of the spectrum, and the region up to 215-keV will be called the "X-ray" portion of the spectrum. In the gamma-ray portion the relative intensities of the gamma rays from 93.3-keV through 501-keV were determined, while in the X-ray portion the relative intensities of the X-rays and 57-keV gamma ray were determined using the 93.3-keV and 215-keV gamma rays for normalization. The line sources for the two sections were made by filling quartz tubes, which were about 1 inch long with an inside diameter of about 0.2 mm, with approximately 12 mg of HfO$_2$. The material for the gamma-ray portion of the spectrum was enriched to 57 percent in Hf$^{179}$ and contained 30 percent Hf$^{180}$, while the material for the X-ray portion of the spectrum was enriched to 87 percent in Hf$^{179}$ and contained 8.6 percent Hf$^{180}$. The materials for the sources were irradiated with neutrons in the Ames Laboratory Research Reactor in a neutron flux of $7 \times 10^{13}$ neutrons/cm$^2$/sec for 10 hours. The 5.5 hour Hf$^{180m}$ activity was obtained from single neutron capture by the Hf$^{179}$. The 45 day Hf$^{181}$ activity was also present in the sources. The time from the reactor shut down until data were taken was approximately two hours. Data could be taken during approximately two half lives for each
Figure 19. Hf$^{180}$ gamma-ray spectrum taken with a NaI(Tl) detector.
Figure 20. Hf$^{180}$ gamma-ray spectrum taken with a Ge(Li) detector.
source. The thin source for the gamma-ray portion was made by depositing approximately 0.6 mg of Hf$^{180m}$ source material on a strip of 0.00025 thick aluminized mylar over an area of 0.2 cm x 3 cm. The thin source for the X-ray portion was made in a similar fashion except 0.3 mg of Hf$^{180m}$ was deposited on the mylar. In order to correct for the presence of Hf$^{181}$ in the source material, a Hf$^{181}$ source was prepared from neutron capture of Hf$^{180}$. This source was allowed to decay for several days to allow any Hf$^{180m}$ to decay out.

The response functions for the 93.3-keV, 215-keV, 333-keV, 444-keV and 501-keV transitions were obtained with the germanium diffraction crystal and are shown in Figure 21. The pulse-height spectrum from the Hf$^{181}$ source was recorded in the same manner as the composite spectrum. This spectrum was then fit to the observed composite spectrum as a response function in the same manner as the monochromatic response functions. Six sets of data were analyzed with the linear least-squares computer program. Figure 22 displays the composite spectrum along with the response functions and the calculated composite spectrum. The bottom curve again shows the deviation of the experimental composite spectrum from the calculated spectrum. The results of the least-squares fitting yielded the relative intensities which are presented in Table 3.

In the X-ray portion of the spectrum, the $K_{\alpha 1}$, $K_{\alpha 2}$, $K_{\beta 1,3}$, $K_{\beta 2}$ X-ray and the 57-keV gamma-ray response functions were obtained with the quartz diffraction crystal while the response functions for the 93.3-keV and 215-keV gamma rays were obtained with the germanium diffraction crystal. As in the other X-ray intensity measurements, response functions for the $K_{\alpha}$ and $K_{\beta}$ X-rays were obtained by fixing the ratios of the $K_{\alpha 1}$ to $K_{\alpha 2}$ and
Figure 21. Hf$^{180}$ response functions used in the least-squares fitting procedure from 90-keV to 510-keV
The graph shows a gamma-ray spectrum for $^{181}$Ta. The peaks at 126 keV, 345.8 keV, 482 keV, 215 keV, 332 keV, 444 keV, and 501 keV are labeled. The x-axis represents channel number, and the y-axis represents counts x10^3.
Figure 22. Hf$^{180}$ composite spectrum from 90-keV to 510-keV with computed composite spectrum and monoenergetic components.
Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS

180m

Hf$^{180m}$ GAMMA-RAY SPECTRUM

CALCULATED COMPOSITE SPECTRUM

EXPERIMENTAL = ""

RESPONSE FUNCTIONS
Table 3. Relative intensities of the gamma rays above 90-keV from the decay of Hf$^{180m}$

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>Intensity</th>
</tr>
</thead>
<tbody>
<tr>
<td>501.3</td>
<td>136±12</td>
</tr>
<tr>
<td>443.8</td>
<td>904±30</td>
</tr>
<tr>
<td>332.5</td>
<td>1000±25</td>
</tr>
<tr>
<td>215.3</td>
<td>865±20</td>
</tr>
<tr>
<td>93.3</td>
<td>180±5</td>
</tr>
</tbody>
</table>

K$_{\beta_1,3}$ to K$_{\beta_2}$ X-rays at their respective values from the tables of Wapstra et al. (7). These response functions are shown in Figure 23. Figure 24 shows the low energy pulse-height spectrum obtained with the thin source which was enriched to 87 percent in Hf$^{179}$. It can be seen that the Hf$^{181}$ contribution is considerably reduced over what it was in Figure 19. This enabled the Hf$^{180}$ X-ray and 57-keV gamma-ray intensities to be determined to a higher degree of precision. The contribution from the Compton distribution from the higher energy gamma rays was determined by fixing the ratios of the Compton distributions from the higher energy gamma rays at the values obtained in the previous experiment and by fitting this distribution along with the other response functions. The Hf$^{181}$ spectrum was again used as a response function in the fitting procedure.

Five sets of data were analyzed with the least-squares computer program which is described in Appendix D. Figure 25 shows the composite spectrum
Figure 23. $^{180}$Hf response functions used in the least-squares fitting procedure below 230-keV
RESPONSE FUNCTIONS

Hf\textsuperscript{180} X-RAY

57-keV

Hf\textsuperscript{180} X-RAY

93.3-keV

215-keV

COUNTS x 10^{-3}

CHANNEL NUMBER
Figure 24. Hf$^{180}$ gamma-ray spectrum below 230-keV
Figure 25. Hf\(^{180}\) composite spectrum below 230-keV with computed composite spectrum and monoenergetic components
along with the response functions and the calculated spectrum. The experimental value for the ratio of $K_{\beta}$ X-ray to $K_{\alpha}$ X-rays was 0.254±0.019 and the expected value (7) is 0.263±0.005. The least-squares fitting procedure yielded the values for the intensities which are given in Table 4. The intensities of the 215-keV and 93.3-keV gamma rays were normalized to their values in Table 3, and this normalization constant was used to determine the relative intensities of the $K_{\beta}$ and $K_{\alpha}$ X-rays and the 57-keV gamma ray with respect to the higher energy gamma rays. The X-ray and gamma-ray relative intensities, normalized such that the 215-keV gamma ray has an intensity of 1000, are presented in Table 5 along with the values of Edwards and Boehm (57) which were obtained by the crystal-diffraction method. For comparison, the ratios of the values obtained by Edwards and Boehm (57) to the present values are given in the fourth column of Table 5. The agreement is within five percent except for the 501.3-keV transition. The present determination for this transition was a direct measurement while that of Edwards and Boehm (57) was inferred from the Hf$^{180m}$ decay scheme and from their other intensity measurements. The present value of 1.25±0.08 for the ratio of the 57-keV gamma ray to the $K$ X-rays is in agreement with the value of 1.6±0.5 obtained from critical absorption by Deutsch and Bauer (58).

The power of the present method for determining gamma-ray relative intensities is demonstrated by the unfolding of the 57-keV gamma ray from the $K_{\alpha}$ and $K_{\beta}$ X-rays. This was only possible because the intensity of the 57-keV gamma ray was approximately the same as the X-ray intensity. Added confidence in the resolving of the one photopeak into the three components, $K_{\alpha}$ X-rays, 57-keV gamma ray and $K_{\beta}$ X-rays, is obtained from the agreement between the experimental and expected ratio of $K_{\beta}$ to $K_{\alpha}$ X-rays.
Table 4. Relative intensities of the gamma rays below 250-keV from the decay of Hf\(^{180m}\)

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>Intensity</th>
</tr>
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<tbody>
<tr>
<td>215.3</td>
<td>1000±23</td>
</tr>
<tr>
<td>93.3</td>
<td>209±6</td>
</tr>
<tr>
<td>K(_{\beta})</td>
<td>96±6</td>
</tr>
<tr>
<td>57</td>
<td>595±23</td>
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<tr>
<td>K(_{\alpha})</td>
<td>379±17</td>
</tr>
</tbody>
</table>

Table 5. Hf\(^{180}\) gamma-ray relative intensities following the decay of Hf\(^{180m}\)

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>Present results</th>
<th>Edwards and Boehm(^a)</th>
<th>Edwards and Boehm(^a) Present results</th>
</tr>
</thead>
<tbody>
<tr>
<td>501.3</td>
<td>136±12</td>
<td>180±55</td>
<td>1.324</td>
</tr>
<tr>
<td>443.8</td>
<td>904±30</td>
<td>866±46</td>
<td>0.958</td>
</tr>
<tr>
<td>332.5</td>
<td>1000±25</td>
<td>1000±42</td>
<td>1.000</td>
</tr>
<tr>
<td>215.3</td>
<td>865±20</td>
<td>882±25</td>
<td>1.020</td>
</tr>
<tr>
<td>93.3</td>
<td>180±5</td>
<td>176±4</td>
<td>0.978</td>
</tr>
<tr>
<td>K(_{\beta})</td>
<td>83±5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>57</td>
<td>513±20</td>
<td>513±17</td>
<td>1.000</td>
</tr>
<tr>
<td>K(_{\alpha})</td>
<td>327±15</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\(^a\)Source: (57).
2. Results and discussion

Following is a description of how the present values for the Hf\textsuperscript{180} internal conversion coefficients were obtained. Because the final three transitions are in cascade with no crossover transitions, the total transition intensities for these transitions are equal. Thus we can write

\[ I_{332} (1 + \alpha_{332}^T) = I_{93} (1 + \alpha_{93}^T). \]

Therefore,

\[ \alpha_{93}^T = \frac{I_{332}}{I_{93}} (1 + \alpha_{332}^T) - 1, \]

where \( I_{332} \) represents the gamma-ray intensity of the 332.5-keV transition and \( \alpha_{332}^T \) the total internal conversion coefficient for the same transition. The notation is similar for the corresponding quantities for the 93.3-keV transition. Since \( \alpha_{332}^T \) has a value of about 0.060 as discussed later, and \( I_{332} / I_{93} \) is large, any uncertainty in \( \alpha_{332}^T \) has only a small effect in determining \( \alpha_{93}^T \). For example, an uncertainty of 10 percent in \( \alpha_{332}^T \) would lead to an uncertainty of 0.7 percent in computing \( \alpha_{93}^T \). The latter coefficient is determined mainly by the ratio of \( I_{332} \) to \( I_{93} \). Therefore, \( \alpha_{93}^T \) can be determined from measurements of the gamma-ray relative intensities compared with \( I_{332} \), along with a correction for \( \alpha_{332}^T \), which is relatively small. The value determined by Edwards and Boehm (57) of 0.060±0.004 was used for \( \alpha_{332}^T \). Since the error in this value is only 6.5 percent, it contributes an uncertainty of 0.5 percent, to the present determination of \( \alpha_{93}^T \). Thus \( \alpha_{93}^T = 4.91±0.23 \) was obtained. The same procedure was not used to determine \( \alpha_{215}^T \) since \( I_{332} / I_{93} \) is near one, and any uncertainty in the value...
of $\alpha_3^{332}$ has a large effect in determining $\alpha_2^{215}$. Once $\alpha_3^{93}$ has been determined, the corresponding internal conversion coefficients for the atomic shells can be obtained from K:L:M+N... ratios obtained by Edwards and Boehm (57). The remainder of the internal conversion coefficients for the 57-, 215.3-, 332.5-, 443.8- and 501.3-keV transitions were determined by taking the ratios of the internal conversion electron intensities of Edwards and Boehm (57) to the present values of the gamma-ray relative intensities. These ratios were then normalized using the value of $\alpha_3^{93}$ which was obtained from the present gamma-ray relative intensity measurements. Thus, the present conversion coefficients depend on the conversion line relative intensities of Edwards and Boehm (57) but have been computed using new measurements of the gamma-ray relative intensities and an independently obtained normalization constant. The conversion coefficients obtained in this manner (59) are presented in Figure 26 along with the coefficients obtained by Edwards and Boehm (57), Gvozdev et al. (60) and Scharff-Goldhaber and McKeown (61). The theoretical values obtained by interpolating values from the tables of Sliv and Band (2) are presented in the last column of the table. The theoretical values of Rose (1) are in good agreement with those of Sliv and Band (2). The large errors in the values of Gvozdev et al. (60) are probably due to the thick sources which were used due to the fact that they used natural Hf instead of enriched Hf. Since the present gamma-ray relative intensities agree with those of Edwards and Boehm (57), it is not surprising that the present internal conversion coefficients also agree closely.

