A computer method for determining by least squares gamma ray relative intensities using a bent-crystal monochromator

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A COMPUTER METHOD FOR DETERMINING BY LEAST SQUARES GAMMA RAY RELATIVE INTENSITIES USING A BENT-CRYSTAL MONOCHROMATOR

by

Joseph Emerson Brown and E. N. Hatch
UNITED STATES ATOMIC ENERGY COMMISSION
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Joseph Emerson Brown and E. N. Hatch

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A COMPUTER METHOD FOR DETERMINING BY LEAST SQUARES GAMMA RAY RELATIVE INTENSITIES USING A BENT-CRYSTAL MONOCHROMATOR

Joseph E. Brown\textsuperscript{t} and E. N. Hatch

ABSTRACT

A method of measuring gamma-ray relative intensities was developed which used a germanium bent-crystal spectrometer as a monochromator. The response function of a NaI detector to the essentially monoenergetic gamma rays, obtained from the diffracted beam of the spectrometer, was recorded in a multichannel analyzer. The composite spectrum due to all the gamma rays was then recorded in the analyzer. A computer program was written to normalize the response functions to unit area and to determine, by minimizing the sum of the squares of the deviations, the linear combination of monoenergetic response functions that best represents the composite spectrum. The intensity of a gamma ray is then proportional to the adjusted area under the monoenergetic response function. The relative intensities of the gamma rays of Tm 169 obtained by electron capture from Yb 169 were measured.

\textsuperscript{*} This report is based on an M.S. thesis submitted by Joseph Emerson Brown to Iowa State University, Ames, Iowa, August, 1964.

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INTRODUCTION

Important information concerning the multipolarity, spin, and parity changes in a nuclear transition may be obtained by comparing experimentally measured internal conversion coefficients with those theoretically predicted (1). In determining these conversion coefficients by taking the ratio of the number of internal conversion electrons emitted to the number of emitted gamma rays for a given transition, the gamma-ray intensities usually have the largest error. Internal conversion line intensities may be measured using a magnetic spectrometer with an error of one or two percent while the gamma-ray intensities are frequently in error from five to ten percent or more. Since the theoretical calculations of internal conversion coefficients are estimated to be accurate to about five percent, it is desirable to measure internal conversion coefficients with an error of not more than five percent for a meaningful comparison. Thus a means must be found to reduce the uncertainty in the gamma-ray intensity measurements. It is usually sufficient to measure relative intensities and then to compute relative internal conversion coefficients. By a suitable normalization procedure absolute internal conversion coefficients may be obtained.

The three most common methods currently in use for determining accurate gamma-ray relative intensities are the
photoelectric conversion, crystal diffraction, and least squares scintillation methods. Hultberg has presented a careful description of the photoelectric conversion technique (2). In this method the 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 emitted electron is

\[ E = E_g - E_b \]

where \( E_g \) is the energy of the entering gamma ray and \( E_b \) is the binding energy of the electron removed from the atom. The number of electrons per second having a given momentum which strike the detector is then recorded. The intensity of the gamma rays can now be calculated if one knows the energy dependence of the cross section for photoelectric absorption and the angular distribution of converted electrons as well as the acceptance angle of the spectrometer. Corrections must also be made for the extension of the source and for absorption effects due to the particular experimental setup. This method takes advantage of the excellent resolution of the beta-ray spectrometer. Recent measurements have yielded conversion coefficients with errors from five to six percent (3, 4, 5).

The second method of determining relative intensities
uses the principle of crystal diffraction. This method has been perfected by Lind (6, 7), Hatch (8), Bergvall (9), and Edwards (10). A bent crystal spectrometer can be used to diffract gamma rays ranging in energy from 30 keV to 600 keV.

It can be shown (10) that, after making appropriate corrections, the intensity of each of the gamma rays emitted from the spectrometer source is proportional to the counting rate when the spectrometer is set at the peak of the diffracted line profile. The intensities must be corrected for self absorption in the source, absorption in the source container, the energy dependent reflectivity of the diffraction crystal, absorption in the air path and the cover of the NaI crystal, and finally for the half life of the isotope since not all of the measurements are made at the same time. One of the particularly troublesome corrections is that due to self absorption in the source. Since the density of the source material, which is frequently a powder, is not known well, the uncertainty in this correction is sometimes larger than that of the other corrections. This technique leads to relative intensities that are in error from two to twenty percent (11, 12).

The last standard method of measuring gamma intensities is the least squares scintillation method. Reynolds (13), Salmon (14), Trombka (15, 16), and Heath (17) have written computer programs to compute relative intensities using this
technique. This method requires the use of a multichannel analyzer, amplifier, preamplifier, photomultiplier tube, and a suitable detector such as a NaI crystal. The basic assumption is that the intensity of a gamma ray is proportional to its area under the pulse height spectrum as recorded on a multichannel analyzer. It is found that if an isotope emits several gamma rays each contributes counts to the composite spectrum as if it were the only gamma ray present. The composite spectrum is then merely the accumulation of the counts due to the various gamma rays. In order to apply this method one needs to obtain the response function of the detector to individual gamma rays of all energies of interest present in the source. In practice it is only possible to obtain a limited number of monoenergetic gamma emitters. It then becomes necessary to devise an interpolation scheme to obtain the response function for a gamma ray of any energy. A computer program is then written to take suitably normalized monoenergetic response functions and determine by a linear least squares adjustment how much each gamma ray contributes to the area under the composite spectrum and thus what its intensity is. Corrections are made for the efficiency of the detector and for absorption in the media between the source and detector. The method is limited by the lack of a sufficient number of monoenergetic emitters. Most of those available are not truly monoenergetic gamma
emitters, or they are very short lived and thus difficult to obtain and to work with. As a result it is difficult to obtain accurate experimental response functions for gamma rays of a given energy. Recent measurements have quoted errors from one to twenty percent (16, 17).

The new method described here utilizes some of the better features of the crystal diffraction and least squares scintillation methods with a subsequent improvement in flexibility and accuracy. Briefly, the technique consists of exposing a very thin, weak line source to the collimator and NaI crystal of a bent crystal spectrometer with the diffraction crystal removed. This allows the composite spectrum to be recorded in a scatter free geometry with few absorption corrections under essentially the same geometrical conditions as the monoenergetic response functions will be measured. The weak source is then removed and a one curie line source is used to diffract the gamma rays of the isotope according to their energies. Thus the spectrometer serves as a monochromator. The response of the detector to each of these monoenergetic gamma rays is then recorded on the multichannel analyzer. An IBM 7074 computer is then used to normalize the monoenergetic response functions and determine the area under the complex spectrum attributable to each gamma ray. Corrections are made for absorption in the air path and cover on the NaI crystal as well as for the efficiency of the crystal. The troublesome corrections due to self absorption in the
source and source holder as well as that for the energy dependent reflectivity of the crystal are avoided. Since the response functions are experimentally determined, there is no need to interpolate to obtain response functions for many cases of interest. If it is desirable to interpolate between two functions to obtain the response functions for a short-lived isotope with some weak lines, it will be possible to choose, from a library of response functions that is being built up, several that are very close in energy to the desired one. This technique presently produces relative intensities that are in error by three to twelve percent.
THE LEAST SQUARES SCINTILLATION METHOD

