GAMMA-GAMMA DIRECTIONAL CORRELATION IN TITANIUM-46 (Ti\(^{46}\))

by

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GAMMA-GAMMA DIRECTIONAL CORRELATION IN TITANIUM-46 (Ti$^{46}$)

INTRODUCTION

One of the most important problems of nuclear physics today is to understand the nature of nuclear forces. The nature of the interactions involved still being not completely known, one of the lines of attack to improve the situation is the determination of the properties of nuclear energy levels. Some of these properties are energy, lifetime, total angular momentum, parity, magnetic moment, electric quadrupole moment and partial width. All these properties are defined for excited levels as well as for stable or unstable ground states; however the experimental determination of these properties is quite different for stable and unstable states because of the obvious reasons involving time and the quantity of material available. The determination of the dynamical moments (spins) and parities associated with excited states is much more difficult, because at present in all but a few cases direct methods are inapplicable. Much work has consequently been carried out to establish the character of the nuclear decay schemes, from which spins and parities may be inferred. So as to make the spin and parity assignments unique, it is
imperative to use as many different methods as possible for their determination. The experimental values of these properties can then be compared with the theoretical values based upon certain assumptions about the nuclear forces.

One of the well-established experimental results, as listed by Evans (14, p. 357), which a nuclear model should be able to explain is that even-even nuclei have zero spin. One of the models that does so is the shell model of the nucleus according to which an even number of identical nucleons having the same \( l \) and \( j \) will always couple to give even parity, zero total angular momentum, and zero magnetic moment. Nuclear spins different from zero manifest themselves through magnetic moments, and any deviations from spherical symmetry give rise to electric quadrupole moments.

Some of the methods used to determine the spins and parities of the short-lived nuclear excited states are:

1. Deuteron stripping reactions at low energy provide a method for deducing spins and parities of some nuclear states. Some measurements of \( (d,p) \) and \( (d,n) \) cross-sections have been made (14, p. 248) and the results give the \( l \)-values of the captured nucleon resulting in a direct relation between the parities of the target nucleus and the excited states of the product nucleus. The results also set limits for the spins of the final states and they are least ambiguous when the deuteron energies employed are sufficient for coulomb effects to be small, but still low enough to be in the low energy region.
2. Spins and parities may also be inferred from the study of beta decay. From the shape of the beta spectra and the Gamow-Teller selection rules (which appear to describe most cases of beta decay) or Fermi selection rules one can decide upon the class of transition and the changes in spin and parity if the $\textit{ft}$ value of the transition, determined experimentally, is known. Consequently assignment of spin and parity of the excited nuclear level can be made if the spin and parity of the parent nucleus are already known.

3. A light nucleus may de-excite itself from its low-lying excited states either by gamma ray emission or by emitting electrons by the process of internal conversion or internal pair formation. By studying the nature of the radiation emitted and keeping in mind the fact that even-even nuclei have zero spin and even parity one can assign limits to the spins and parities of the excited states of such nuclei. It has been shown (38) that the magnitudes of the internal conversion and pair production coefficients are dependent on the spin and parity changes in the transition.

4. Correlation measurements of the polarization of the electric vector of the gamma rays may be used to determine the multipolarity of the gamma rays and hence the parity change of the transition involved which can be used to infer the parity of the nuclear states under investigation.

5. Directional correlation measurements between two or more gamma rays, if the lifetime of the intermediate state is short, determines the multipole order of the gamma rays and gives information about the spins and parities of the nuclear states involved. The directional correlation measurement alone never allows complete establishment of the decay scheme and has to be supplemented by other methods.

One basic requirement of the theory of angular correlation is that the orientation of the nuclear spin
of the intermediate nuclear state must remain essentially constant during the lifetime of that state. This limits the method to cases where the lifetime of the intermediate state is extremely short so that the nucleus does not precess appreciably during this time; if the lifetime of the state is longer than $10^{-10}$ sec., the electromagnetic fields of the electron shells and the surroundings tend to make the nucleus precess. Too much of this effect makes the spin direction of the intermediate state no longer well-defined when referred to the direction of the first gamma ray emitted. Precession can therefore smear out an otherwise anisotropic angular correlation making it isotropic, or more nearly so.

Although the experimental difficulties and more limited application have slowed down the progress of angular correlation work, yet it has established itself as one of the most important tools in the field of nuclear spectroscopy.