In Figure 27 are displayed the ratios of the present determinations to the theoretical values of Sliv and Band (2) for the K-shell conversion coefficients of the Hf$^{180}$ E2 transitions. The errors were obtained by
Figure 26. Hf$^{180}$ internal conversion coefficients following the decay of Hf$^{180m}$
<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>Conversion Line</th>
<th>Conversion electron intensities (Edwards and Boehm)</th>
<th>Edwards and Boehm®</th>
<th>Present results</th>
<th>Conversion coefficients Gvozdev and Rusinov®</th>
<th>Scharff-Goldhaber and McKean®</th>
<th>Theoretical^d</th>
</tr>
</thead>
<tbody>
<tr>
<td>57.5</td>
<td>L_I + L_II</td>
<td>0.248±0.014</td>
<td>0.458±0.036</td>
<td>0.456±0.040</td>
<td>L_I 0.308±0.025</td>
<td>0.163 69</td>
<td></td>
</tr>
<tr>
<td></td>
<td>L_III</td>
<td>0.045±0.006</td>
<td>0.084±0.012</td>
<td>0.082±0.014</td>
<td>L_II 0.057±0.010</td>
<td>0.055±0.010 22</td>
<td></td>
</tr>
<tr>
<td></td>
<td>L_Total</td>
<td>0.29±0.012</td>
<td>0.543±0.036</td>
<td>0.541±0.040</td>
<td>0.33±0.10</td>
<td>0.225 91</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Total</td>
<td>0.378±0.013</td>
<td>0.698±0.045</td>
<td>0.696±0.048</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>93.3</td>
<td>K</td>
<td>0.205±0.012</td>
<td>1.10±0.09</td>
<td>1.05±0.09</td>
<td>Total 1.3±0.4</td>
<td>E2</td>
<td></td>
</tr>
<tr>
<td></td>
<td>L_Total</td>
<td>0.582±0.017</td>
<td>3.13±0.19</td>
<td>2.99±0.22</td>
<td></td>
<td>2.75</td>
<td></td>
</tr>
<tr>
<td></td>
<td>M+N+...</td>
<td>0.169±0.012</td>
<td>0.909±0.08</td>
<td>0.868±0.08</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Total</td>
<td>0.956±0.023</td>
<td>5.14±0.24</td>
<td>4.91±0.23</td>
<td></td>
<td>E2</td>
<td></td>
</tr>
<tr>
<td>215.3</td>
<td>K</td>
<td>0.114±0.005</td>
<td>0.123±0.009</td>
<td>0.122±0.009</td>
<td>0.15±0.05</td>
<td>0.137</td>
<td></td>
</tr>
<tr>
<td></td>
<td>L_Total</td>
<td>0.072±0.006</td>
<td>0.077±0.007</td>
<td>0.077±0.008</td>
<td></td>
<td>0.070</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Total</td>
<td>0.221±0.010</td>
<td>0.237±0.017</td>
<td>0.236±0.018</td>
<td></td>
<td>E2</td>
<td></td>
</tr>
<tr>
<td>332.5</td>
<td>K</td>
<td>0.040±0.0016</td>
<td>0.038±0.003</td>
<td>0.037±0.003</td>
<td>0.055±0.014</td>
<td>0.042</td>
<td></td>
</tr>
<tr>
<td></td>
<td>L_Total</td>
<td>0.015±0.0012</td>
<td>0.014±0.0015</td>
<td>0.014±0.0014</td>
<td></td>
<td>0.0132</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Total</td>
<td>0.063±0.0025</td>
<td>0.060±0.004</td>
<td>0.058±0.004</td>
<td></td>
<td>E2</td>
<td></td>
</tr>
<tr>
<td>443.8</td>
<td>K</td>
<td>0.017±0.0010</td>
<td>0.0189±0.0017</td>
<td>0.0177±0.0016</td>
<td>0.026±0.007</td>
<td>0.020</td>
<td></td>
</tr>
<tr>
<td></td>
<td>L_Total</td>
<td>0.004±0.0005</td>
<td>0.0044±0.0007</td>
<td>0.0041±0.0006</td>
<td>0.0063±0.0016</td>
<td>0.0049</td>
<td></td>
</tr>
<tr>
<td></td>
<td>M+N+...</td>
<td>0.0014±0.0028</td>
<td>0.0015±0.0003</td>
<td>0.0014±0.0003</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Total</td>
<td>0.0227±0.0012</td>
<td>0.0249±0.0022</td>
<td>0.0232±0.0020</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>501.3</td>
<td>K</td>
<td>0.0070±0.0009</td>
<td>0.0370±0.012</td>
<td>0.048±0.008</td>
<td>0.035±0.014</td>
<td>0.121 0.038</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Total</td>
<td>0.0104±0.0011</td>
<td>0.0549±0.018</td>
<td>0.071±0.011</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

^aSource: (57).
^bSource: (60).
^cSource: (61).
^dSource: (2).
^eThis value was obtained from the present gamma-ray intensities.
statistically adding the experimental error and a five percent error in the theoretical value. The $\alpha_K^{93}$ agrees closely with the theoretical value, but the previously observed deviation of the experimental from the theoretical values for the 215.3-, 332.5- and 443.8-keV transitions remains. In fact, while this deviation for the coefficients of Edwards and Boehm was 10 percent, the present coefficients are slightly lower and the deviation of the present values from the theoretical values is 11 percent for these three E2 transitions. This deviation is larger than has been recently found in other precise measurements for E2 transitions. Before any statement can be made about the possible theoretical origins of these deviations, such as due to the K electron wave functions overlapping the nucleus, an independent measurement of the conversion electron intensities would be necessary to determine conclusively if these deviations are real. The present experimental $\alpha_K$ for the 501-keV transition agrees closely with the theoretical $\alpha_K$ for an E3 multipolarity and is in agreement with the E3, M2 mixture obtained from an angular correlation experiment by Bodenstedt et al. (62) of 96.5 percent E3 and 3.5 percent M2. Scharff-Goldhaber and McKeown (61) have recently made accurate measurements of the $L_1$, $L_{II}$ and $L_{III}$ conversion coefficients for the 57-keV transition. They report the $L_1$ and $L_{II}$ coefficients are higher than the theoretical conversion coefficients for an E1 transition while the $L_{III}$ coefficient is in agreement with the theoretical value. They point out that no admixture of M2 can account for this difference. Paul et al. (63) have shown that the difference is not due to parity mixing. Scharff-Goldhaber and McKeown (61) conclude that the anomalously high $L_1$ and $L_{II}$ conversion coefficients are due to penetration effects in this extremely K-forbidden E1 transition.
Figure 27. Comparison of the present results for the K conversion coefficients of the E2 transitions in Hf$^{180}$ to the theoretical values of Sliv and Band (2)
C. K Internal Conversion Coefficients in Gd$^{155}$

The present investigation was carried out to measure accurately the gamma-ray relative intensities of the 105-, 86- and 60-keV gamma rays and the Gd X-rays from the decay of Eu$^{155}$ and to use these accurately determined intensities to determine the K conversion coefficients for the 105- and 86-keV transitions. The level structure exhibited by Gd$^{155}$ following Eu$^{155}$ decay is shown in Figure 28 (32, p. 5-5-52, 1963).

1. Analysis of the Gd$^{155}$ X-ray and gamma-ray spectrum

The line source for the bent-crystal spectrometer was made by double neutron capture of Eu$_2$O$_3$ which was enriched to 95 percent in Eu$^{153}$. A quartz capillary 1 inch long and 0.012 inches inside diameter was filled with 10 mg of enriched Eu$_2$O$_3$. The capillary was then irradiated with neutrons in the Materials Testing Reactor at Arco, Idaho, in a neutron flux of $5 \times 10^{14}$ neutrons/cm$^2$/sec for 28 days. The thin source for the composite spectrum could not be produced in the same manner because of the large amount of Eu$^{154}$ which would be present in the source material. Instead, Eu$^{155}$ material was purchased from Oak Ridge National Laboratory Isotope Sales Division. This material had been produced by beta decay to Eu$^{155}$ following single neutron capture of Sm$^{154}$. The source material had been allowed to decay for more than two years to allow the 15.2 day Eu$^{156}$ activity to die out. The Eu$^{156}$ had been produced by beta decay of 9.4 hour Sm$^{156}$ following double neutron capture of Sm$^{154}$ and by single neutron capture of Eu$^{155}$. The thin source was made by depositing the Eu$^{155}$ material on a strip of aluminized mylar 0.00025 inches thick and 0.2 cm x 3 cm. In Figures 29 and 30 are shown
Figure 28. Decay scheme of Eu$^{155}$ (32, p. 5-5-52, 1963)
Nal(Tl) and Ge(Li) spectra from the thin Eu$^{155}$ source. It can be seen in the Ge(Li) spectrum that there is a gamma ray at approximately 123-keV. This gamma ray is due to Eu$^{152}$ and Eu$^{154}$ present in the source material. Since the Eu$^{155}$ source material was purchased in solution from Oak Ridge, it was not possible to determine the amount of material deposited on the strip of mylar. To check the effect of the source thickness, a very thin source was made by evaporating Eu$^{155}$ onto 1.75 mg/cm$^2$ aluminum. This source was 0.75 inches long and 2 mm wide. The ratios of K X-rays to the 86-keV plus 105-keV gamma rays was determined. The ratio for the evaporated source was 0.52 while that for the drop source was 0.53. From this it was concluded that there was no appreciable effects due to the thickness of the drop source. The evaporated source was not used for the composite spectrum because of the long counting times that would be necessary due to the weak intensity of the source.