The least squares scintillation method of determining gamma-ray relative intensities depends on the various ways that gamma rays may interact with matter (15, 16, 17). We restrict the discussion to gamma rays with energies less than 1.02 MeV, the pair production threshold. The two primary ways that gamma rays interact with matter in this energy region are through photoelectric absorption and Compton scattering. Although there is no sharp cutoff, photoelectric absorption is most important at low energies while Compton scattering is most important above 0.5 MeV. Photoelectric absorption occurs when a gamma ray ejects an electron from a K, L, or M shell in an atom. The gamma-ray photon transfers all of its energy to the electron and disappears. In order to conserve momentum this process must take place near a third body to which the electron is bound. A certain amount of energy is required to remove the electron from the atom against the electrostatic attraction of the nucleus. The energy $E$ of the emitted electron is given by

$$E = E_g - E_b$$

where $E_b$ is the electron binding energy referred to above, and $E_g$ is the energy of the incident gamma ray. After the electron is ejected from the atom, an electron from an outer shell will fill the vacancy left by the ejected electron causing the emission of an X-ray or Auger electron.
At higher energies Compton scattering becomes important. This is the process whereby a photon interacts with an electron that is essentially free by transferring part of its energy to the electron and scattering in such a way as to conserve energy and momentum. The energy $E_s$ of the scattered gamma ray is given by

$$E_s = \frac{E_g}{1 + \frac{E_g (1 - \cos \Theta)}{mc^2}}$$

where $E_g$ is the energy of the incident gamma ray and $\Theta$ is the angle that the scattered gamma ray makes with the direction of the incident gamma ray. Maximum energy transfer to the electron occurs when $\Theta = \pi$. For this value of $\Theta$ the energy of the backscattered photon is given by

$$E_{bs} = \frac{E_g}{1 + \frac{2E_g}{mc^2}}$$

and the maximum energy transferred to the electron is given by

$$E_c = E_g - \frac{E_g}{1 + \frac{2E_g}{mc^2}}.$$ 

The minimum energy transfer occurs as $\Theta$ goes to zero. The probability is greatest for large and small angle scattering while it is least for scattering at $\Theta = 90$ degrees.

When the detector is a NaI crystal containing thallium ions as an activator, the electrons which have received energy through photoelectric absorption or Compton scattering excite electron hole formation in the valence band. Due to
the thallium ions, there are accessible electronic energy levels called luminescence centers in the normally forbidden region between the conduction and valence bands. Transitions from the conduction band through these luminescence centers is then possible with resulting emission of light photons. The decay time of the light pulse in the crystal is longer than the interaction time of the gamma ray. Consequently a gamma ray may be scattered several times, photoelectrically absorbed, and may have the following X-ray emitted and absorbed before the light pulse decays. Thus the intensity of the light produced in the crystal in this manner is proportional to the energy which the gamma ray loses in the crystal.

A photomultiplier tube is optically coupled to the NaI crystal. After the pulse of light is produced in the crystal it is reflected to strike the active surface of the phototube. Electrons are ejected from this surface in proportion to the intensity of the light pulse. These electrons are multiplied through several stages. The result is that a voltage pulse is created at the output of the phototube. This voltage pulse is proportional to the energy lost by the gamma ray in the crystal.

The preamplifier and linear amplifier amplify and transmit the voltage pulse to a multichannel analyzer. The multichannel analyzer may have 400 channels or memory locations. If the maximum voltage pulse that it can receive is 10 volts then each channel is $\frac{10}{400} = 0.025$ volts wide. The function of the multichannel analyzer is to record and store in a memory
the number of voltage pulses that are received according to their magnitudes. Thus the analyzer will store in channel 1 the number of pulses received that lie between 0.000 and 0.025 volts, in channel 2 all those between 0.025 and 0.050 volts, and in channel i the number of voltage pulses received between $V_i$ and $V_i + 0.025$ volts. $V_i$ is given by

$$V_i = (i - 1) (0.025), \quad i = 1, \ldots, 400.$$ 

The result of all these effects is that when monoenergetic gamma rays strike a NaI crystal coupled to the electronic system mentioned above, a distribution called a response function is formed in the multichannel analyzer. This response function consists of the number of counts in a channel versus channel number. A typical response function is shown in Figure 1. The response function consists of two parts for energies less than 1.02 MeV. The higher energy portion consists of the photopeak or full energy loss peak and the iodine escape peak if this is resolved from the photopeak. These peaks are nearly Gaussian in shape with some deviation at the low energy side due to the escape peak. The photopeak obviously results from those gamma rays which lose all of their energy in the crystal. The channel number of the center of this peak is proportional to the energy of the monoenergetic gamma rays that struck the crystal. The iodine escape peak results from those gamma rays that transferred all of their energy to electrons in the detector by undergoing photoelectric absorption. The iodine X-ray that followed the emission of
Figure 1. A typical monoenergetic response function of a NaI crystal to a 1 MeV gamma ray.
the electron, however, escaped from the crystal. From equation 1 the energy of the center of this peak is

\[ E = E_g - E_b \]

where here \( E_b \) is the binding energy of the K electron in iodine.

The second feature of the response function of monoenergetic gamma rays is a broad hump called the Compton continuum which is lower in energy than the photopeak. The voltage pulses that are counted in this region are due to those gamma rays which are Compton scattered, with the scattered photons escaping from the crystal so that only a part of their energy is lost in the crystal. The maximum energy transferred to an electron in the crystal through the Compton effect is as we have seen in equation 4

\[ E_c = E_g - \frac{E_g}{1 + \frac{2E_g}{mc^2}}. \]

Thus, counts due to Compton scattering occur in channels proportional to \( E_c \), the Compton edge, down to channel 1. For some experimental geometries there is a small peak in the Compton continuum due to gamma rays that scatter through 180 degrees in matter surrounding the crystal. These backscattered rays are then detected by the crystal. The energy of this peak is given by equation 3

\[ E_{bs} = \frac{E_g}{1 + \frac{2E_g}{mc^2}}. \]
Prior to the work of Reynolds (13) and others (14, 15, 16, 17), the accepted method of determining gamma-ray relative intensities using a scintillation method was the spectrum peeling method. In this method the response function of the highest energy gamma ray was fitted to the highest energy photopeak in the composite spectrum. The counts due to this gamma ray were then subtracted from all channels and the procedure was repeated for the second highest energy photopeak. This procedure was continued until all of the response functions were subtracted. The intensity of each gamma ray is proportional to the area under the photopeak in the fitted response function. Girgis et al. (18) and West and Johnston (19) have described this method. The two disadvantages of this method are that, first, it cannot be applied well if there are several overlapping peaks and, second, the errors tend to accumulate as the peeling continues so that the last intensities have larger errors.

The following paragraphs describe the approach to measuring relative intensities developed by Reynolds (13), Salmon (14), Trombka (15, 16), and Heath (17). The development below follows that of Trombka (14, 15) and relies on the fact that the complex spectrum is merely a linear combination of response functions due to the presence of gamma rays of various energies. The number of counts in the photopeak of each of these monoenergetic response functions is related in the
following way to the intensity of the gamma ray giving rise to the response function.

\[ N_{pn} = I_n \cdot t \cdot \frac{W}{4\pi} \cdot e^{-udn} \cdot \frac{E_n}{P_n} \]  

**equation 5**

where

- \( N_{pn} \) = the number of counts in the photopeak of gamma ray \( n \) with energy \( H_n \)
- \( I_n \) = the number of gamma rays of energy \( H_n \) emitted per second
- \( t \) = the time the crystal is exposed to the source
- \( \frac{W}{4\pi} \) = the solid angle subtended by the crystal divided by the total solid angle
- \( e^{-udn} \) = Fraction of the gamma rays of energy \( H_n \) that are not scattered out of the beam or absorbed before reaching the NaI crystal

\[ E_n = \text{Total number of counts detected in the monoenergetic spectrum divided by the total number of gamma rays of energy } H_n \text{ which are incident upon the scintillation crystal} \]

\[ P_n = \text{Number of counts detected in the photopeak and iodine escape peak divided by the total number of counts detected for a gamma ray of energy } H_n \]

\[ F_n = e^{-udn} \cdot E_n \cdot P_n \cdot \]

The relative intensity of two gamma rays of energy \( H_n \) and \( H_m \) is given by

\[ \frac{I_n}{I_m} = \frac{N_{pn} \cdot F_m}{N_{pm} \cdot F_n} \]  

**equation 6**

since the solid angle subtended by the crystal and the time observed are the same for all gamma rays and thus cancel out.

The problem of determining relative intensities is the problem of determining \( \frac{N_{pn}}{N_{pm}} \). The \( e^{-udn} \) may be determined by looking
up the absorption coefficients in the appropriate tables (20, 21). The $E_n$ may be obtained from a list by Vegors et al. (22). $P_n$ must be determined experimentally for the source to crystal distance of a given experiment under conditions of negligible scattering.