Keister and Schmidt (26) first proposed a decay scheme for Sc$^{46}$, but the scheme most commonly accepted is shown in Figure 1, as reported by Strominger, Hollander, and Seaborg (46, p. 622). The half life of the intermediate excited state of Ti$^{46}$ has been measured by various workers. Bertolini (5) measured the lifetime of the first excited state of Ti$^{46}$ using
Figure I. Decay Scheme of Sc$^{46}$
a high resolution nanosecond time analyzer and found an upper limit of $10^{-11}$ sec. Azuma (3) using a fast coincidence apparatus with a resolving time of 2.5 nanoseconds found the upper limit of the lifetime to be less than $5 \times 10^{-11}$ sec. These values check well with those of other investigators who used different methods e.g. coulomb excitation. Hence there should be negligible perturbation of the spin orientation of the intermediate state of Ti$^{46}$ and the nature of the angular correlation should be preserved.

Keister and Schmidt (26) from their study of the beta spectrum of Sc$^{46}$ characterized the transition by $J = 2$ (no change in parity) i.e. a second forbidden transition. This result led them to assign to the ground state of Sc$^{46}$ a probable spin of $4^+$. Their conclusion supported earlier proposals for the decay scheme of Sc$^{46}$ (21, p. 185). This conclusion has further been strengthened by the values of log $ft$ for the two beta transitions observed in the case of Sc$^{46}$. Log $ft$ values of 6.2 and 12.9 for the transitions to the second and first excited levels of Ti$^{46}$ respectively, suggest the spin and the parity for the ground state of Sc$^{46}$ to be either $4^+$ or $5^+$ provided Ti$^{46}$ levels are $4^+$ and $2^+$ respectively. However the spin assignment of 5 to the ground state of Sc$^{46}$ gives a negative value
for the beta-gamma circular polarization coefficient which is in contradiction to the experimental results of Bohm and Wapstra (8). Furthermore the shell model predicts a $\frac{f}{2} - \frac{f}{2}$ configuration for the odd-proton and neutron, and hence even parity for the ground state of Sc$^{46}$. The assignment of spin 4 to the nuclear state in question is also consistent with Nordheim's rule (34) for the coupling of odd-neutron and odd-proton groups belonging to the same Schmidt group (14, p. 156). All this evidence establishes the fact that the ground state of Sc$^{46}$ has a spin of 4 and has even parity.

Experimental studies of the internal conversion coefficients of the transitions from the decay of Sc$^{46}$ have been undertaken by various investigators. In each case the measurements were made on either K-, K + L-, K + L + M-, or K$_{\text{total}}$ conversion coefficients for the gamma rays of energies 0.89- and 1.12-Mev emitted, following beta decay of Sc$^{46}$, by the residual Ti$^{46}$ while returning to the ground state. Peacock and Wilkinson (36) used a 180° spectrometer of fair resolution. Moon, Waggoner, and Roberts (33) made similar measurements with a double-coil thin lens spectrometer whereas Porter and Cook (37) used a 13-cm radius of curvature semi-circular magnetic spectrometer. Sturken, O'Friel, and
Webber (47) investigated the problem using two different methods. Recently Hultberg, Frey, and Hamilton (24) measured the coefficients using an iron-free double focusing spectrometer. The results of the different investigators are:

<table>
<thead>
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<th>Reference</th>
<th>Nature of Coeff.</th>
<th>0.89-Mev</th>
<th>1.12-Mev</th>
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<tr>
<td>(36)</td>
<td>( a_K )</td>
<td>8</td>
<td>4</td>
</tr>
<tr>
<td>(33)</td>
<td>( a_{K+L+M} )</td>
<td>1.74( \pm )0.08</td>
<td>1.51</td>
</tr>
<tr>
<td>(37)</td>
<td>( a_{K+L+M} )</td>
<td>1.36</td>
<td>0.607</td>
</tr>
<tr>
<td>(49)</td>
<td>( a_{K+L+M} )</td>
<td>1.9</td>
<td>0.88</td>
</tr>
<tr>
<td>(47)</td>
<td>( a_{total} )</td>
<td>2.60( \pm )0.50</td>
<td>1.34( \pm )0.15</td>
</tr>
<tr>
<td>(27)</td>
<td>( a_{total} )</td>
<td>1.55( \pm )10%</td>
<td>1.61</td>
</tr>
<tr>
<td>(24)</td>
<td>( a_{K+L+M} )</td>
<td>1.6 ( \pm ) 5%</td>
<td>0.96 ( \pm ) 5%</td>
</tr>
</tbody>
</table>

Although all the results mentioned above tend to indicate that both the gamma ray transitions following the 0.36-Mev beta ray from the ground state of Sc\(^{46}\) are electric quadrupole, there remains some doubt, on the basis of these observations, whether the transitions involved are pure E2 or not. This is especially so for the 1.12-Mev gamma ray whereas the internal conversion coefficient measurements together with the beta decay evidence
indicate clearly the E2 nature of the 0.89-Mev transition. Furthermore it can be seen that the values of internal conversion coefficients are not very dependent upon the mixing, in the transition, of different multipolarities. However, in directional correlation measurements the correlation function is very sensitive to the mixing ratios.