It can be seen from Figure 29 that the gamma-ray spectrum mainly consists of the 105-, 86- and 60-keV gamma rays and the Gd X-rays. Weak transitions at 26- and 45-keV are also present. The other transitions which are shown on the decay scheme are extremely weak and can be ignored when the gamma-ray relative intensities are determined. Response functions were measured for the 105-, 86- and 60-keV gamma rays and the $K_{\alpha_1}$, $K_{\alpha_2}$, $K_{\beta_1,3}$ and $K_{\beta_2}$ X-rays. $K_{\alpha}$ and $K_{\beta}$ response functions were again obtained by fixing the ratios of the $K_{\alpha_1}$ to $K_{\alpha_2}$ and $K_{\beta_1,3}$ to $K_{\beta_2}$ X-rays. Since the 26-keV gamma-ray intensity is small and since the absorption corrections for 26-keV are very large, no attempt was made to determine accurately the relative intensity of the 26-keV gamma rays. However, a response function was included to improve the fit to the experimental data. This response function
Figure 29. Gd$^{155}$ gamma-ray spectrum taken with a NaI(Tl) detector
Figure 30. \textsuperscript{155}Gd gamma-ray spectrum taken with a Ge(Li) detector.
was obtained from the shape of the photopeak of the $K_{\alpha_1}$ X-ray and from the energy calibration in the X-ray region. The 45-keV gamma ray lies between the $K_\alpha$ and $K_\beta$ X-rays. No attempt was made to unfold this gamma ray from the X-rays. The X-ray intensity was, however, corrected for the 45-keV gamma rays. Since the gamma rays at 123-keV are the most intense gamma rays in the decay of Eu$^{152}$ and Eu$^{154}$ and since the intensity of the 123-keV gamma rays in the Eu$^{155}$ source was small, no correction was made for the Compton distributions from higher energy gamma rays. A correction was made, however, for the Gd X-rays due to the conversion of the 123-keV transition.

The germanium diffraction crystal was used to obtain the response functions shown in Figure 31. Four sets of data were analyzed with the least-squares computer program. Figure 32 shows one of the fits obtained. The weighted average values are presented in Table 6 along with the previously reported values. The present X-ray intensities have been corrected for the 45-keV gamma rays and the Gd X-rays due to the conversion of the 123-keV transition. The 45-keV intensity was taken from Hatch and Boehm (64). The correction for the Gd X-rays from the conversion of the 123-keV transition was made in the following way. From the relative intensity of the 123-keV gamma ray which was obtained from the least-squares fitting procedure and from the K conversion coefficient for this transition, the number of K X-rays was calculated from $N_{123} = \omega_K^{123} \cdot I_{123}$, where $\omega_K$ is the K fluorescent yield. The corrections for the Eu$^{152}$ and Eu$^{154}$ X-rays and the 45-keV gamma rays were small and about equal. The ratio of $K_\beta$ to $K_\alpha$ X-rays was determined to be $0.234 \pm 0.012$, while the expected value from Wapstra et al. (7) is $0.244 \pm 0.007$. 
Figure 31. Gd$^{155}$ response functions used in the least-squares fitting procedure
Gd Kα X-RAYS

Eu¹⁵⁵ RESPONSE FUNCTIONS

Gd Kβ X-RAYS

60-keV

86-keV

106-keV

CHANNEL NUMBER

COUNTS x 10⁻³
Figure 32. $\text{Gd}^{155}$ composite spectrum with computed composite spectrum and monoenergetic components
Table 6. \textsuperscript{155}Gd gamma-ray relative intensities following the decay of \textsuperscript{155}Eu

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>Present results</th>
<th>Subba Rao\textsuperscript{a}</th>
<th>Hatch and Boehm\textsuperscript{b}</th>
<th>Vergnes\textsuperscript{c}</th>
</tr>
</thead>
<tbody>
<tr>
<td>105</td>
<td>68.3±2.7</td>
<td>68.5±5</td>
<td>64</td>
<td>65</td>
</tr>
<tr>
<td>86</td>
<td>100±3</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>60</td>
<td>4.3±0.3</td>
<td>5.0±0.6</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>K\textsubscript{B}</td>
<td>15.3±0.6</td>
<td>97.5±0.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>K\textsubscript{A}</td>
<td>65.6±2.2</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\textsuperscript{a}Source: (65).
\textsuperscript{b}Source: (64).
\textsuperscript{c}Source: (66).

2. Results and discussion

The present values for the gamma-ray intensities are in agreement, within the experimental errors, with all of the previously reported values except the lower of the two values reported by Subba Rao. The X-ray relative intensity presented here is 16 percent lower than the value reported by Subba Rao (65). Recently Subba Rao (4) has reanalyzed his data and has reported readjusted values for the conversion coefficients. The magnitude of the change would correspond to a change in his X-ray intensity which would bring it into good agreement with the present value.

The K internal conversion coefficients for the 86-keV and 105-keV transitions were determined in the following way. Since the energies of the
26-keV and 45-keV gamma rays are too low for K conversion, the K X-rays are
due predominantly to the 60-, 86- and 105-keV transitions

\[ N_X = N_X^{60} + N_X^{86} + N_X^{105} \]

Equation 11

where \( N_X \) is the total number of K X-rays and \( N_X^{60}, N_X^{86}, \) and \( N_X^{105} \) are the num-
ber of K X-rays due to 60-, 86- and 105-keV transitions respectively. The
number of K X-rays from the 60-keV transition can be determined from

\[ N_X^{60} = \alpha_K \omega_K I_{60} \]

Equation 12

where \( \omega_K \) is the fluorescent yield, \( I_{60} \) is the relative intensity of the 60-
keV gamma ray and \( \alpha_K^{60} \) is the K internal conversion coefficient for the 60-
keV transition. The \( \alpha_K^{60} \) can be obtained from the mixing ratio of 5/95
which has been determined from the Tb decay (67) and the theoretical con-
version coefficients of Rose (1). Using these values, \( \alpha_K^{60} \) is 6.89. Sub-
stituting this value into Equation 12 along with the fluorescent yield and
the intensity of the 60-keV gamma ray from Table 7, the number of X-rays
due to the 60-keV transition was determined to be 27.3±3.3. Using this
value in Equation 11, the number of X-rays from the 86- and 105-keV tran-
sitions was determined. The number of K conversion electrons for each of
these transitions was then determined from

\[ e_K^{86} + e_K^{105} = \frac{N_X^{86} + N_X^{105}}{\omega_K} \]

and the ratio of \( e_K^{86}/e_K^{105} \) obtained from Subba Rao (65). Using these electron
intensities and the gamma-ray relative intensities from Table 6, the K con-
version coefficients for the 86- and 105-keV transitions were determined and
are presented in Table 7 along with the values of Subba Rao (65) and the
Table 7. Gd$^{155}$K internal conversion coefficients following the decay of Eu$^{155}$

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>Present results</th>
<th>Subba Rao$^a$</th>
<th>Subba Rao$^b$</th>
<th>Theoretical$^c$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>E1</td>
<td>M2</td>
</tr>
<tr>
<td>105</td>
<td>0.23±0.03</td>
<td>0.29±0.054</td>
<td>0.23±0.03</td>
<td>0.21</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>11</td>
</tr>
<tr>
<td>86</td>
<td>0.43±0.06</td>
<td>0.49±0.075</td>
<td>0.35±0.04</td>
<td>0.37</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>26</td>
</tr>
</tbody>
</table>

$^a$Source: (65).

$^b$Source: (4).

$^c$Source: (2).

Theoretical values of Sliv and Band (2). The values in column 4 were reported to be from a re-analysis of the data reported in (65). It can be seen that the present value and the revised value of Subba Rao are in good agreement with the theoretical values for pure E1 transitions.

D. Relative Intensities of the 104-, 142- and 246-keV Gamma Rays in Eu$^{155}$

The relative intensities of the 104-, 142- and 246-keV gamma rays from the decay of 22 minute Sm$^{155}$ have been reported with values differing by 45 percent and with errors of 10 percent or more (68-73). The present investigation was undertaken to determine accurately the relative intensity of these gamma rays so they may be used to obtain the internal conversion coefficients for these transitions.
1. Analysis of the Eu\(^{155}\) gamma-ray spectrum

The level structure in the nucleus Eu\(^{155}\) following the decay of Sm\(^{155}\) is shown in Figure 33 (32, p. 5-5-51, 1963). The Sm\(^{155}\) beta decays to Eu\(^{155}\) with a half-life of 22 minutes and the Eu\(^{155}\) in turn beta decays to Gd\(^{155}\) with a half-life of 1.81 years. In Figure 34 is shown a NaI(Tl) pulse-height spectrum from the decay of Sm\(^{155}\).

Because of the short half-life of Sm\(^{155}\), it was not possible to irradiate the Sm\(^{154}\) and load it into the source holder manually as had previously been done. The procedure used instead was to use the rabbit system described in section III. The line source for the bent-crystal spectrometer consisted of 22 mg of Sm\(_2\)O\(_3\), enriched to 99.2 percent in Sm\(^{154}\), in a quartz capillary 1 inch long and 0.018 inches inside diameter. The quartz capillary was placed in the V groove of the nose cone of the beryllium rabbit which is shown in Figure 7. The rabbit was then inserted into the pneumatic tube and sent into the reactor. The sample was irradiated for 20 minutes in a neutron flux of \(9 \times 10^{12}\) neutrons/cm\(^2\)/sec. At the end of the twenty minute irradiation the rabbit was automatically brought down into the transfer system and on into the source position of the spectrometer as described in section III. The time from withdrawal of the rabbit from the reactor to the beginning of the recording of the response functions was approximately 12 seconds. The (400) planes of germanium were used to diffract the gamma rays. Background was recorded before and after the recording of the response function. After the response function for the 246-keV gamma ray was measured, the rabbit was sent back into the reactor for another 20 minutes of irradiation. The 142-keV gamma-ray response function was then recorded in
Figure 33. Decay scheme of Sm$^{155}$ (32, p. 5-5-51, 1963)
Figure 34. Eu$^{155}$ gamma-ray spectrum
the same manner as the 146-keV gamma ray. Immediately after the 142-keV response function was measured, the 104-keV response function was measured. The thin source for the composite pulse-height spectrum was made by irradiating 0.1 mg enriched Sm in solution for 20 minutes in a neutron flux of $4 \times 10^{13}$ neutrons/cm$^2$/sec. This source material was then deposited on a strip of aluminized mylar 0.00025 inches thick and 0.2 cm x 3 cm. Background for this source was recorded before and after the measurement of the composite spectrum. Because of the half-life of the source, no attempt was made to determine the relative intensities of the X-rays. Because of the difference in half lives of the Eu$^{155}$ and Sm$^{155}$ there was an extremely small amount of Eu$^{155}$ in the source. By allowing the 22 minute Sm$^{155}$ to decay away, it was found that there was a small amount of 47 hour Sm$^{153}$ present in the source material. As can be seen in Figure 35 the gamma-ray spectrum from the decay of Sm$^{153}$ mainly consists of a 103-keV transition. This gamma ray required a small correction to the intensity of the 104-keV gamma-ray when the Sm$^{155}$ data was taken immediately after irradiation.