Trombka (15, 16) uses the photopeak area as a measure of the gamma-ray intensity since under his experimental conditions there are expected to be spurious counts due to scattering. Thus the photopeak should be the least distorted part of the spectrum because any scattered pulses would be degraded in energy and appear at a lower energy portion of the spectrum. This procedure is also useful if it is desired to set the discriminator on the multichannel analyzer so that all pulses below a certain cutoff voltage are ignored.

In order to determine $\frac{N_{pm}}{N_{pm}}$, the response function of a NaI crystal to a source with gamma rays of $p$ different energies is recorded in a 400 channel multichannel analyzer. Let this composite spectrum be represented by

$$R_i, i = 1, \ldots, 400$$

where $R_i$ is the total number of counts in channel $i$ due to all $p$ gamma rays. Let

$$A_{in}, i = 1, \ldots, 400, n = 1, \ldots, p$$

be the number of counts in channel $i$ in the normalized response function of a monoenergetic gamma ray of energy $H_n$. The response function is normalized so that the area under
the photopeak is unity. The area under the photopeak is defined in this method to be the area under a Gaussian curve fitted to the high energy side of the photopeak. If the iodine escape peak is resolved, its area is added to that of the photopeak. Let

\[ B_n, \ n = 1, \ldots, p \]

be the area under the corresponding photopeak in the complex spectrum attributable to a gamma ray of energy \( H_n \) as determined by a linear least squares adjustment. Thus

\[ B_n = Npn \]

according to our previous definition. Let

\[ X_i, \ i = 1, \ldots, 400 \]

be the independent, random error in channel \( i \) due to statistical fluctuations in \( R_i \). We assume here that the \( A_{in} \) are known such that their fluctuations are small compared with those of \( R_i \). We therefore have

\[ R_i = \sum_{n=1}^{p} A_{in} B_n + X_i \]

\[ X_i = R_i - \sum_{n=1}^{p} A_{in} B_n \]

equation 7

Assuming that the error \( X_i \) is random, the probability \( p_i \) that the error \( X_i \) is between \( X_i \) and \( X_i + dX_i \) is given by

\[ p_i = \frac{1}{\sqrt{2\pi s_i}} e^{-\frac{(X_i^2)}{2s_i^2}} \ dX_i \]

where \( s_i \) is the standard deviation in the number of counts in
channel i due to random fluctuations in $R_i$. Let $P$ be the probability that 400 errors will be observed such that $X_1$ lies between $X_1$ and $X_1 + dX_1$, while $X_2$ lies between $X_2$ and $X_2 + dX_2$, ..., $X_{400}$ lies between $X_{400}$ and $X_{400} + dX_{400}$. Thus we have

$$P = \prod_{i=1}^{400} P_i$$

$$= e^{-\sum_{i=1}^{400} \frac{1}{2s_i^2} (X_i - 2 \cdot \frac{dX_i}{2s_i})^2} \prod_{i=1}^{400} \frac{dX_i}{\sqrt{2\pi s_i}}$$

since the error in each channel is independent, the $dX_i$ are arbitrary and the $s_i$ are fixed. The principle of maximum likelihood (23) asserts that the best possible values of the $B_n$ are those which maximize $P$. This can be understood for the following reason. $P$ is a function of the $B_n$, and values of $B_n$ which make $P$ negligibly small would not be expected to give rise to the measured errors after making only one observation. It is expected that the values of the errors which were observed in one measurement were observed because their probability of occurrence was high. $P$ will be a maximum when

$$\sum_{i=1}^{400} \frac{X_i^2}{2s_i^2}$$

is a minimum since the $s_i$ are fixed and the $dX_i$ are arbitrary. We can omit the 2 in the denominator since a constant multiple has no effect on the minimizing process. We are lead to minimizing the quantity
\[
T = 400 \sum_{i=1}^{400} \frac{(R_i - \sum_{n=1}^{p} A_{in} B_n)^2}{s_i^2}
\]

\[
= \sum_{i=1}^{400} w_i (R_i - \sum_{n=1}^{p} A_{in} B_n)^2
\]

equation 8

where

\[
w_i = \frac{1}{s_i^2} = \frac{1}{R_i}
\]

since

\[
s_i^2 = R_i
\]

Thus the criterion for an acceptable set of \(B_n\) is that the weighted sum of the squares of the errors in each channel between the observed and computed composite spectrum must be a minimum. The weights have the effect of normalizing the observed deviations, \(X_i\), in each channel by the standard deviation of the counts, \(R_i\), in each channel so that only significant departures from the expected deviations, \(R_i\), contribute to \(T\). The minimum value of \(T\) will occur for those values of \(B_n\) for which

\[
\frac{\partial T}{\partial B_k} = 0, \quad k = 1, \ldots, p.
\]

equation 9

Setting the computed partial derivatives equal to zero we are led to a set of normal equations.

\[
\sum_{i=1}^{400} w_i (R_i - \sum_{n=1}^{p} A_{in} B_n)A_{ik} = 0, \quad k = 1, \ldots, p.
\]
Let $A_{ki}^T = A_{ik}$ where the $T$ superscript stands for transpose.

Thus we have

$$
\sum_{i=1}^{400} A_{ki} w_i R_i - \sum_{i=1}^{400} A_{ki}^T w_i \sum_{n=1}^P A_{in} B_n = 0, \quad k = 1, \ldots, p.
$$

We rewrite this expression in matrix notation where capital letters stand for matrices.

- $A$ is a $(400 \times p)$ matrix of the $A_{in}$ with each column being a monoenergetic response function.
- $A^T$ is the transpose of $A$.
- $W$ is a $(400 \times 400)$ diagonal matrix with the $w_i$ on the diagonal.
- $B$ is a $(p \times 1)$ vector of the $B_n$.
- $R$ is a $(400 \times 1)$ vector of the $R_i$.

In matrix notation we have

$$
A^T W R - A^T W A B = 0
$$

$$(A^T W A) B = A^T W R
$$

$$
B = (A^T W A)^{-1} A^T W R \quad \text{equation 10}
$$

for $A^T W A$ nonsingular. Thus to solve for $B$ all one needs to do is to take the inverse of the matrix product $A^T W A$ and multiply that times the matrix product $A^T W R$. Having computed the $B_n$ the relative intensities may be calculated by substituting the $B_n = N_{pn}$ in equation 6. These calculations may be programed for a computer.

Here we depart from the method of development of Trombka (16, 17) in order to determine an estimate of the variances and covariances of the $B_n$. The approach given here is due to Scheffé (24). We continue to assume that the error $X_i$ is
random and due to the fluctuations in $R_i$, and thus that the fluctuations in the $A_{in}$ are much smaller than the fluctuations in $R_i$. We define the following quantities.

$E(R_i)$ is the expected value of the random variable $R_i$

$$E(R) = \begin{pmatrix} E(R_1) \\ \vdots \\ E(R_{400}) \end{pmatrix}$$

$E(R)$ is the expected value of the matrix $R$ defined above.

$$\text{Cov}(R_i, R_j) = E((R_i - E(R_i))(R_j - E(R_j)))$$

$$(VR)_{ij} = (\text{Cov}(R_i, R_j))_{ij} \text{ is the } i,j \text{ element of the covariance matrix, } VR, \text{ for } R. \ VR \text{ is a (400X400) diagonal matrix.}$$

$VR = E((R - E(R))(R - E(R))^T)$.

Thus the covariance matrix, $VR$, has the variance of the $R_i$ on the diagonal and zero for the off diagonal elements since the fluctuations in each channel are assumed to be independent of those in any other channel.