The various determinations discussed above show more or less conclusively that the first and second excited states of Ti$_{46}^{46}$ are $2^+$ and $4^+$, respectively. These assignments have been strengthened further by the angular correlation experiments of Novey (35); Metzger and Deutsch (31); Brady and Deutsch (9); Hayashi, Kawamura, and Aoki (23); and Bertolini, Bettoni, and Lazzarini (4). However it was felt worthwhile to re-measure the directional correlation with the improved techniques associated with a scintillation spectrometer. Furthermore it seems fruitful to try to determine transition mixing ratios which can then be used to test nuclear models and in this way possibly aid our understanding of the nature of nuclear forces.
THEORY OF DIRECTIONAL CORRELATION

The probability of emission of a nuclear particle by a radioactive nucleus depends on the angle between the nuclear spin axis and the propagation vector of the radiation. In general, the radiation pattern from a radioactive source is isotropic because the ensemble of nuclei have random orientations in space. The experimenter may reduce the random orientation of nuclei with finite moments by cooling the source to a very low temperature in a strong magnetic field or electric field gradient. The interaction between the nuclear magnetic moment and the magnetic field or that between the nuclear electric quadrupole moment and the electric field gradient produces a preferred orientation or alignment of the nuclear spin axes. This results in an anisotropic radiation pattern.

An alternative procedure consists of choosing only those nuclei whose spins lie in a preferred direction. This is possible if the nucleus decays by a cascade process with a sufficiently short-lived intermediate state. The first radiation is observed in a fixed direction, thereby selecting an ensemble of nuclei which have a nonisotropic distribution of spin orientations. The succeeding second radiation will then exhibit a certain angular correlation with respect to the first. Figure 2 shows the typical
The Cascade

The Experiment

The Result

The Theory

FIGURE 2. An Example of a Directional Correlation Experiment
aspects of a directional correlation measurement. The correlation function $W(\phi)$ is the probability that radiation 2 is emitted in the direction $\vec{k}_2$ relative to the direction $\vec{k}_1$ of radiation 1. Experimentally, one measures the number of true coincidences $N$ between the two radiations as a function of the angle $\Theta$ between the axes of the detectors. Because of the finite size of the detectors, $N(\Theta)$ is proportional to an average of the true correlation $W(\phi)$ over a range of angles $\Delta \phi$ distributed around $\Theta$. Therefore $N(\Theta)$ must be normalized and corrected to yield $W(\phi)_{\text{exp}}$. The comparison of $W(\phi)_{\text{exp}}$ with theory provides the desired information about the nuclear spins and multipole order of the radiation involved in the cascade.

It is essential to understand why a correlation exists; that is, why the coincidence counting rate depends on the angle between the propagation vectors of the two radiations. Consider a quantum mechanical transition in which a radiation is emitted from a state characterized by the quantum numbers $j_1$, $m_1$, leading to a state $j$, $m$. The radiation will have angular momentum $L$ and projection quantum numbers $M = m_1 - m$. In actual practice one observes the summation of transitions from all substates $m_1$ to all substates $m$. The angular distribution of the radiation is, for each $M$, dependent on $\phi$; however, it turns out the summation over $M$, which is a summation over
all substates, is independent of $\emptyset$. Hence the observed radiation pattern in a single transition from a non-oriented source is isotropic.

The situation is different for the observation of coincidence radiations. If one considers a single fixed value of $M$, then the pattern is not isotropic because a particular direction in space is being considered. This direction is the axis of quantization with reference to which $M$, $m_\parallel$ and $m$ are measured. If, of all the radiations emitted in all directions, one observes only that which proceeds in a certain direction, then this is equivalent to selecting a certain set of values of $M$. If one chooses the axis of quantization to lie along the direction of emission of the first radiation, $M$ will equal $\pm 1$ with $M = 0$ omitted. This follows from the fact that a plane wave can have only intrinsic spin along the direction of propagation. One does not sum over all values of $M$ and hence not all sublevels of $m$ will be equally populated. If this final state for the first transition is the initial state for the second transition, the emitted radiation pattern for the second transition is anisotropic.

The general theory of angular correlation has been successively developed by various authors starting with Hamilton (22). The progress in the theory after Hamilton's pioneering work was chiefly due to Lloyd (30),
Alder (2, p. 235), Falkoff (15), Deutsch (12, p. 196) and Goertzel (20). An excellent review article has been given by Biedenharn and Rose (6, p. 729).