2. Results and discussion

Figure 35 shows the three response functions used in the fitting procedure. Because of the poor statistics of the 142- and 246-keV gamma rays, seven sets of data were analyzed. In Figure 37 is displayed one of the fits obtained. The points are the experimental spectrum, and the smooth curve is the calculated spectrum. The dashed curves are the response functions which add up to the calculated spectrum. In this case only the low energy tails of the response functions can be distinguished from the experimental data and the calculated spectrum. The bottom curve is again the deviation
GAMMA-RAY SPECTRUM FROM THE DECAY OF Sm$^{153}$

Figure 35. Eu$^{153}$ gamma-ray spectrum
Figure 36. Eu$^{155}$ response functions used in the least-squares fitting procedure
Figure 37. $\text{Eu}^{155}$ composite spectrum below 250-keV with composite spectrum and monoenergetic components.
of the experimental spectrum from the calculated spectrum. The weighted averages for the gamma-ray intensities from the seven sets of data are presented in Table 8 along with the previously reported values. The present values for the intensity of the 104-keV and 246-keV gamma rays are in agreement with the values of Funke et al. (68) and Kracik et al. (69). The present value for the 142-keV gamma ray is higher than any of the previously reported values. It is interesting to note that the ratio of the 246-keV gamma ray to the 142-keV gamma ray for the present measurement and for the recent measurement of Potnis et al. (70) are in good agreement. The gamma-ray relative intensities of Funke et al. (68) and Potnis et al. (70) were obtained with Ge(Li) detectors. The 104-keV and 142-keV gamma rays are on top of the large Compton distribution from the 246-keV gamma rays. This can add a large uncertainty to the determination of the intensities of these gamma rays, particularly the 142-keV gamma rays.

Schmid and Burson (73) have measured conversion coefficients for the 104-keV and 142-keV transitions from beta-gamma coincidences. Kracik et al. (69) have measured the conversion coefficients for the 104-keV and 246-keV transitions by measuring the percentage of the total decays which are due to conversion electrons from the 104-keV and 246-keV transitions by measuring the conversion line intensities relative to the continuous beta spectrum with a magnetic spectrometer. The gamma-ray intensities can then be normalized by assuming the 104-keV and 246-keV transitions are the only ones which lead to the ground state. This means that the total transition intensity for these transitions must be 100 percent. Because of the half life of the source, this was a very difficult experiment to do. Both of
Table 8. Eu$^{155}$ gamma-ray relative intensities following the decay of Sm$^{155}$

<table>
<thead>
<tr>
<th>Reference</th>
<th>Energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>104</td>
</tr>
<tr>
<td>Present results</td>
<td>2000±90</td>
</tr>
<tr>
<td>Potnis et al. (70)</td>
<td>2000</td>
</tr>
<tr>
<td>Funke et al. (68)</td>
<td>2000±200</td>
</tr>
<tr>
<td>Kracik et al. (69)</td>
<td>2000</td>
</tr>
<tr>
<td>Funke et al. (71)</td>
<td>2000</td>
</tr>
<tr>
<td>Sund et al. (72)</td>
<td>2000</td>
</tr>
<tr>
<td>Schmid and Burson (73)</td>
<td>2000</td>
</tr>
</tbody>
</table>

the reported values for the 104-keV transition are higher than the theoretical prediction for a pure E1 transition, Schmid and Burson's being 29 percent higher and Kracik et al. being 76 percent higher. Also, Schmid and Burson's value for the 142-keV transition is 69 percent higher than the theoretical prediction for a pure E1 transition. The value of Kracik et al. (69) for the 246-keV transition is in agreement with a pure M1 transition. The errors in the reported values are from 22 percent to 40 percent. It is hoped that by using the present accurately determined gamma-ray intensities along with conversion electron intensities measured with a Si(Li) detector, accurate conversion coefficients can be determined for these transitions. A more meaningful comparison can then be made with the
theoretical predictions. The Si(Li) measurements are presently being carried out.*

E. Concluding Remarks

Gamma-ray and X-ray relative intensities and internal conversion coefficients from five different isotopes were measured and reported in this investigation. The gamma-ray and X-ray relative intensities were measured with uncertainties from 2 percent to 19 percent and the internal conversion coefficients had uncertainties from 3 percent to 14 percent. The K internal conversion coefficients of the E2 transitions in Yb$^{170}$ and Er$^{166}$ were measured in an attempt to clear up the controversy over these values. The present results for the K internal conversion coefficients for these transitions are approximately 5 percent higher than the theoretical values.

The relative intensities of the gamma rays from the decay of Hf$^{180m}$ determined in this investigation are in good agreement with the reported values of Edwards and Boehm (57) except for the 501-keV transition. The present value was a direct measurement while the value of Edwards and Boehm (57) was deduced from their other data. Because of the good agreement of the gamma-ray relative intensities and since the internal conversion electron ratios of Edwards and Boehm (57) were used in the present investigation, the values of the internal conversion coefficients reported here are

*These measurements are presently being carried out at the Ames Laboratory by D. F. Boneau.
in agreement with those of Edwards and Boehm (57). The 10 percent deviation in the K-conversion coefficients of the E2 transitions reported by Edwards and Boehm remains.

The gamma-ray relative intensities reported in the present studies of the 105-keV, 86-keV and 60-keV transitions from the decay of Eu$^{155}$ are in agreement with most of the reported values for these transitions. The K internal conversion coefficients for the 105-keV and 86-keV transitions determined in this investigation are in good agreement with the revised values of Subba Rao (4) and with the theoretical predictions for pure E1 transitions.

The gamma-ray relative intensities of the 104-keV and 246-keV transitions from the decay of Sm$^{155}$ reported in the present work are in agreement with the values for these transitions reported by Funke et al. (68) and Kracik et al. (69). The present value for the 142-keV transition is slightly higher than the previously reported values.
V. LITERATURE CITED


VI. ACKNOWLEDGMENTS

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VII. APPENDIX A: EFFECTS DUE TO SOURCE WIDTH AND POSITION

The present method for measuring the gamma-ray relative intensities assumes that the composite spectrum and response functions are measured under identical conditions. Since it was desirable to use a thin composite source that was wider than the line source and since the geometry with respect to the room was slightly different for the thin source and line source, an investigation was made to determine the effects of varying the thin source width and position. The procedure used was to record a "normal" pulse-height spectrum and a pulse-height spectrum under the condition being checked. The "normal" pulse-height spectrum was recorded with the thin source located at the position where the data which are described in this dissertation were recorded. The two spectra were then normalized to a constant area and subtracted. The type of effects looked for were due to scattering and a change in the photopeak to total ratio. For a constant source to crystal distance the effects due to scattering would be much larger than those due to a change in photopeak to total ratio. This is not necessarily true when the source to crystal distance is changed drastically.

If the differences fluctuate statistically around zero it was assumed that there was no observable difference in the spectra and that either source position was equally good. If the difference remains positive or negative over several channels it was assumed that there were effects due to either scattering and/or a change in the photopeak to total ratio. If these cases were used in the least-squares fitting procedure the goodness of fit parameter $S^2$, would be large and the data would be rejected as
The effect of varying the source width was determined by recording the composite spectra from Yb\textsuperscript{169} sources of 0.5 mm, 3 mm and 5 mm wide and subtracting each of these from a second spectrum obtained from the 0.5 mm wide Yb\textsuperscript{169} source. The spectra were normalized such that the total area was a constant. These differences are shown in Figure 38. The investigation of the scattering due to the slight difference in geometry between the composite source and the source used for the response functions was carried out in the following way. The pulse-height spectrum from a thin Eu\textsuperscript{155} source 2 mm wide was recorded at three different positions on the focal circle. These positions were the normal source position, two inches and six inches to the left of the normal source position. The spectra were again normalized so that the total area was a constant and were subtracted from the spectrum recorded at the normal source position. Figure 39 displays these differences.

Four other effects of source width and position were investigated. These are the effect of the thin source not being perpendicular to the collimator, scattering due to the crystal clamping block, the effect of not being at the transmission maximum and moving the source closer to the detector. This last effect was checked with the hope of being able to use much weaker sources. These effects were again determined by recording spectra from a thin Yb\textsuperscript{169} source and subtracting. Figure 40 shows these differences. The top curve was obtained from spectra from a 5 mm wide disk source recorded at 0\degree and rotated through 70\degree with respect to the collimator. The next curve was obtained from a 2 mm wide source without a clamping
Figure 38. Comparison of the gamma-ray spectra obtained with thin sources of three different widths
Figure 39. Comparison of the gamma-ray spectra obtained with the thin source in three different positions on the focal circle.
Figure 40. Comparison of the gamma-ray spectra obtained with the thin source located at its normal position on the focal circle and with it rotated through 70°, with a blank clamping block between the source and detector, with the collimator set at the transmission half maximum and with the thin source 130 cm from the detector.
Y\textsuperscript{90} GAMMA-RAY SPECTRUM

- Escape peak
- 95.4 keV
- 177.7 keV
- 198.4 keV
- 308.1 keV

Counts per 3

- Spectrum recorded without clamping block minus spectrum recorded with clamping block
- Spectrum recorded at transmission minimum minus spectrum recorded at transmission half maximum
- Spectrum recorded on focal circle (26 cm) minus spectrum recorded at 130 cm
block and with a clamping block which did not have a diffraction crystal. The third curve is the difference of spectra recorded at the transmission maximum and at half maximum. The bottom curve is the difference of spectra recorded from the source at 261 cm from the detector (on the focal circle) and at 130 cm. Because of the focussing properties of the collimator, the collimator plates were removed in the latter case.

As can be seen in Figures 38, 39, and 40 the difference for the various source widths and positions are statistical. Therefore, there is either very little scattering in these cases or the collimator does a very effective job in eliminating the scattering. Because of this, it was possible to keep the thickness of the thin source for the composite spectrum at a minimum by making it wider. The optimum width was approximately 2 mm. Also it seems that the assumption of identical geometries for the line source and the thin source is a good one. As can be seen from the lower two curves in Figure 40 the differences are not statistical in these cases. It appears to be critical that the collimator and detector are set at the transmission maximum. However, this can easily be done by recording the count rate as a function of detector position. Also, it appears that it is not possible to move the source closer to the detector to achieve higher counting rates.
VIII. APPENDIX B: CALCULATION OF THE EFFICIENCY OF THE NAI CRYSTAL

This appendix describes the calculation of the efficiency of a 3 inch x 3 inch NaI(Tl) crystal for a line source 1 inch long and 261 cm from the detector. These calculations were done in collaboration with Michael Yester.