We now show that if

$$B = CR$$

where $B$ and $R$ are as before and $C = (ATWA)^{-1}ATW$ is assumed to be known without error ($E(C) = C$) then

$$VB = CVRC^T$$

where $VB$ is the (20X20) covariance matrix for $B$ with the variances of the $B_n$ on the diagonal and the covariances on the off diagonal positions. By definition
\[ V_B = E(((B - E(B)) (B - E(B)))^T) \]

but
\[ E(B) = C E(R) \]

since \( C \) is known without error. Thus
\[
V_B = E((C R - C E(R))(C R - C E(R))^T)
\]
\[ = E(C (R - E(R))(C (R - E(R)))^T) \]
\[ = C E((R - E(R)(R - E(R))^T) c^T \]

since \( E(C) = C \)

and \( (C (R - E(R)))^T = (R - E(R))^T c^T \)

therefore
\[ V_B = C V_R c^T. \]

Now \((V_R)_{ij} = R_i \delta_{ij} = \frac{1}{w_i} \delta_{ij} = (W^{-1})_{ij} \) since \( W W^{-1} = I \).

Since \( W \) is diagonal, \( W = W^T \).

Thus
\[
V_B = (A^T W A)^{-1} A^T W W^{-1} ((A^T W A)^{-1} A^T W)^T
\]
\[ = (A^T W A)^{-1} A^T I W^T (A^T)^T((A^T W A)^{-1})^T \]
\[ = (A^T W A)^{-1} A^T W A (A^T W A)^{-1} \]
\[ = (A^T W A)^{-1} \]

since \( A^T W A \) and thus \((A^T W A)^{-1}\) is symmetric. Thus the
variances of the \( B_n \) appear on the diagonal of the matrix
\((A^T W A)^{-1}\) which was computed and used in equation 10 to compute the \( B_n \).

The covariance of \( B_i \) and \( B_j \) appears in the i, j position on this matrix where i is not equal to j.

Actually Scheffé (24) shows that it is possible to improve the estimation of the error in the \( B_n \) in the following way. If we had assumed that the weights for determining the minimum of \( T \) as given by equation 8 were
where here $m$ is an arbitrary constant to be determined, the $m$ would have canceled out when the partial derivatives $\frac{\partial T}{\partial B_k}$ were set equal to zero in equation 9. Hence multiplying the weights by a constant does not alter the fitting procedure to determine the $B_n$. In determining the covariance matrix $V_B$, however, we would now have

$$V_B = m (AT W A)^{-1}.$$ Scheffé (24) shows that an unbiased estimate of $m$ is given by

$$m = \frac{\sum_{i=1}^{400} w_i (R_i - \sum_{n=1}^{P} A_n B_n)^2}{400 - P}$$

The best estimate of the covariance matrix $V_B$ is then

$$V_B = \sum_{i=1}^{400} w_i \frac{(R_i - \sum_{n=1}^{P} A_n B_n)^2}{400 - P} (AT W A)^{-1}$$

The quantity $m$ can be used as a figure of merit to determine how well the computed composite spectrum equals the observed composite spectrum. Moore (25) argues that under the assumptions that the $w_i$ are the known reciprocals of the variances of the $R_i$, that the $R_i$ are normally or Gaussian distributed, and that the $B_n$ are estimated by using the least square estimate above then

$$\sum_{i=1}^{400} w_i \frac{(R_i - \sum_{n=1}^{P} A_n B_n)^2}{400}$$

has a chi-square distribution. This implies that at least
fifty percent of the time one would expect T to be less than or equal to the number of degrees of freedom 400 - p. Thus the expected value of m is one. If m is considerably more than one it is likely that errors are occurring other than those due to statistical fluctuations in R_1.

The method described above avoids the difficulties presented by the spectrum stripping method. The accuracy associated with the measurement of a low energy gamma ray is improved. In the spectrum stripping method one subtracts the response function of the highest energy gamma ray from the composite spectrum and then repeats the procedure for the next lowest energy gamma ray. Each time the subtraction is performed some error is made. This error tends to accumulate so that the lowest energy lines usually have the largest error. In the least squares scintillation procedure all gamma rays have their response functions fitted simultaneously and the accuracy of the intensity of a gamma ray of a given energy is limited just by the statistical fluctuations in the counts in its peak in the composite spectrum. Since the calculations are done on a computer, a composite spectrum can be analyzed much more quickly by this method than by the spectrum stripping method. Overlapping peaks can be readily handled by this approach as long as their gamma rays are not too weak. Finally this method permits the calculation of an estimate of the error in the E_n.

Trombka (15, 16) and Heath (17) have devised interpolation
procedures to generate monoenergetic response functions $A_{\text{in}}$ for gamma rays of any energy. Because of the difficulty in obtaining monoenergetic gamma emitters that are long lived, it is necessary to interpolate over fairly large energy regions with a subsequent increase in the error in a response function.
THE LEAST SQUARES SCINTILLATION TECHNIQUE
USING A BENT-CRYSTAL MONOCHROMATOR

The method proposed here is basically an extension of the scintillation method due to Reynolds (13), Salmon (14), Trombka (15, 16) and Heath (17), described in the previous chapter. In this method a bent-crystal spectrometer is used as a gamma-ray monochromator to diffract the various gamma rays according to their energies. It is then possible to expose the NaI crystal to gamma rays of one energy only and to measure their response functions. This procedure eliminates the need for interpolating to obtain response functions, as was done previously, as well as eliminating the need for certain corrections necessary in the diffraction method. In this chapter the theory of the bent crystal spectrometer will be briefly reviewed, and the changes required in the development of the least squares method as given in the previous chapter will be presented.

The theory of the bent-crystal spectrometer has been described by DuMond (26, 27, 28) and others (9, 29). Essentially the bent-crystal spectrometer consists of three components: a source, a curved germanium, quartz or other diffraction crystal, and a collimator-detector system (Figure 2). The crystal is bent so that its radius of curvature is the diameter $R$ of the focal circle. The bending has the following effect. If one were to extend the diffracting planes
Figure 2. A schematic drawing of a bent-crystal spectrometer.
of the crystal, they would intersect the focal circle at the point on the focal circle opposite the crystal on the diameter R. Dumond (26) shows that if a source is placed on the focal circle at the point S such that the Bragg condition,
\[ n\lambda = 2d \sin \theta, \ n = 1, 2, \ldots \], is satisfied, the diffracted gamma rays will appear to come from a virtual image on the focal circle which is a reflection of the source position through the diameter R. In the Bragg equation, \( n \) is the order of the reflection, \( \lambda \) is the wavelength of the gamma ray, \( d \) is the spacing of the diffracting planes, and \( \theta \) is the angle between the gamma ray and the diffracting planes in the crystal.

The collimator consists of many closely spaced lead sheets tapered such that a projection of the plates would intersect the focal circle at the virtual image. The collimator is needed to shield the NaI detector, which is immediately behind it, from the gamma rays of the direct beam. The collimator serves another useful purpose. Figure 2 shows an exaggerated separation between the plates for the purpose of illustration. In reality the plates are very closely spaced. A gamma ray which is Compton-scattered from one of these plates has a very small probability of passing through the collimator to the crystal without intersecting another plate and thus being further scattered and absorbed. As a result, when the source, crystal, and collimator-detector geometry are such that
and the collimator points at the virtual image, gamma rays of essentially one energy strike the crystal.

Since it is now possible to measure the monoenergetic response functions,

\[ A_{i,n} = 1, \ldots, 400, \quad n = 1, \ldots, p, \]

the necessity for devising an interpolation technique to generate them is eliminated. Furthermore, it is found in chapter 4 that there are a negligible number of scattered gamma rays reaching the crystal. Thus it is more accurate to normalize these monoenergetic response functions so that their total area is unity. This procedure eliminates the need for fitting a Gaussian to the photopeak of the response function. The process of fitting a Gaussian to the photopeak is a somewhat subjective operation. A decision must be made concerning exactly how many points will be chosen for the fitting operation and where on the response function these points should be taken. The problem is complicated by the fact that the peaks become broader as the energy of the gamma ray increases. If a fitted Gaussian which does not match the experimental curve exactly is used as a measure of the area under the photopeak, then there will be a corresponding error in the determination of the relative intensities. For this reason it was decided to measure the monoenergetic response function, add up the number
of counts in each channel, and then divide the number of counts in each channel by the total number of counts. This procedure will normalize the area under the monoenergetic spectrum to unity.