The theory may be summarized in the following manner: The initial, intermediate and final quantum states may be characterized by \( \psi_1, \psi_i \) and \( \psi_2 \), respectively. Let \( H_1 \) be the interaction term in the Hamiltonian causing the emission of the first quantum and \( H_2 \) be that responsible for the emission of the second quantum. One assumes negligible perturbation of the intermediate state by the extra nuclear fields. This holds true if the lifetime of the state is very small compared to the Larmor period. An application of second-order perturbation theory (41, p. 168) leads to the probability that a nucleus, decaying through the cascade \( 1 \rightarrow i \rightarrow 2 \), emits two radiations in the directions specified by the propagation vectors \( \mathbf{k}_1 \) and \( \mathbf{k}_2 \), respectively. This probability is the correlation function

\[
W(\mathbf{k}_1, \mathbf{k}_2) = S \sum_{m_1 m_2 m} \left| \langle j_2 m_2 | H_2 | j m \rangle \langle j m | H_1 | j_1 m_1 \rangle \right|^2
\]

(1)

where \( j_i \) is the angular momentum of the \( i \)th state, \( m_i \) is the projection of \( j_i \) on the axis of quantization, \( j \) is the angular momentum of the intermediate state, and \( S \) denotes a summation over non-observed quantities such as the polarization quantum numbers if the experiment does not
involve polarization measurements. Equation (1) can be brought into a useful form by splitting the matrix elements into multipole components according to the angular momentum $L, M,$ and parity of the radiation. Then a transformation is performed, carrying the co-ordinate system of quantization over into the co-ordinate system of the radiation. The matrix elements of distinct multipole order are then split into geometrical factors and reduced matrix elements using the Wigner-Eckart theorem. Finally, the total expression is reduced using the properties of the Clebsch-Gordan coefficients (41, p. 38) and Racah algebra (41, p. 107). For pure radiation the correlation function may be written

$$W(\theta) = \sum_v a_v P_v(\cos \theta)$$

where

$$a_v = F_v(L_1 j_1 j) F_v(L_2 j_2 j)$$

$$F_v(L_1 j_1 j) = (-1)^{j_1 - j - 1} (2j + 1)^{1/2} (2L_1 + 1) C(L_1 L_2 j; 1, -1)$$

$$\times W(jj j_1 L_1; v j_1)$$

$P_v(\cos \theta)$ is the Legendre polynomial of order $v$; $L_1, j_1$ and $j$ are the angular momentum of the $i^\text{th}$ quantum, $i^\text{th}$ quantum state, and the intermediate state of the cascade, respectively, $C(L_1 L_2 j; 1, -1)$ is the Clebsch-Gordan
coefficient with $v = L_1 + L_2$, and $W(jjL_1L_2; vj_1)$ is the Racah coefficient with $v_{\text{max}}$ the smallest value of $2j$, $2L_1$, $2L_2$. The coefficients $F_v$ have been tabulated by Biedenharn and Rose (6, p. 746).

For a cascade where one component is mixed $(j_1(L_1L'_1)j(L_2)j_2)$ there results (6, p. 747)

$$W(\emptyset)_{\text{mix}} = W_I(\emptyset) + \delta^2 W_{II}(\emptyset) + 2\delta W_{III}(\emptyset)$$

where $\delta$ is the mixing ratio and $\delta^2$ is the ratio of the total angle integrated intensity of the $L'_1$-pole to that of the $L$-pole radiation. The magnitude $\delta$ is the ratio of the reduced matrix elements:

$$\delta = (j \| T_{L'_1} \| j_1)/(j \| T_{L_1} \| j_1)$$

$W_I(\emptyset)$ and $W_{II}(\emptyset)$ are found from Equation (2). They are

$$W_I(\emptyset) = \sum_v F_v(L_1j_1j) F_v(L_2j_2j) P_v(\cos \emptyset)$$

and

$$W_{II}(\emptyset) = \sum_v F_v(L'_1j_1j) F_v(L_2j_2j) P_v(\cos \emptyset)$$

$W_{III}(\emptyset)$ is the contribution due to the interference between $L_1$ and $L'_1$ and is given by
\[
W_{III}(\varnothing) = \sum_{\substack{v \neq 0 \text{ even}}} a_v^{III} P_v(\cos \varnothing)
\]

with

\[
a_v^{III} = F_v(L_1L'_1j_1j) F_v(L_2j_2j)
\]

and

\[
F_v(L_1L'_1j_1j) = (-1)^{j-j_1-1} \frac{1}{((2j+1)(2L_1+1)(2L'_1+1))^{\frac{3}{2}}} C(L_1L'_1v; l, -1) \\
\times W(jjL_1L'_1; vj_1)
\]