The efficiency is defined as the fraction of the gamma rays which are emitted from a source that are detected by the NaI(Tl) crystal. The fraction of the particles with energy E that strike a crystal of thickness t and absorption coefficient \( \tau \) that will be absorbed is given by

\[
(1 - e^{-\tau(E)t})
\]

The efficiency for a line source is then given by

\[
T(E) = \frac{1}{4\pi L} \int_0^L \int_0^{2\pi} \int_0^{\Theta_{\text{max}}} (1-e^{-\tau(E)t}) \sin \Theta d\Theta d\Phi dx \quad \text{Equation 13}
\]

where \( 4\pi L \) is the total solid angle. Due to the symmetry, Equation 13 can be rewritten as

\[
T(E) = \frac{1}{4\pi L} (2) (2) \int_0^{L/2} \int_{-\pi/2}^{\pi/2} \int_0^{\Theta_{\text{max}}} (1-e^{-\tau(E)t}) \sin \Theta d\Theta d\Phi dx \quad \text{Equation 14}
\]

As can be seen in Figure 41 the integral over \( \Theta \) must be divided into two parts corresponding to a gamma ray exiting through the bottom or the side of the crystal. Therefore,
Figure 41. Source-detector geometry for a line source of length L and a distance $H_0$ from a NaI(Tl) crystal of thickness $t_0$ and a radius $r_0$. 
\[ T(E) = \frac{1}{4\pi L} \int_0^{L/2} \int_0^{\pi/2} \left[ \int_0^{\theta_1} (1-e^{-\tau_1}) \sin \theta d\theta \right. \]
\[ + \left. \int_{\theta_1}^{\theta_2} (1-e^{-\tau_2}) \sin \theta d\theta \right] d\phi dx . \quad \text{Equation 15} \]

First consider only the case in which the gamma ray leaves through the bottom of the crystal. Consider the line source as a series of point sources a distance \(x\) from the axis of the crystal, where \(0 \leq x \leq L/2\). \(\theta_1\) is the angle between the perpendicular from the point source to the crystal and the line PR in Figure 41. By finding \(P'R\), \(\theta_1\) can be determined from the relation

\[ \theta_1 = \tan^{-1} \left( \frac{P'R}{H_0 + t_o} \right) . \quad \text{Equation 16} \]

From triangle ORU in Figure 42 we have

\[ \overline{RU}^2 = \overline{OR}^2 - \overline{OU}^2 , \]

but \(\overline{OR}^2 = r_0^2\),

and \(\overline{OU}^2 = (x-P'U)^2\),

so \(\overline{RU}^2 = r_0^2 - (x+P'U)^2\),
therefore $RU^2 = r^2_0 - x^2 - 2xP'_U - (P'_U)^2$.  

Equation 17

From the triangle $P'RU$ in Figure 42

$RU^2 = P'R^2 - P'_U^2$.  

Equation 18

Combining Equations 17 and 18 we get

$r^2_0 - x^2 - 2xP'_U = P'R^2 - P'_U^2$,  

or $r^2_0 - x^2 - 2xP'_U = P'R^2$,  

but $P'_U = P'R \sin \phi$,  

so $r^2_0 - x^2 - 2xP'R \sin \phi = P'R^2$.  

Equation 19

Rearranging Equation 19 and solving for $P'R$ we obtain

$P'R = -x \sin \phi + \sqrt{x^2 \sin^2 \phi - (x^2 - r^2_0)}$.  

Equation 20

Using Equation 20 in Equation 16 we get

$\theta_1 = \tan^{-1} \left[ \frac{-x \sin \phi + \sqrt{x^2 \sin^2 \phi - (x^2 - r^2_0)}}{H_0 + t_0} \right]$.  

Equation 21

The distance $t_1$ a gamma ray will travel through the crystal can be found from Figure 41

$t_1 = SR$,  

but $SR = t_0 / \cos \theta$.  

Figure 42. Geometry used in the plane normal to the NaI(Tl) crystal to determine the equations for the efficiency
so \( t_1 = t_0 / \cos \theta \). \hspace{1cm} \text{Equation 22}

Now consider the case where the gamma ray exits through the side of the crystal. It is evident from Figure 46 that

\[ \theta_2 = \tan^{-1} \left( \frac{(O') Q}{H_0} \right) \], \hspace{1cm} \text{Equation 23}

but \( Q'O = P'R \).

Therefore, substituting Equation 20 into Equation 24 we have

\[ \theta_2 = \tan^{-1} \left[ \frac{-x \sin \phi + \sqrt{x^2 \sin^2 \phi - (x^2 - r_0^2)}}{H_0} \right] \]. \hspace{1cm} \text{Equation 25}

From Figure 41 it can be seen that

\[ t_2 = S'T = P'T - PS' \],

but \( P'T = \frac{P'T}{\sin \theta} = \frac{P'R}{\sin \theta} = \frac{-x \sin \phi + \sqrt{x^2 \sin^2 \phi - (x^2 - r_0^2)}}{\sin \theta} \)

and \( PS' = \frac{H_0}{\cos \theta} \)

so \( t_2 = \frac{-x \sin \phi + \sqrt{x^2 \sin^2 \phi - (x^2 - r_0^2)}}{\sin \theta} = \frac{H_0}{\cos \theta} \). \hspace{1cm} \text{Equation 26}

Combining Equations 21, 22, 24 and 26 with Equation 15 we have
\[
T(E) = \frac{1}{\pi L} \int_0^{L/2} \int_{-\pi/2}^{\pi/2} \frac{\tan^{-1}\left( \frac{-\xi \sin \phi + \sqrt{x^2 \sin^2 \phi - (x^2 - r_0^2)} - H_0}{H_0 + t_0} \right)}{\left[ 1 - e^{-\frac{\tau t_0}{\cos \theta}} \right] \sin \theta \, d\theta \, d\phi}
\]

Equation 27

where \( L \) = source length

- \( r_0 \) = radius of the NaI(Tl) detector
- \( \tau(E) \) = absorption coefficient for energy \( E \)
- \( t_0 \) = thickness of NaI(Tl) crystal.

Equation 27 was numerically integrated using the trapezoidal rule and an IBM 7074 computer. The input parameters for the computer program were \( r_0, t_0, H_0, L, E, \tau \) and the number of intervals to be used for each integral. The absorption coefficients \( \tau(E) \) were taken from the tables of Vegors et al. (23).
As a check on the accuracy of the computer program, a comparison was made with the calculations of Jegors et al. (23) for $H_0 = 10$ cm and $L = 0.75$ inches. The difference was less than 1 percent for energies above 50-keV. The differences were probably due to the different methods used in the numerical integration. When $H_0 = 261$ cm the variables change much more slowly and it is believed that the agreement would be even better at this distance.

The dependence of the efficiency on the source length and the distance between the source and crystal was determined. In Table 9 are given the efficiencies at four energies for source lengths of 0.25, 0.50 and 1.00 inches and a source to crystal distance of 261 cm. As can be seen, the efficiency is independent of the source length in the given ranges. The variation of efficiency as a function of source to crystal distance is given in Figure 43. At 500-keV the efficiency only changes by 0.02 percent for a variation in the source to crystal distance of 2 cm.

Table 9. The efficiency of a 3 inch x 3 inch NaI(Tl) crystal for a source to crystal distance of 261 cm as a function of energy and source length

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>Source Length</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.25&quot;</td>
</tr>
<tr>
<td>50</td>
<td>1.0000</td>
</tr>
<tr>
<td>150</td>
<td>0.9975</td>
</tr>
<tr>
<td>300</td>
<td>0.9949</td>
</tr>
<tr>
<td>500</td>
<td>0.9041</td>
</tr>
</tbody>
</table>
Figure 43. Relative efficiency of a 3'' x 3'' NaI(Tl) crystal as a function of energy and distance from the source.
Figure 44 is the relative efficiency as a function of energy of a 3 inch x 3 inch NaI(Tl) crystal for a line source 1 inch long and 261 cm from the crystal. An error of ±1 percent was assigned to the values read from the graph.
Figure 44. Relative efficiency of a 3" x 3" NaI(Tl) crystal 261 cm from a one inch line source as a function of energy.
In this appendix the equations used in the determination of the standard deviations of the gamma-ray relative intensities will be derived.

To determine the standard deviations of the $B_i$, several theorems about expected values will be needed. The proofs below follow those of Stevenson (74). We define the expected value of any function of the variable $z$ as

$$E[f(z)] = \int p(z) f(z) dz$$

where $p(z)$ is the distribution function of $z$. The distribution function $p(z)$ of the variable $z$ is defined such that $p(z)dz$ is the probability that a measurement value of $z$ will be between $u$ and $u+du$. $p(z)$ is normalized such that

$$\int_{-\infty}^{\infty} p(z)dz = 1.$$ 

Also define the covariance of two variables $z$ and $h$ by

$$\text{cov}(z,h) = E[(z-E(z))(h-E(h))].$$

The variance of $z$ is defined by $\text{var}(z) = E[(z-E(z))^2] = \sigma_z^2$, where $\sigma_z$ is called the standard deviation of $z$. Then for a constant $a$, the relationships follow:

$$E(a) = \int_{-\infty}^{\infty} ap_z(x)dx = a ,$$

$$E(az) = \int_{-\infty}^{\infty} ap_z(x)dx = aE(z) ,$$

Equation 28

Equation 29
\[ E(a+z) = \int_{-\infty}^{\infty} (a+x)p_x(x)dx = a + E(z) \text{ ,} \quad \text{Equation 30} \]

\[ \text{Var}(a) = E \left( (a - E(a))^2 \right) = E \left( (a - a)^2 \right) = 0 \text{ ,} \quad \text{Equation 31} \]

\[ \text{Var}(az) = E \left( (az - E(az))^2 \right) = E \left( (az - aE(z))^2 \right) = E \left( a^2 (z - E(z))^2 \right) = a^2 \text{ var}(z) \text{ ,} \quad \text{Equation 32} \]

\[ \text{Var}(a+z) = E \left( (a+z - E(a+z))^2 \right) = E \left( (a+z - a - E(z))^2 \right) = \text{Var}(z) \text{ .} \quad \text{Equation 33} \]

Also,
\[ E(z+h) = \int_{-\infty}^{\infty} x p_{z+h}(x)dx \text{ ,} \]

but \[ p_{z+h}(x) = \int_{-\infty}^{\infty} p_h(v)p_z(x-v)dv \text{ ,} \]

then \[ E(z+h) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} x p_h(v)p_z(x-v)dvdx \text{ .} \]

Reversing the order of integration and letting \( x = v-v+h \) we get
\[ E(z+h) = \int_{-\infty}^{\infty} p_h(v) \left[ \int_{-\infty}^{\infty} (x-v)p_z(x-v)dx + v \int_{-\infty}^{\infty} p_z(x-v)dx \right] dv \text{ .} \]
\[ dx = d(x-v) \text{ for the inner integrals since } v \text{ is held fixed.} \]

\[ E(z+h) = \int_{-\infty}^{\infty} p_h(v) \left[ \int_{-\infty}^{\infty} (x-v)p_z(x-v)d(x-v) + v \int_{-\infty}^{\infty} p_z(x-v)d(x-v) \right] dv \]
\[ = \int_{-\infty}^{\infty} p_h(v) \left[ E(z) + v \right] dv = E(z) \int_{-\infty}^{\infty} p_h(v)dv + \int_{-\infty}^{\infty} vp_h(v)dv \]
\[ E(z+h) = E(z) + E(h) \text{ .} \quad \text{Equation 34} \]

\[ \text{cov}(z,h) = E \left[ (z - E(z)) (h - E(h)) \right] = E \left[ z h - z E(h) - h E(z) + E(z) E(h) \right] \]
\[ = E(z h) - E(h) E(z) - E(z) E(h) + E(z) E(h) \]
\[ = E(z h) - E(z) E(h) \text{ .} \quad \text{Equation 35} \]
\[ \text{var}(z+h) = E \left[ (z+\text{E}(z)+h-\text{E}(h))^2 \right] \]

\[ = E \left[ (z-\text{E}(z))^2 + 2(z-\text{E}(z))(h-\text{E}(h)) + (h-\text{E}(h))^2 \right] \]

\[ = E \left[ (z-\text{E}(z))^2 \right] + E \left[ (h-\text{E}(h))^2 \right] + 2E \left[ (z-\text{E}(z))(h-\text{E}(h)) \right] \]

\[ \text{var}(z+h) = \text{var}(z) + \text{var}(h) + 2\text{cov}(z,h). \quad \text{Equation 36} \]

We will also need to show that the expected values of \( B_j \) determined from the normal equations are the true values. To show this we will follow the method of Kenny and Keeping (75, p. 309 ff.).