We now have the following relationship between the total number of counts detected and the intensity of a monoenergetic gamma ray. This compares with equation 5.

\[ N_n = I_n \frac{w}{4\pi} e^{-\mu_d n} E_n \]  

Equation 13

where

- \( N_n \) = the total number of gamma rays of energy \( H_n \) detected in time \( t \)
- \( I_n \) = the number of gamma rays of energy \( H_n \) emitted per second
- \( t \) = the time that the crystal is exposed to the source
- \( \frac{w}{4\pi} \) = the fraction of the total solid angle subtended by the crystal
- \( e^{-\mu_d n} \) = the fraction of gamma rays in the beam not scattered or absorbed
- \( E_n \) = the number of counts produced in the analyzer divided by the total number of gamma rays that struck the face of the crystal.

The relative intensity of two gamma rays is given by

\[ \frac{I_n}{I_m} = \frac{N_n e^{-\mu_d m} E_m}{N_m e^{-\mu_d n} E_n} \]  

Equation 14

The photopeak-to-total ratio that appeared in equation 5 is not needed here if the total number of counts in a monoenergetic spectrum is used as the measure of the intensity. If,
however, the lower level discriminator is set to suppress the counts of lower energy, a factor similar to \( P \), which would give the curve-to-total ratio, would have to be determined for each gamma ray and included.

The problem now reduces to determining \( N_n, n = 1, \ldots, p \). The derivation presented in the previous chapter for determining the minimum value of the quantity given by equation 8

\[
T = \sum_{i=1}^{400} w_i \left( R_i - \sum_{n=1}^{p} A_{in} B_n \right)^2
\]

may be taken over exactly as presented if we now identify the \( B_n, n = 1, \ldots, p \) with the total area under the complex spectrum attributable to a gamma ray of energy \( H_n \).

Thus we have, using the same notation as before, from equations 10 and 12

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

\[
V_B = \sum_{i=1}^{400} w_i \left( R_i - \sum_{n=1}^{p} A_{in} B_n \right)^2 \frac{1}{400 - p} (A^T W A)^{-1}
\]

\[
B_n = N_n
\]

The relative intensities may be calculated by substituting these values of \( N_n \) into equation 14.

In the energy region below 600 keV this procedure utilizes the better features of the method developed by Reynolds (13) and others (14, 15, 16, 17). It eliminates any errors that might have been due to the use of incorrectly calculated response functions. As will be described in chapter 4, the dis-
tance from the source to the IaI crystal is 261 centimeters in
the present work. This contrasts with the source to crystal
distance of 10 centimeters used in the work by Trombka (15,
16) and Heath (17). For a source-to-crystal distance of 261
centimeters there is an increase (over that obtained at closer
distances) in the ratio of number of counts detected in the
photopeak to the total number of counts detected. This is due
to the fact almost all of the gamma rays which strike the crystal
are going essentially parallel to the axis of the crystal and
thus have a high probability of losing all of their energy in
the crystal. Further, the collimator effectively screens the
crystal from those gamma rays which have been scattered. The
reduction of the Compton continuum of a gamma ray of a given
energy means that the background is less for the lower energy
gamma rays. It therefore is possible to get a better signal-
to-noise ratio with a subsequent improvement in accuracy.

The present method eliminates several corrections re-
quired in the crystal diffraction method of measuring gamma
intensities. The correction for the energy dependent re-
fectivity of the crystal is eliminated because the diffrac-
tion crystal is not used in the recording of the composite
spectrum. Since the intense source is not used for determin-
ing the composite spectrum there is no need to make the un-
certain correction for self absorption in the source and the
source container. Finally the correction for the half life of
the isotope is not necessary.
EXPERIMENTAL CONDITIONS

This chapter will describe the experimental conditions under which the data were taken. The equipment that was used will be discussed and the experimental techniques employed will be presented. The equipment used consisted of a bent-crystal spectrometer, a linear amplifier, a voltage stabilizer, and a 400-channel analyzer.

The bent-crystal spectrometer used here is patterned after the one described by Seppi et al. (29). It consists of three main components: a lead-shielded source holder, a curved germanium crystal unit coupled to the sine screw assembly, and the collimator-detector assembly which rides on a curved track (Figure 3). In this spectrometer the source holder remains fixed and the collimator-detector assembly moves so as to maintain the Bragg condition. Two types of source holders were designed and built as shown in Figures 4 and 5. Essentially, the source holder consists of a device to hold a one-inch-long quartz capillary tube containing a one-curie source in a vertical position. The source is positioned 2 meters from the diffraction crystal or 2.61 meters from the NaI detector. Surrounding the source holder is a 5 inch lead shield to absorb the gamma radiation, thereby permitting free movement of personnel near the intense source.

The diffraction crystal unit consists of a germanium crystal mounted on a post in such a way that the crystal can
Figure 3. A line drawing of a bent-crystal spectrometer.
Figure 4. A bent-crystal spectrometer source holder and shield.
Figure 5. A bent-crystal spectrometer source holder and shield.
rotate about a vertical axis through its center. A rigid arm is attached to the crystal mount and is coupled to a precision screw mechanism. The crystal itself is a 3 by 2 3/4 inches by 1 1/2 millimeters thick single crystal of germanium. The crystal is cut so that the (400) planes are used in the diffraction. The crystal is polished and then clamped between two steel clamping blocks cut so that the crystal is bent to a 2 meter radius as described by DuMond (26). The crystal is mounted so that in the zero position the arm which leads to the precision screw is at right angles to the precision screw. The crystal is caused to rotate by turning the precision lead screw. The arm is constrained to move as the screw is turned, rotating the crystal. It is then possible to relate the number of turns of the screw or the linear distance the arm has moved to the sine of the angle through which the crystal has rotated. Because of this characteristic, the label of sine screw mechanism was given to the precision screw and its mounting. Seppi et al. (29) give a description of the operation of the sine screw mechanism.

The collimator-detector system rests on a platform that is constrained to rotate on a curved track whose center is the axis of rotation of the germanium crystal. The collimator consists of 30 lead plates 0.040 inches thick. The plates are 0.056 inches apart near the NaI crystal and 0.040 inches apart near the germanium crystal. If the plates of the collimator were extended they would intersect on the focal circle. Im-
mediately behind the collimator is a Harshaw Integral Line Assembly. This unit, which consists of a 3X3 inch NaI(Tl) crystal and a photomultiplier tube, was hand picked for optimum resolution. Behind the Integral Line Assembly is an RIDL Model 10-17 transistorized preamplifier. The collimator-detector system is electrically coupled by a Selsyn generator and motor to the precision lead screw. When the crystal turns through an angle $\theta$ the collimator-detector system turns through an angle $2\theta$. It can be seen from Figure 2 that this is precisely the relationship which must exist if diffraction is to be observed as described earlier. The only function of the spectrometer in this experiment is to serve as a gamma ray monochromator so that the crystal may be exposed to mono-energetic gamma rays and the monoenergetic spectrum may be recorded.

The electronic components are connected to a Stabiline voltage regulator which corrects for fluctuations in the line voltage. An RIDL Model 30-19 linear amplifier was used to amplify the voltage pulses from the preamplifier. A Cosmic Radiation spectrastat was used to provide the high voltage power. The multichannel analyzer was the RIDL Model 34-12B which has 400 channels. Output from the analyzer was in the form of punched paper tape. This tape was converted to IBM punched cards by an IBM tape to card converter. The experimental setup described above made it possible to take large
quantities of data quickly and efficiently.

The sources used in the experiment were of two types: a line source with an activity of about 0.2 curies was used to obtain the response functions of the monoenergetic gamma rays; and a weak, 1-millicurie source was used for the measurement of the composite gamma-ray spectrum. The strong source was obtained by filling a one inch long quartz capillary with 1.8 milligrams of Yb 168 enriched to 19.5%. The capillary had an inside diameter of .004 inches and an outside diameter of .040 inches. The Yb was in the chemical form Yb₂O₃. The source was irradiated in the reactor at the Argonne National Laboratory in a neutron flux of 10¹³ n/cm² for a period of two weeks. At the time of its arrival its activity was approximately one curie, but at the time the final measurements were taken its activity was down to about .2 curies. This strong line source was used to obtain the monoenergetic response functions of the NaI crystal.

The source described above was not used to obtain the composite spectrum for several reasons. In the first place the source is too intense to expose the crystal to the direct beam. In the second place our method of determining gamma-ray relative intensities seeks to avoid the corrections for the self absorption in the source and the source holder. Finally the gamma rays which would be backscattered in the dense shield behind the source would reach the crystal and produce back-
scatter peaks which are not present in the monoenergetic spectrum. They are not present in the monoenergetic spectrum since gamma rays that are backscattered are much lower in energy and would not satisfy the same focussing or Bragg condition as the original gamma ray. Thus a source, crystal, detector-collimator geometry which would accept the original gamma ray will not accept the scattered gamma ray. From these considerations it is apparent that it is important to minimize the amount of mass immediately behind the source in order to minimize the backscattering in the complex spectrum.

For these reasons it was decided to use a second weaker source to record the complex spectrum. The weak source was made by machining a brass ring 1 3/4 inches in diameter and 3/16 inches high. A piece of aluminized mylar 0.00025 inches thick was creased to produce a line. The mylar sheet was then glued to the brass ring with an epoxy base resin. Two drops of cement were placed one inch apart on the crease to define the ends of the line source. The Yb$_2$O$_3$, which had been irradiated for the same length of time as the strong source, was taken into solution with dilute HCl. The liquid was evaporated to dryness and a known volume of distilled water to which a wetting agent had been mixed was added. One drop of the solution containing 1 millicurie of Yb 169 was then deposited in the center of the fold on the aluminized side of the mylar. It was found that the source adhered better to the aluminized
mylar than to the plain mylar. The drop was then spread out to fill the crease made previously. The wetting agent assisted the liquid in spreading out uniformly. The two spots of glue defined the ends of the source such that a one inch line source was produced centered in the brass ring. A 0.00025 inch thick piece of plain mylar was then glued to the other side of the brass ring making a completely sealed source with an extremely thin backing. The brass ring was then mounted in the lucite ring holder as is indicated in Figure 6. The source was placed on the focal circle at the same distance from the detector as the strong source. For the recording of the composite spectrum the diffraction crystal was removed and the collimator-detector was rotated until maximum intensity was observed. The collimator was left in place since it was effective in reducing scattering.

The data was taken in the following manner. The curved germanium diffraction crystal was removed and the weak source was placed in position. The collimator-detector system was then rotated until a maximum in intensity was obtained. The composite spectrum of the NaI detector was then recorded in the multichannel analyzer in this position. Following the measurement of the composite spectrum, the collimator and detector were rotated until the background counting rate was reached. A background reading was then taken. After the background measurement the weak source was removed and the
Figure 6. Line source and source holder used to measure composite spectrum.
germanium diffraction crystal was replaced. The spectrometer was then adjusted until a maximum in intensity was noted for a gamma ray of a given energy. The spectrometer was then set at the screw division reading corresponding to this maximum and the monoenergetic response function of the NaI crystal was recorded in the multichannel analyzer. Figure 7 shows the order of appearance of the various orders of the several lines in the Tm 169 spectra and the points where the monoenergetic response functions were measured. After the monoenergetic response functions were measured, the precision screw was turned several revolutions; and avoiding neighboring lines, the background was measured. This was done on both sides of the screw division setting where the response function was measured.

This technique of measuring background is very effective since a very slight movement of the screw is needed to reach background counting rates. Because the detector carriage has moved very slightly the geometry is essentially the same as it was before, and the same scattered radiation from the room should be detected as when the response functions were taken. It is therefore possible to obtain accurate background readings and hence to eliminate the effect of the background from the response functions. In measuring the first order 130.53 keV monoenergetic spectrum an interesting effect occurred. A second order 261.0 keV peak appeared in the spectrum and did not appear in the background. It was no problem to eliminate
Figure 7. Screw division settings for a germanium bent-crystal spectrometer corresponding to the various orders of gamma and X-rays of Tm 169 following electron capture from Yb 169. * indicates screw division settings for measurement of monoenergetic response functions.
the peak by setting the value of the monoenergetic spectrum and the background equal to zero for those multichannel analyzer channels higher in energy than the photopeak of the 130.53 keV line. This procedure is generally possible since photopeaks due to higher order diffraction of gamma rays of greater energy will always appear in channels higher in energy than the photopeak of interest.

Using the tables of Grodstein (20) and McGinnies (31), corrections to the relative intensities obtained from the composite spectrum were made for scattering and absorption in the air path between the source and crystal as well as for absorption in the aluminum cap on the NaI crystal. It was found that corrections due to absorption in the 0.00025 inch mylar covering on the weak source were negligible. The efficiencies of the NaI crystal were determined from tables by Vegors et al. (32) and Bell et al. (30). The listing by Vegors et al. (32) gives the efficiencies for point, disc, and line sources for a variety of source-to-detector distances. It was found that at 10 centimeters source-to-crystal distance the relative efficiencies in the energy region 50-600 keV did not differ by more than .5% whether or not the figures for the line or point source were used. It is expected that at 261 centimeters the difference would be even less. Using the two sets of efficiencies for a point source (31, 30) it was
possible to determine the way in which the efficiencies for a given energy were changing as a function of source-to-crystal distance. From this information it was possible to interpolate graphically and thus determine the efficiencies as a function of energy at a source-to-detector distance of 61 centimeters. It was estimated that the uncertainties in the NaI detector efficiencies were about 1%. The total error due to all corrections was estimated to be 1.5% for the Tm 169 gamma-ray relative intensities.
EXPERIMENTAL RESULTS

The relative intensities of the Tm 169 gamma rays emitted following electron capture from Yb 169 are presented in Table 1. These intensities were determined by a weighted average of the three sets of data for which the fit parameter m from equation 11 was less than two. The values of the weights used in computing the average value of the relative intensities of each of the gamma rays were the reciprocals of the squares of the estimated errors in the relative intensities for each measurement. Recent measurements of the relative intensities of the same gamma rays by Alexander and Boehm (12) and Grabowski et al. (31) are displayed for comparison. The measurements of Alexander and Boehm (12) and Grabowski et al. (31) were obtained by measuring the intensities of the diffracted line profiles from bent-crystal spectrometers. The Tm 169 measurements by Alexander and Boehm (12) are thought to be the most accurate relative intensity determinations which have been obtained for this isotope with a bent-crystal spectrometer. Their method is, of course, quite different from the present method in which the intensities are obtained by unfolding the composite pulse height distribution from the NaI detector using the response functions obtained from monoenergetic gamma rays in the various diffraction peaks.
Table 1. Relative intensities of Tm 169 gamma rays following electron capture from Yb 169