Let \( A^TWA = D \), and \( G = A^TWR \).

The normal equations then become \( DB = G \) and \( B = D^{-1}G \) or \( B_j = \sum_{k=1}^{C} (D^{-1})_{jk}G_k \).

Equation 38

The expected value of \( B_j \) is then determined from Equation 29

\[ E(B_j) = \sum_{k=1}^{C} (D^{-1})_{jk}E(G_k) \quad \text{Equation 39} \]

where \( E(G_k) = \sum_{i=1}^{C} a_{ki}^{T} E(R_i) \). If \( E(R_i) = \sum_{l=1}^{Q} a_{i1}^{T} n_l \) where \( n_l \) are the true values of the \( B_1 \), then

\[ E(B_j) = \sum_{k=1}^{C} \sum_{i=1}^{Q} \sum_{l=1}^{Q} (D^{-1})_{jk} a_{ki}^{T} a_{i1}^{T} n_l = \sum_{l=1}^{Q} \left[ n_l \sum_{k=1}^{C} \sum_{i=1}^{C} (D^{-1})_{jk} a_{ki}^{T} a_{i1}^{T} \right] \]
but from Equation 37 \( \sum_{i=1}^{C} a_{ki} T \omega_{ki} a_{i1} = D_{k1} \) so \( E(B_j) = \sum_{i=1}^{Q} \sum_{k=1}^{n} (D^{-1})_{jk} D_{k1} \). Equation 40

Using \( \sum_{k=1}^{C} (D^{-1})_{jk} D_{k1} = \delta_{j1} \) in Equation 40 we get

\[
E(B_j) = \sum_{i=1}^{Q} \eta_{i} \delta_{j1} = \eta_{j} .
\] Equation 41

We can now proceed to calculate the variances of the \( B_j \). To do this we need to calculate the covariance matrix of the vector \( B \). We will follow the approach due to Scheffe (76, pp. 8-12). Define the expected value of a matrix to be the matrix formed from the expected values of its components. For a vector \( V \),

\[
E(V) = \begin{bmatrix} E(V_1) \\ E(V_2) \\ \vdots \\ E(V_n) \end{bmatrix}
\]

and \( \text{cov}(V) = E \left[ (V - E(V))(V - E(V))^T \right] \).

For a constant matrix \( G \) and if \( W = GV \) then by applying Equation 12 we get

\[
\text{cov}(W) = E \left[ (W-E(W))(W-E(W))^T \right] + E \left[ G(V-E(V))(V-E(V))^T G^T \right] \\
= GE \left[ (V-E(V))(V-E(V))^T \right] G^T = G \text{cov}(V) G^T .
\] Equation 42
For simplicity let $A_{ij} = \frac{a_{ij}}{\sqrt{\text{var}(R_j)^{1/2}}}$ and $\psi_j = \frac{R_j}{\sqrt{\text{var}(R_j)^{1/2}}}$

Equation 43

where $\text{var}(R) = \sigma^2(R)$ from the definition of standard deviation.

Then Equation 10 becomes $B = (A^T A)^{-1} A^T \Lambda$. Equation 44

Assume the covariance matrix of $\psi$ is $\text{cov}(\psi) = \sigma^2 I$

where $I$ is the identity matrix. Then Equation 42 becomes, with $W = B$, $V = \psi$ and $G = (A^T A)^{-1} A^T$,

$$\text{cov}(B) = (A^T A)^{-1} A^T \cdot \text{cov}(\psi) \cdot \left[(A^T A)^{-1} A^T\right]^T$$

$$= \sigma^2 (A^T A)^{-1} A^T \cdot I \cdot A \cdot \left[(A^T A)^{-1}\right]^T.$$ 

Since $A^T A$ is symmetrical, $(A^T A)^{-1}$ is symmetrical. Therefore,

$$\text{cov}(B) = \sigma^2 (A^T A)^{-1} A^T \Lambda \cdot (A^T A)^{-1} = \sigma^2 (A^T A)^{-1}.$$ 

Using Equation 43 one gets

$$\text{cov}(B) = \sigma^2 (A^T W A)^{-1}.$$ Equation 45

We must now find an unbiased estimate of $\sigma^2$. Following the method of Kenney and Keeping (75, pp. 311-312) we define the residual $r_j$ by

$$r_j = \psi_j - T_j$$
where $y_j$ is, as before, the weighted observed value, and $T_j$ is the weighted value computed from the least-squares solution. Define the error $\delta_j$ by

$$\delta_j = y_j - \beta_j$$

where $\beta_j$ is the true value of $y_j$. Then by applying Equation 34 we get

$$E(y_j^2) = E[(\beta_j + \delta_j)^2] = E[(\beta_j^2 + 2\beta_j\delta_j + \delta_j^2)]$$

$$= E(\beta_j^2) + E(2\beta_j\delta_j) + E(\delta_j^2),$$

but from Equation 28 $E(\beta_j^2) = \beta_j^2$, and $E(\delta_j) = E(y_j - \beta_j) = E(y_j) - \beta_j$ from Equation 30, but from Equation 41 $E(\delta_j) = 0$. Also $E(\delta_j^2) = \sigma^2$ from the definition of variance. Therefore,

$$E(y_j^2) = \beta_j^2 + \sigma^2.$$  

Equation 46

If the true values of the $\beta_i$ are $n_i$, then

$$\beta_j = \sum_{i=1}^{Q} a_{ji} n_i / \sqrt{\text{var}(R_j)}^{1/2}.$$  

Therefore $E(y_j^2) = \sum_{i=1}^{Q} \sum_{k=1}^{Q} n_i n_k a_{ji} a_{jk} / \sqrt{\text{var}(R_j)}^{1/2} \sqrt{\text{var}(R_j)}^{1/2} + \sigma^2$.

Sum over all $j$ and apply Equation 34

$$\sum_{j=1}^{C} E(y_j^2) = E\left(\sum_{j=1}^{C} (y_j^2)\right) = C\sigma^2 + \sum_{i=1}^{Q} \sum_{k=1}^{Q} n_i n_k \sum_{j=1}^{C} a_{ji} a_{jk} / \sqrt{\text{var}(R_j)}^{1/2} \sqrt{\text{var}(R_j)}^{1/2}.$$
But from Equation 37,

\[ \sum_{j=1}^{C} a_{ji} a_{jk} / \text{var}(R_j) = D_{ik}, \]

so \( E( \sum_{j=1}^{C} (v_j)^2 ) = C \sigma^2 + \sum_{i=1}^{Q} \sum_{k=1}^{Q} n_i n_k D_{ik} \). \hspace{1cm} \text{Equation 47}

The normal equations were \( DB = AWAB = AWWR = G \),

so \( G_i = \sum_{k=1}^{Q} D_{ik} B_k \). Multiply both sides by \( B_i \) and sum over \( i \)

\[ \sum_{i=1}^{Q} B_i G_i = \sum_{i=1}^{Q} \sum_{k=1}^{Q} D_{ik} B_i B_k. \]

Taking expectation values and applying Equations 29, 35 and 28 respectively, we get

\[ E( \sum_{i=1}^{Q} B_i G_i ) = \sum_{i=1}^{Q} \sum_{i=1}^{Q} D_{ik} E( B_i B_k ) = \sum_{i=1}^{Q} \sum_{i=1}^{Q} D_{ik} \left[ n_i n_k + \text{cov}(B_i B_k) \right]. \]

Substituting from Equations 45 and 37 we have

\[ E( \sum_{i=1}^{Q} B_i G_i ) = \sum_{i=1}^{Q} \sum_{i=1}^{Q} D_{ik} \left[ n_i n_k + \sigma^2(D^{-1})_{ik} \right]. \]

or \( E( \sum_{i=1}^{Q} B_i G_i ) = \sum_{i=1}^{Q} \sum_{k=1}^{Q} D_{ik} n_i n_k + Q \sigma^2 \), since \( \sum_{k=1}^{Q} D_{ik}(D^{-1})_{ik} = \sum_{k=1}^{Q} \delta_{ik} = 1. \)
Thus,

\[ \sum_{i=1}^{Q} \sum_{k=1}^{Q} \eta_i \eta_k D_{ik} = E( \sum_{i=1}^{Q} B_i G_i ) - Q \sigma^2 . \]  

Equation 48

Substituting Equation 48 into Equation 47 we get

\[ E( \sum_{j=1}^{C} (\psi_j)^2 ) - E( \sum_{j=1}^{C} B_j G_j ) = (C-Q) \sigma^2 . \]  

Equation 49

Also, since \( r = \psi - T = \psi - AB/[\text{var}(R)]^{1/2} \),

\[ A^T r/[\text{var}(R)]^{1/2} = A^T \psi/[\text{var}(R)]^{1/2} - A^T AB/\text{var}(R) = G - A^T AB/\text{var}(R) = 0. \]

Therefore

\[ \sum_{j=1}^{C} r_j^2 = \psi^T r = \psi^T (\psi - AB/[\text{var}(r)]^{1/2}) = \psi^T \psi - AB/\text{var}(r) = 0. \]

Substitute in Equation 49

\[ \sigma^2 = E( \frac{1}{C} \sum_{j=1}^{C} r_j^2/(C-Q) ) . \]  

Equation 50
Let \( S^2 = \frac{C}{\sum_{j=1}^{3} \frac{r_j^2}{(C-Q)}} = \frac{C}{\sum_{j=1}^{3} \omega_j^2 (R_j - \frac{Q}{a_{jk}B_k})^2/(C-Q)} \),

Equation 51

then \( E(S^2) = \sigma^2 \), and \( S^2 \) is an unbiased estimate of \( \sigma^2 \).

Thus \( N_i = B_i \pm \sqrt{S^2(A^TWA)^{-1}} \).

Equation 52

Following the method of Beers (27, pp. 46-48) we will now show that for large \( N \), \( \text{var}(R_i) = R_i \). Let the probability that \( N \) particles are observed in time \( t \) be \( P_N \). Then, if \( t \) is divided into \( d \) equal intervals so small that the probability of emission of two particles in the same interval is negligible, the probability of the emission of one particle in a given interval is \( \overline{N}/d \) where \( \overline{N} \) is the average number of particles. The probability of emission of \( N \) particles in the first \( N \) intervals and none in the remaining \( d-N \) intervals is given by \( (\overline{N}/d)^N (1-\overline{N}/d)^{d-N} \). The number of ways of distributing \( N \) particles in \( d \) intervals is given by \( d(d-1)...(d-N+1) \). The number of ways of interchanging the particles is \( N! \). The probability of obtaining \( N \) counts is then given by \( P_N = d(d-1)...(d-N+1)/N! (\overline{N}/d)^N (1-\overline{N}/d)^{d-N} \), the binomial distribution law. If \( d \to \infty \), then \( d(d-1)...(d-N+1) \to d^N \) and \( (1-\overline{N}/d)^{d-N} \to e^{-N} \). Thus, \( P_N = \frac{\overline{N}^N}{N!}, \) the Poisson distribution.