<table>
<thead>
<tr>
<th>Energy keV</th>
<th>Rel. intensities</th>
<th>Standard deviation</th>
<th>Expt. % total</th>
<th>Expt. % total</th>
<th>Ref.</th>
<th>Ref.</th>
<th>R₁</th>
<th>R₂</th>
<th>R₃</th>
</tr>
</thead>
<tbody>
<tr>
<td>93.60</td>
<td>7.4 ± .5 ± 6</td>
<td>± .4 ± 5</td>
<td>7.2</td>
<td>6.6</td>
<td>1.03</td>
<td>1.12</td>
<td>1.09</td>
<td></td>
<td></td>
</tr>
<tr>
<td>109.78</td>
<td>47 ± 1 ± 3 ± 5</td>
<td>± 5 ± 1 ± 1</td>
<td>50</td>
<td>51</td>
<td>.95</td>
<td>.93</td>
<td>1.02</td>
<td></td>
<td></td>
</tr>
<tr>
<td>118.20</td>
<td>5.4 ± .4 ± 8</td>
<td>± .5 ± 1 ± 6</td>
<td>5.2</td>
<td>4.8</td>
<td>1.04</td>
<td>1.12</td>
<td>1.08</td>
<td></td>
<td></td>
</tr>
<tr>
<td>130.53</td>
<td>32 ± 1 ± 3 ± 3</td>
<td>± 1 ± 6 ± 1</td>
<td>31</td>
<td>32</td>
<td>1.02</td>
<td>.99</td>
<td>1.00</td>
<td></td>
<td></td>
</tr>
<tr>
<td>177.24</td>
<td>62 ± 2 ± 4 ± 1</td>
<td>± 2 ± 2 ± 1</td>
<td>62</td>
<td>58</td>
<td>1.00</td>
<td>1.07</td>
<td>1.07</td>
<td></td>
<td></td>
</tr>
<tr>
<td>197.97</td>
<td>100 ± 2 ± 2 ± 2</td>
<td>± 0 ± 1 ± 0</td>
<td>100</td>
<td>100</td>
<td>1.00</td>
<td>1.00</td>
<td>1.00</td>
<td></td>
<td></td>
</tr>
<tr>
<td>261.00</td>
<td>4.7 ± .6 ± 13</td>
<td>± .5 ± 12</td>
<td>4.8</td>
<td>4.6</td>
<td>.97</td>
<td>1.01</td>
<td>1.04</td>
<td></td>
<td></td>
</tr>
<tr>
<td>307.70</td>
<td>28 ± 1 ± 4 ± 1</td>
<td>± 1 ± 3 ± 3</td>
<td>28</td>
<td>31</td>
<td>1.00</td>
<td>.92</td>
<td>.904</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

a From reference Alexander and Boehm (12).

b From reference Grabowski et al. (31).

c Ratio of relative intensities of present measurements to those of Alexander and Boehm (12).

d Ratio of relative intensities of present measurements to those of Grabowski et al. (31).

e Ratio of relative intensities of Alexander and Boehm (12) to those of Grabowski et al. (31).
The total deviations listed for column 3 for the present measurements were obtained by adding a 1.5% systematic error (due mainly to the NaI efficiency correction) to the standard deviations obtained from three runs. The fourth column in the table lists the percentage deviation for the total standard deviation in each measurement. The fifth and sixth columns give the standard and percent standard deviations determined from the three sets of data.

For comparison purposes the last three columns of the table give the ratios of the present measurements to those of Alexander and Boehm (12), \( R_1 \), the present measurements to those of Grabowski et al. (31), \( R_2 \), and finally the ratios Alexander and Boehm's (12) measurements to those of Grabowski et al. (31). Figure 8 shows these ratios plotted for visual display. It will be noted that the best agreement is obtained for \( R_1 \). The dashed lines indicate the square root of the sum of the squares of the reported errors in the present measurements and in those of Alexander and Boehm (12). Both \( R_2 \) and \( R_3 \) have the same shape except for the 110 keV gamma ray. This indicates that the present measurements agree with those of Alexander and Boehm (12) that the measurements of Grabowski et al. (31) are either too large or too small.

The accuracy of the present method is very apparent in the excellent agreement of these intensities with those of Alexander and Boehm (12). All of the values obtained agree
Figure 8. Comparison of reported values.
within the sum of the square of the errors assigned to each measurement. The largest deviation is about 5% for the 109.78 keV gamma ray, and it is believed that the present result is the more accurate one. The agreement with the results of Grabowski et al. (31) is also reasonably good, although they list no errors for their measurements and their measurements appear to be less accurate than those of Alexander and Boehm (12). As seen in Figure 8 there does not seem to be any systematic correlation with energy of the differences for the various sets of measurements.

It is apparent from Table 1 that the relative intensities of the 336.5 keV, 240.4 keV, 156.66 keV, 117.25 keV, and the 63.12 keV lines were not measured. The first four gamma rays are from ten to one hundred times weaker than the weakest line in the present measurement. It would be expected that only at maximum source strength would there be a possibility of determining the intensities of these lines. At the time the present measurements were taken the source was not strong enough to permit the determination of these intensities. The 63.12 keV line appears at roughly the same place in the composite spectrum as do the Tm 169 X-rays. The combination of these lines forms one large hump. If one were to measure the intensity of the 63.12 keV line the intensities of all of the X-rays would have to be determined also. It was not possible to stabilize the electronics sufficiently
with the equipment available to allow the time required to measure all of the X-ray response functions and the 63.12 keV line in addition to the other gamma rays. As it becomes possible to stabilize the electronics for long periods of time it should be possible to improve the statistics enough to determine the intensities of these lines also.

Since it was decided to eliminate the contributions to the composite spectrum from all X-rays and gamma rays lower in energy than the 93.60 keV line, it was necessary to normalize on a number of counts less than the total number of counts and to compute a curve-to-total ratio analogous to the peak-to-total ratio of equation 5. This procedure was discussed in conjunction with equation 14 of Chapter 3. The only gamma ray affected by this calculation was the 93.60 keV line. Since its iodine escape peak fell below the cutoff energy, the curve-to-total ratio was 5% smaller than the others.

Figure 9 shows two typical monoenergetic response functions used in the determination of the relative intensities. The small number of counts in the Compton continuum compared to the number in the photopeak is apparent. The present measurements yield a peak to total ratio 10% greater than that reported by Heath (32) for the 307.7 keV line. This trend continued for the other gamma rays with the present values converging to Heath's (32) at lower energies. The reason
Figure 9. Typical monoenergetic response functions for 307.7 keV and 197.97 keV gamma rays.
for this difference is attributed to the greater source-to-detector distance in the present case. Since our source is 361 cm from a 3X3 inch NaI crystal, essentially all of the gamma rays that strike the crystal are going parallel to the axis of the crystal and thus have a high probability of losing all of their energy in the crystal. The source-to-crystal distance was 10 cm in Heath's (32) work.

Figure 10 summarizes the entire experiment. The top curve which has been displaced 200 counts for clarity is the experimentally determined composite spectrum. The next lower curve is the computed composite spectrum. The monoenergetic response functions which added together to produce this computed composite spectrum are shown below with dashed lines. It is seen that the computed composite spectrum follows the experimental composite spectrum very well. The lower portion of the figure presents the deviations between the experimental composite spectrum and the computed composite spectrum divided by the square root of the number of counts in the experimental composite spectrum. It is seen that the deviations are purely statistical in nature. A demonstration of the power of the method is that the weak 118.20 keV line is fitted accurately even though it is swamped by the neighboring lines.

Trombka (15, 16) has given standard deviations of about 10% for his intensity measurements, and Heath's (17) test
Figure 10. Tm-169 composite spectrum from 75 keV to 350 keV with computed composite spectrum and monoenergetic components.
case is quoted at around 1%. Alexander and Boehm (12) give errors of 5% while Hultberg (2, 3) quotes errors of 5%. It is estimated that by improving the stability of the detection electronics and increasing the source strengths so that better counting statistics will be obtained, the present method should yield relative intensity measurements accurate to at least 3% for gamma rays in those cases for which there are approximately ten gamma rays or less reasonably separated in energy. It is believed that the present method will provide the most accurate and convenient method for obtaining gamma-ray relative intensities for many decay schemes of interest.
LITERATURE CITED


APPENDIX

The computer program used to determine the relative intensities of a maximum of 20 gamma rays on a 400 channel analyzer is described in this appendix. Basically the program is designed to perform the following calculation as given by equation 10

$$B = (A^T W A)^{-1} A^T W R.$$  

In order to accomplish this, the program first causes a parameter card to be read in which sets the size of the A matrix and defines certain options. The program then reads in the matrices A and R. Next the weight matrix W is determined. Depending on the value of IW, a parameter read in, it will read the weights as data, set them equal to one, or compute them. Since the background is subtracted from the observed composite spectrum before the fitting begins, the expected deviation in the number of counts in channel i is

$$s_i = \sqrt{R_i^2 + Rb_i^2}, \quad i = 1, \ldots, 400$$

where $R_i$ is the measured number of counts in the complex spectrum and $Rb_i$ is the number of counts of background in channel i. We have then

$$w_i = \frac{1}{\sqrt{R_i^2 + Rb_i^2}}$$

if the weights are computed.

After determining W the program will either normalize the Ain on the photopeak area or the total area depending
on the value of a parameter, \( N \). If the normalizing is on the photopeak area an Ames Lab library subroutine NLLS is used to fit a Gaussian to the leading edge of the photopeak after the average background is subtracted. The \( A_{in} \) are then divided by the area under the Gaussian. If the normalization is to be on the total area then the average background is subtracted, and the number of counts in each channel of a monoenergetic spectrum is divided by the total number of counts in the monoenergetic spectrum. This is the method of normalization used for the present measurements.

Following the normalization the matrix product \( A^T W A \) is formed and an Ames Lab library subroutine MATINV is called to take its inverse. The inverse is then multiplied times the matrix product \( A^T W R \). Having determined \( B \) the program now determines the fit parameter \( m \) as defined by equation 11. The fit parameter \( m \) is then printed out and the product \( m(A^T W A)^{-1} \) is formed and printed out.

Finally a subroutine COMP is called to compute the relative intensities by finding the largest \( B_n \) and normalizing on this intensity. COMP also computes all the corrections and the estimated error in the relative intensities.

The program will print the normalized \( A_{in} \), the \( A_{in} B_n \), the \( R_i \), and the \( \sum_n A_{in} B_n \), \( i = 1, \ldots, 400 \), as desired.

A test case was run through in which two Gaussians
normalized to unity were added channel by channel to form a composite spectrum. The peaks of the two Gaussians were 1 channel apart and each had approximately the same amplitude. The program computed the areas under the composite spectrum within 0.1%.

The flow chart and listing of the program follows. The first program given is the main program. It is followed by a subroutine COMP which calculates the relative intensities knowing the $B_n$ as computed in the main program. Following COMP is a subroutine NLLS which is used to fit a Gaussian to a photopeak if the normalization is on the photopeak area. NLLS calls the next subroutine SUBRT as a part of its calculation. In order to compute the inverse of a matrix the Ames Lab Computer Services Group subroutine MATINV is used which is not listed. Following the programs a portion of the output showing the fit parameter $m$, $m(\text{ATWA})^{-1}$, and the relative intensities with their standard deviations is given.
Figure 11. Flow chart for computer program.
Figure 12. Main program.
111 READ 1110, (A(I,J), I=1, IA)
120 READ 1110, (W(I), I=1, IA)

IF (1W) 121, 125, 120
120 READ 1110, (W(I), I=1, IA)
GO TO 400
121 DO 122,1+1,IA
OMEGA = IDENTITY MATIX
GO TO 400
125 DO 129, I=1,IA
CALCULATE OMEGA FROM RHO
GO TO 129
128 W(I)=1./ZZ
129 CONTINUE

DO 134 I=1,IA
BZ=O.
134 READ 1110, (B(I,J), I=1, IA)

137 BZ=BZ+A(J, I)
135 DO 134, I=1,IA

136 PRINT 1122
1122 FORMAT (17H DIVIDING BY BZ=0)
STOP 89

133 DO 139 IX=1, IA
READ 1120, (IU(IX), IX=1, IA)

NDFP=IU-IL+1
401 READ 1110, (B(J), J=1, IA)
403 NFDP=400,403

IF (1W) 405, 404, 403
139 GP1(1)=GP(1)

138 SIGMA=GP(1)/SQRT(1.3862944)
GP(1)=SIGMA
DO 139 I=1,3

Figure 12. Continued.
Figure 12. Continued.
C DO 220 I=1,JA
STORE AWR AND AWA FOR MATINV
REL 066

C DO 225 I=1,JA
C AI(J,J) = AWR(I) + AWR(I)
REL 068

C DO 230 I=1,JA
INVERT AWA AND SAVE FOR B
REL 070

C CALL MATINV( C, JA, B, 20, 1, DET, WORK1, WORK2, WORK3, WORK4)
REL 071

C IF (DET .LE. 0.0) CALL SINGULAR MATRIX MESSAGE
REL 073

C 1 VECTOR (/X F9.0,01F10.0/)
REL 077

C DO 235 J=1,JA
REL 079

C PRINT 1235, (C(J,J),J=1,JA)
REL 080

C FORMAT (1520.8)
NOT SINGULAR
REL 083

C A(-1) = A =I AND PRINT
REL 084

C DO 250 I=1,JA
REL 085

C DO 255 J=1,JA
REL 086

C FORMAT (8H A(I,J))
C I=1 . JA
REL 087

C PRINT 1250, I, (A(I,J),J=1,JA)
REL 090

C FORMAT (13H I=1,JA)
REL 091

C CONTINUE
REL 093

C PRINT 1260, (B(J,J),J=1,JA)
REL 094

C PRINT (13H BETA VECTOR FID-5E24.81)
REL 095

C FIAJA = IA - JA
REL 096

C IF (FIAJA) 262, 261, 262
REL 097

C PRINT 2652
REL 098

C 2652 FORMAT (8H FIAJA=01
REL 099

C FM = 0.
DO 502 I=1,JA
DO 502 I=1,JA
REL 099

C DO 503 J=1,JA
REL 099

C FM = 0.
DO 505 J=1,JA
REL 099

C FM = 0.
DO 505 J=1,JA
REL 099

C PRINT 2752, J, (A(I,J),J=1,IA)
REL 099

C 2752 FORMAT (6H FID-5E24.81)
REL 099

C FORMAT (13H FID-5E24.81)
REL 100

C DO 270 N=1,JA
REL 101

C BA(I) = BA(I)+AI(J)
REL 102

C DO 270 N=1,JA
REL 103

C FM = FM + WII * BARAA
REL 104

C FM = FM / FIAJA
REL 105

C PRINT 1280, FM
REL 106

C 1280 FORMAT (6H FID-15.8/13H M=(A(T)*W=A) 10% 30H LOWER TRIANGULAR PRED
REL 107

C INTENDED ONLY //1
C DO 290 I=1,JA
REL 108

C DO 295 J=1,JA
REL 110

Figure 12. Continued.
Figure 12. Continued.
Figure 13. Subroutine COMP
Figure 14. Subroutine NLLS
SUBROUTINE SUBRT (J,W,X,Y,GP1,DERIV,YC,F1, WI)

DIMENSION W(250),X(250),Y(250),GP1(8), DERIV(8)

YC=GP1(1)* EXPF(-.5*((X(J)-GP1(2))/GP1(3))**2)

F1=Y(J) - YC

DERIV(1)=EXPF (-.5*((X(J)-GP1(2))/GP1(3))**2)

DERIV(2)=(GP1(1)* (X(J)-GP1(2)) /GP1(3)**2)*DERIV(1)

DERIV(3)=(X(J)-GP1(2))/GP1(3)*DERIV(2)

WI=W(J)

RETURN

END

Figure 15. Subroutine SUBRT.
Figure 16. Portion of output from run 1.
<table>
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<tr>
<th>Row</th>
<th>0.40728759E-04</th>
<th>0.17373414E-04</th>
<th>0.18654939E-04</th>
<th>0.2088211E-04</th>
<th>0.11728356E-04</th>
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<td>0.10708272E-03</td>
<td>0.89067268E-05</td>
<td>0.31275824E-05</td>
<td>0.23955930E-05</td>
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<td>0.19735911E-05</td>
<td>0.4610687E-06</td>
<td>0.11728356E-04</td>
<td>0.11728356E-04</td>
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<td>0.1021613E-06</td>
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<td>0.1377414E-06</td>
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<tr>
<td>Row 5</td>
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<td>0.7165050E-07</td>
<td>0.3849887E-06</td>
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<td>0.11728356E-04</td>
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<td>0.2777414E-06</td>
<td>0.49706935E-06</td>
<td>0.15787063E-06</td>
</tr>
</tbody>
</table>

**Figure 18. Portion of output from run 3.**