Now use Stirlings approximation in the form \( N! = \sqrt{2\pi} N^{N+1/2} e^{-N} \) (this approximation has an error of less than 1 percent when \( N>10 \)) then \( N! = \sqrt{2\pi\overline{N}} \overline{N} \overline{N} e^{-\overline{N}} \), and \( P_N = 1/\sqrt{2\pi\overline{N}} \). Let \( Y = \ln P_N \) then

\[
Y = -N+Nln\overline{N}-ln\sqrt{2\pi} -(N+1/2)lnN+N .
\]
\[
\frac{dY}{dN} = \ln(N + \frac{1}{2}) - \ln(N) - 1.
\]

When \( P_N \) is a maximum \( \frac{dY}{dN} = 0 \) so \( \ln(N - 1) - 1 - 2N - \ln(N + 1) = 0 \) and \( \ln(N - 1) - e^{1/2N} - \ln(N) = 0 \) yielding \( N - Ne^{1/2N} = 0 \). Since \( N \) is postulated to be large, \( e^{1/2N} \) is essentially unity, and the maximum occurs close to \( N = \bar{N} \). Expand \( Y \) about \( \bar{N} \) in a Taylor series and retain only the first two nonvanishing terms.

\[
Y(N) = Y(\bar{N}) + \frac{dY}{dN}(N - \bar{N}) + \frac{1}{2} \frac{d^2Y}{dN^2}(N - \bar{N})^2 + \ldots
\]

Since the maximum occurs close to \( \bar{N} \), the first derivative of \( Y \) with respect to \( N \) is zero.

\[
Y(N) = -\ln(\sqrt{2\pi\bar{N}}) + \frac{1}{2}(N - \bar{N})^2 \frac{d^2Y}{dN^2}
\]

\[
\frac{d^2Y}{dN^2} = -\frac{1}{N - 1 - 2(-1/N^2)}
\]

so \( Y(N) = -\ln(\sqrt{2\pi\bar{N}}) + \frac{1}{2}(N - \bar{N})^2 \left[ \frac{1}{N - 1 - 2\bar{N}^2} \right] \).

Since \( \bar{N} \) is large, \( 1/\bar{N} \gg 1/2\bar{N}^2 \) and we drop the \( 1/2\bar{N}^2 \) term. Taking antilogarithms we get

\[
P_N = 1/\sqrt{2\pi\bar{N}} \ e^{-\frac{(N - \bar{N})^2}{2\bar{N}}}
\]

Letting \( N - \bar{N} = x \), we then have

\[
P_N = 1/\sqrt{2\pi\bar{N}} \ e^{-\frac{x^2}{2\bar{N}}}.
\]
but from Equation 5 \( P(x) = \frac{1}{\sqrt{2\pi \sigma^2}} e^{-\frac{x^2}{2\sigma^2}} \), so if on the average \( \bar{N} \) counts are observed in a time \( t \), the standard deviation in the number of counts is \( \sigma = \sqrt{\bar{N}} \). If it is assumed \( N \) is near \( \bar{N} \), \( \sigma = \sqrt{N} \). Thus the weights in Equations 10 and 52 are

\[
\omega_i = \frac{1}{R_i}. 
\]

Equation 53

In practice, because of background radiation, the weights given in Equation 53 must be modified. If \( N_i \) are the true number of counts in the experimental composite spectrum, then \( N_i = R_i - M B R_i \), where \( M \) is the ratio of the time for which the composite spectrum was recorded to the time for which the background was recorded, and \( B R_i \) is the number of counts recorded in the background spectrum. Then, \( \Delta N_i = \Delta R_i - M \Delta B R_i \), \( (\Delta N_i)^2 = (\Delta R_i)^2 + M^2 (\Delta B R_i)^2 - 2M \Delta R_i \Delta B R_i \). Take the average of both sides of the equation

\[
\frac{(\Delta N_i)^2}{\sigma(N_i)} = \frac{(\Delta R_i)^2}{\sigma(R_i)} + M^2 (\Delta B R_i)^2, 
\]

since the \( \Delta R_i \) and \( \Delta B R_i \) are independent. Therefore, assuming \( R_i \) and \( B R_i \) are large,

\[
(\sigma(N_i))^2 = (\sigma(R_i))^2 + M^2 (\sigma(B R_i))^2 = R_i + M^2 B R_i 
\]

and \( \omega_i = \frac{1}{R_i + M^2 B R_i} \).

Following the method of Fry (77, pp. 285-289) we will now show that \( S^2 \) has a \( x^2 \) distribution and can be used as a figure of merit. We had

\[
P(x_1, x_2, x_3, \ldots)dx_1 dx_2 dx_3 \ldots = K e^{-\sum_{i=1}^{C} \frac{x_i^2}{2\sigma_i^2}} dx_1 dx_2 \ldots 
\]
where \( x_i = R_i - \sum_{j=1}^{Q} a_{ij} B_j \).

But \( P(x_1, x_2 \ldots) dx_1 dx_2 \ldots = P(x_1(t_1, t_2, \ldots), x_2(t_1, t_2, \ldots) \ldots) \)

\[
\left| \frac{a(x_1, x_2, \ldots)}{a(t_1, t_2, \ldots)} \right| dt_1 dt_2 \ldots \]

\( = P(t_1, t_2, \ldots) dt_1 dt_2 \ldots \) (77, pp. 153-163).

Let \( q_i^2 = x_i^2 / \sigma_i^2 \), then

\[
P(q_1, q_2, \ldots) dq_1 dq_2 \ldots = Ke^{\sum_{i=1}^{C} 1/2 q_i^2} dq_1 dq_2 \ldots .
\]

For simplicity let \( C = 3 \). Also, let \( q_1^2 + q_2^2 + q_3^2 = r^2 \).

Suppose we have been given a set of the a's and have computed the sum of the squares of the q's and found it to be \( S^2 \). We want to see how reasonable the estimates of the a's are. To do this we compute the chance that another experiment, conducted so that its probabilities were really equal to the ones estimated from the experimental data, would lead to a result that is at least as improbable as the one under discussion. To do this we need to add together the probabilities of all admissible sets of values which are less likely to occur than the experimental ones. Since we have a decreasing exponential, the points which correspond to these sets all lie outside \( r = S \). Hence we need only add the probabilities corresponding to all admissible
points for which \( r > S \). In \( q \) space \( q_1^2 + q_2^2 + q_3^2 = r^2 \) is a sphere centered at the origin and with radius \( r \). All points on it have the same probability. The \( q \)'s are deviations measured in such units that equal vector deviations are equally likely, no matter what their directions. We integrate over all admissible values which lie outside a sphere of radius \( S \). Before doing this, we must know what regions contain these admissible values. This is determined from the auxiliary equations, which will be of the form

\[
\frac{C}{j=1} d_j q_j = 0.
\]

For example, if \( C = 3 \) the auxiliary equation is \( d_1 q_1 + d_2 q_2 + d_3 q_3 = 0 \). This is a plane passing through the origin of the coordinate system. All admissible points must lie on such a plane and the integral is no longer a volume integral outside a certain sphere but a surface integral outside a certain circle. For two such equations it will be a line which intersects two such planes. In general, a single condition on the variables reduces the space of \( C \) dimensions to one of \( C-1 \) and we must integrate over all those portions of this space which are further from the origin than a certain predetermined amount \( S \). \( Q \) conditions reduces the space to one of \( C-Q \) dimensions. In our case the \( Q \) auxiliary equations are

\[
\frac{C}{i=1} \frac{a_{i1}}{\sigma_i(R_i)} q_i = 0, \quad \frac{C}{i=1} \frac{a_{i2}}{\sigma_i(R_i)} q_i = 0, \quad \ldots, \quad \frac{C}{i=1} \frac{a_{iQ}}{\sigma_i(R_i)} q_i = 0.
\]

In one dimension

\[
P = 2K \int_S^\infty e^{-r^2/2} \, dr.
\]
In two dimensions
\[ P = 2\pi K \int_{S}^{\infty} e^{-r^2/2} \, r \, dr. \]

In three dimensions
\[ P = 4\pi K \int_{S}^{\infty} e^{-r^2/2} \, r^2 \, dr. \]

In \( c' \) dimensions, where \( c' = C-Q \),
\[ P_{c'}(>S^2) = K' \int_{S}^{\infty} e^{-r^2/2} \, r^{c'-1} \, dr, \]
but \( 1 = K' \int_{0}^{\infty} e^{-r^2/2} \, r^{c'-1} \, dr. \)

Let \( r^2/2 = u \), then
\[
1 = K' \int_{0}^{\infty} e^{-u} \left( 2u \right)^{c'-1} \frac{du}{\sqrt{2u}} = (2)^{\frac{c'-2}{2}} K' \int_{0}^{\infty} e^{-u}(u)^{\frac{c'-2}{2}} \, du = \]
\[
\frac{(2)^{\frac{c'-2}{2}}}{\Gamma\left(\frac{c'}{2}\right)} K'. \]

So \( K' = 1/\left[2^{\frac{c'}{2}} \Gamma^{\left(-\frac{c'-2}{2}\right)}\right]. \)

Then \( P_{c'}(>S^2) = \frac{1}{c'^{-1} - \frac{c'-2}{2} \Gamma\left(-\frac{c'-2}{2}\right)} \int_{S}^{\infty} e^{-r^2/2} \, r^{c'-1} \, dr. \) Equation 54

Therefore, the distribution \( P(r) \, dr \) is
\[ p(r)dr = \frac{1}{\left[ \frac{c'-2}{2} \right]!} \frac{e^{-r^2/2}}{r^{c'-1}} \text{ dr.} \]

We want the \( S^2 \) distribution. Let \( r^2 = S^2 \), then \( \frac{3r}{\delta S^2} = \frac{1}{2(S^2)^{1/2}} \)

and \( P(S^2)d(S^2) = \frac{1}{\left[ \frac{c'-2}{2} \right]!} \frac{e^{-S^2/2}}{(S^2)^{c'/2}} \text{ d}(S^2) \).

This is identical with the \( x^2 \) distribution

\[ P(x^2)d(x^2) = \frac{1}{\left( \frac{c'-2}{2} \right)!} \frac{e^{-x^2/2}}{(x^2)^{c'/2}} \exp(-x^2/2) \text{ d}(x^2). \]

so \( S^2 = \sum_{i=1}^{Q} \left[ \frac{R_i - \sum_{j=1}^{Q} a_{ij} B_j}{\sigma_i} \right]^2 \) has a \( x^2 \) distribution.

The integral in Equation 54 has been tabulated (77, p. 469). By knowing \( x^2 \) and the number of degrees of freedom, one can determine the "goodness" of the fit. The \( S^2 \) in Equation 54 is an estimate which corrects for variances which were not included in the weighting. If the weighting factors for the least-squares solution are chosen to correspond with the true values of the variances of the \( R_j \), then the value of \( \sigma^2 \) is 1 and from Equation 53 we get

\[ E(x^2) = C-Q \text{ and } E(x^2)/(C-Q) = 1. \]
In summary, the squares of the standard deviations of the gamma-ray relative intensities can be determined by multiplying the diagonal elements of \((A^{T}WA)^{-1}\) by \(S^2\), where \(S^2\) is given by Equation 51. The value of \(S^2\) can be used as a figure of merit and has an expected value of 1.
X. APPENDIX D: FLOW CHART AND REVISED COMPUTER PROGRAM

In this appendix the flow chart and listing of the IBM 360-50 computer program used to determine the gamma-ray relative intensities will be given. The program given here is a revised version of the one given in Reference 22. A maximum of 8 gamma rays recorded with a 400 channel analyzer can be analyzed at one time.

To determine the relative intensities the following equation must be solved

\[ B = (A^TWA)^{-1} A^TWR. \]

This is done by reading a parameter card which specifies the number of channels and the number of gamma rays and certain other options. The program then reads the matrices A, R and the background for R. Next the weights are determined. They can be read in, set equal to 1 or calculated. During the present measurements the weights were always calculated. Two sets of background for the response functions are then read in, averaged, and subtracted from the response functions. The program will then either normalize the response functions on the photopeak area or the total area. The total area was always used in these calculations. The curve to total ratio is then determined for each response function. Following the normalization \( A^TWA \) is formed, and an Ames Laboratory library subroutine MATINV is called to find \( (A^TWA)^{-1} \). This inverse is then multiplied times \( A^TWR \). Next, the goodness of fit parameter, \( S^2 \), defined by Equation 51 is calculated. The subroutine COMP then computes the relative intensities and standard
deviations in the relative intensities by computing the corrections and the standard deviation of the corrections. The response functions, calculated spectrum, experimental spectrum and deviation of the calculated spectrum from the experimental spectrum are then printed out. The flow chart and listings for the main program, and subroutines COMP, NLLS and SUBRT are given in Figures 45, 46, 47, 48 and 49. Subroutine NLLS is used when normalizing on the photopeak area. NLLS calls subroutine SUBRT.
Figure 45. Flow chart of the linear least-squares computer program
ISU DISK RES SPOOLED BPS FORTRAN

/AJOB   2 MIN
/BEGIN COMPILATION

INPUT -
JA NUMBER OF COLUMNS (13)
IN =?, CALCULATE W (OMEGA) (12)
+= READ W (OMEGA)
- = SET W = IDENTITY MATRIX
IWA = +, WEIGHTS IN NLLS = 1.0
= 0 WEIGHTS IN NLLS COMPUTED
INV = 0, CALCULATE A(-1) = A(1) AND PRINT (13)
++ = SKIP A(-1) = A(1) CALCULATION
IWRAP = 0, PRINT BA(I), I=1,JA
=-, + SKIIP PRINT

IN = +, NORMALIZE ON TOTAL AREA
= 0 NORMALIZE ON PHOTOPAK AREA
IWA = +, PRINT NORMALIZED A MATRIX
= 0, PRINT A(1) = NORM A MATRIX

TITLE IDENTIFICATION OF DATA (1,45)
CARD2 A(I,J) A MATRIX, READ BY COLUMNS (175,45)
CARD3 R(I) RHOD ARRAY (DIAGONAL ELEMENTS) (175,4)
CARD4 R(I) BACKGROUND
CARD5 W(I) OMEGA VECTOR (OPTIONAL MAY BE CALCULATED)
CARD6 2 VECTORS OF BACKGROUND A VECTOR AT A TIME FOR FIRST COLUMN
CARD7 BACKGROUND FOR SECOND COLUMN OF A FFT

S.0001 DIMENSION A(400,1),A(400,8),R(400),AWA(20,20),F(20,20),
IAWR(20),R(20),TITLE(12),AI(20),20,WORK1(20),FAI(20),
WORK2(20),WORK3(20),WORK4(20),CH0P(20),S0C0P(20),FRA00(20)
S.0002 DIMENSION RA(400),HP(400)
S.0003 DIMENSION X(250),R(20),STOP(25),R(12),G0(4),GP(20),
1ST0E(3,20),VARF2(I,20),NSIGN(I),GP0(20),5P1(20),AREA(20),
2E(20),ST0E(20),F(20),SDF(20)
S.0004 COMMON A1,A2,R,W,AWA,G,AI,AWR,P,IA,JA,TITLE,X,P,STOP,H,M,G50,F1,
1ST0E,VARF2,NSIGN,GP,G01,AREA,STOP,F,ST0E,F,20,0,ERROR
S.0005 EQUIVALENCE (BA(I),1), (HP(1),2)

S.0006 READ TITLE AND INDICATORS
S.0007 1100 READ (1,1100)A,J,IA,IN,IWA,IN,IPR,IN,IPA,TITLE
S.0008 1100 FORMAT (813,12A4)
C
S.0009 WRITE (3,1101)TITLE
S.0010 1191 FORMAT (1H1,9X,12A4)
C
S.0011 IF (IA)300,105,401
S.0012 300 STOP A9
S.0013 105 STOP
C
S.0014 READ IN DATA
S.0015 601 READ (1,602) IZERN
S.0016 602 FORMAT (15I)
S.0017 110 READ (1,1400)BAC,(AWR(J),J=1,JA)
S.0018 1400 FORMAT (5E15.8)
S.0019 DO III J=1,JA
S.0020 111 READ (1,11101)(A(I,J),I=1,IA)

Figure 46. Main program
Figure 46. (Continued)
Figure 46. (Continued)
S.0137 1124 FORMAT (19H A ARRAY BY COLUMNS,/(5E24.8))
S.0138 GO TO 130
S.0139 142 WRITE (3,1126)
S.0140 1126 FORMAT (16H SINGULAR MATRIX)
S.0141 STOP 89
S.0142 143 WRITE (3,1127)NIT
S.0143 1127 FORMAT (20H DID NOT CONVERGE IN 14,114 ITERATIONS)
S.0144 STOP 89
S.0145 144 WRITE (3,1128)
S.0146 1128 FORMAT (20H XTX MATRIX SINGULAR)
S.0147 STOP 89
S.0148 145 WRITE (3,1129)
S.0149 1129 FORMAT (19H GUESS ON PARAMETER,I?,4W40)
S.0150 STOP 89
S.0151 00 700 1 = 1,JA
S.0152 AWR(I)=0.
S.0153 DO 200 J=1,JA
S.0154 200 AWA(I,J)=0.
C
S.0155 DO 210 I=1,JA
S.0156 DO 210 J=1,JA
C
S.0157 A(I,J)=A(J,I)*W(J)
S.0158 AWR(I)=ATWR(I)+AWR(I)
C
S.0159 DO 210 K=1,JA
S.0160 AWA(I,K)=AWA(I,K)+ATW*A(J,K)
C
S.0161 DO 220 I=1,JA
S.0162 B(I)=AWR(I)
S.0163 DO 220 J=1,JA
S.0164 220 C(I,J)=AWA(I,J)
C
S.0165 CALL MATINV(C,JA,5,20,1,0,0,WORK 1,WORK 2,WORK 3,WORK A)
C
S.0166 IF (DET)240,235,240
S.0167 SINGULAR MATRIX MESSAGE
S.0168 WRITE (3,1235)I(R(I),1=1,JA)
S.0169 IF (INV)260,245,260
S.0170 IF (MULT)280,260,280
C
S.0171 1250 WRITE (3,1276)J,(A(I,J),1=1,JA)
S.0172 1260 FORMAT (6H A(I,J),2H)/(5E24.8))
S.0173 CONTINUE
C
S.0174 IF (MULT)280,260,280
S.0175 A(I,J)=0.
S.0176 DO 255 J=1,JA
S.0177 A(I,J)=A(I,J)+AWA(I,K)*C(K,J)
S.0178 WRITE (3,1250)J,(A(I,J),1=1,JA)
S.0179 1250 FORMAT (13H0IDENTITY ROW 14, 6F17.9/(1F7.0))
S.0180 CONTINUE
C
S.0181 255 WRITE (3,1250)J,(A(I,J),1=1,JA)
S.0182 1260 FORMAT (13H1 BETAVECTOR 7X 5E20.8/(12X 5F20.8))
S.0183 5\textsuperscript{1/2}A_JA=IA-JA

Figure 46. (Continued)
Figure 46. (Continued)
SUBROUTINE COMP(B,AWA,P,STOP,R1,STOP,J,E,COMP,SDCHP)

DIMENSION B(20),AWA(20),P(20),STOP(20),E(20),F(20),SOF(20),
1U(20),SDU(20),D(20),SO(20),EF(20),SDEY(20),SDF(20),
1STDRI(20),SA(20),SDF(20),SDCHP(20)

READ (1,2000)JA,N

READ (1,20fI11(E(I),F(I),SOF(I),T(I))=J,A)

READ (1,2002) ( (U(I,J) , SDU(I,J), D(I,J),S0(I,J),I=1,JA)

IF (8(11)402,500,401

WRITE (3,2013)I

FORMAT (3H B(,I?,11H» NfCATIVE)

STOP 89

WRITE (3,2100)I

FORMAT (3H 0(,I3,8H) = ZFRON

STOP B9

CONTINUE

DO 423 1=1,JA

Fl1)=F(I)+CHOP(I)

S0F(I)=SDCHP(I)+SOF(I)

WRITE (3,440)

FORMAT (32HK RATIO OF CUTOFF TO TOTAL C'PVE/Z)

WRITE (3,425)(E(I),F(I),S0(I),T(I))=1,JA)

AMAX=R(I)

IMAX=1

DO 403 I=2,JA

IF (8MAX-B(I))404,40 3,40 3

BMAX=B(I)

rMAX=I

CONTINUE

0MAX=0.

0UMAX=0.

DO 407 1=1,N

DUMAX = DUMAX + U(I MAX,I)*D(IMAX,I)

nMAX = DMAX+n(IMAX,I)*D(IMAX,I)

AMAX=EXP(-1.*DUMAX)

AMAX=AMAX*AMAX

BOMP = AMAX*EF(I MAX)*EF(I MAX)*IMAX

COM=AWA(IMAX)/EF(IMAX)*EF(IMAX)*EF(IMAX)

STOP(IMAX)/EF(IMAX)*EF(IMAX)*EF(IMAX)

STOP(IMAX)/EF(IMAX)*EF(IMAX)*EF(IMAX)

STOP(IMAX)/EF(IMAX)*EF(IMAX)*EF(IMAX)

DO 405 I=1,JA

DN=0.

DU=0.

DO 406 J=1,N

DDU=DDU+D(I,J)*DDU(J)

D=DU+DDU

A(I)=EXPO(-1.*DU)

SDF(1)=SDF(1)+SDF(1)*SDF(1)

A(I)=A(I)*STDP(1)

STOP(I)

Figure 47. Subroutine COMP
Figure 47. (Continued)
Figure 48. Subroutine NLLS
Figure 48. (Continued)
BEGIN COMPILATION

SUBROUTINE SUBRT(J,W,X,Y,GPI,DERIV,YC,F1,W1)

DIMENSION W(250),X(250),Y(250),GPI(1),DERIV(V(9))

YC=GPI(1)*EXP(-.5*((X(J)-GPI(2))/GPI(3))**2)

F1=Y(J)-YC

DERIV(1)=EXP(-.5*((X(J)-GPI(2))/GPI(3))**2)

DERIV(2)=(GPI(1)-((X(J)-GPI(2))/GPI(3))**2)*DERIV(1)

DERIV(3)=((X(J)-GPI(2))/GPI(3))**2*DERIV(1)

WI=WI+F1

RETURN
END

SIZE OF COMMON BLOCK PROGRAM 2242

END OF COMPILATION SUBRT

COMPILATION TIME WAS 0004.86 SECONDS

Figure 49. Subroutine SUBRT