The functions \(C(L_1L'_1v; l, -1)\) \(W(jjL_1L'_1; vj_1)\) have been tabulated for the case \(L' = L + 1\) (6, p. 748). Another parameter frequently used in angular correlation work is the anisotropy \(A\), defined by

\[
A = \frac{1 + a_2 + a_4}{1 - a_2 + 3a_4} - 1
\]
EXPERIMENTAL SETUP

The equipment used for the experiment mainly consisted of a gamma-ray spectrometer together with its table. The electronic equipment was similar to the conventional delayed fast-slow coincidence scheme in which the timing and energy selection functions were separated and finally re-combined in a triple coincidence circuit.

Gamma rays were detected by a pair of scintillation counters. These counters generated voltage pulses of negative polarity whose heights were proportional to the detected gamma ray energies. Each pulse was fed to a pre-amplifier which was essentially used for matching the impedance of the counter to that of the signal-carrying cable. The output of each of the two pre-amplifiers was fed to a linear amplifier. At this stage the signals were each split into separate timing and energy selection channels. The output of the pulse height discriminator of each of the linear amplifiers was fed to a fast coincidence circuit to establish time coincidence. The unselected amplified counter pulses (High output) were fed to the pulse height analyzers. The differential pulse height analyzers selected only these pulses with amplitudes between V and V + dV. The outputs of the analyzers were fed to the slow coincidence
channels. The output of the fast coincidence circuit was delayed by an extended delay line in order to match the delay experienced by the output pulses from the analyzers, before being fed to the triple coincidence stage. Hence the triple coincidence circuit gave an output only when the amplitudes of the signals from detectors 1 and 2 were within the window widths of the analyzers. Figure 3 shows a block diagram of the scintillation spectrometer.

The gamma-ray detectors were NaI (Tl) 1.5 inches in diameter by 1.0 inch thick optically coupled to Du Mont K-1719 photomultiplier tubes. The crystal-photomultiplier combination was co-axially positioned in a steel mounting which also acted as a partial electromagnetic shield. In addition, the movable detector had a mu metal shield to eliminate the effect of stray magnetic fields.

The pre-amplifiers were stacked cathode followers which gave a fast rising negative output pulse. The linear amplifiers were the R-C coupled non-overloading type designed for a rise time of 0.2 microsecond. The pulses used for the fast coincidence were the discriminated scaler outputs of the amplifiers and were of 25 volts magnitude, negative polarity and 0.7 microsecond duration. Each differential pulse-height analyzer was fed by the positive pulses from each of the
Figure 3. Block diagram of the scintillation spectrometer
amplifiers. Each pulse was of 1.3 microseconds duration with a long negative exponential undershoot.

The pulse height analyzers were of the usual type. Each generated a negative output pulse of about 15 volts if and only if a positive input signal had an amplitude falling within two preset voltage levels, V and V + dV. This determined first, whether the input pulse had crossed the lower level, V, of the channel, and second, whether or not the pulse had crossed the upper level, V + dV. The lower level crossing generated an output pulse which was vetoed by an anti-coincidence pulse when the upper level, V + dV, was crossed.

The coincidence circuit with a resolving time of about 8 x 10^{-8} seconds consisted of two fast and two slow channels. The fast channels were fed by the scaler pulse outputs of the respective amplifiers. If a coincidence occurred, a large pulse was formed at a common plate junction. This pulse, after shaping, was fed to the triple coincidence bus, having been delayed about five microseconds after the initial event to compensate for delay caused in the slow channels mainly by the pulse height analyzers. Outputs of the two analyzers were fed to the respective channels of the slow coincidence circuit. The outputs of the two fast and two slow channels, all of positive polarity, were fed to the
junction of four diodes and a load resistor. Each of these diodes was so arranged that any one of the diodes could hold the voltage down on the load resistor, even when one or more of the other diodes was cut off. This was attainable by opening the circuit through the diode by a manual switch. When a triple coincidence occurred, a positive pulse was generated on the triple coincidence bus which was fed to the output circuitry resulting in a negative output pulse to energize a scaler.

The high-voltage power supplies were designed to provide a highly stable (0.02% per day) d.c. voltage source for precision scintillation counting. The output voltage changed less than 0.35% for a current increase from zero to maximum load and less than 0.00035% per volt change of line from 100-130 volts. The ripple was less than 0.01% of the output voltage. Each power supply had an input regulating transformer followed by a high voltage transformer, a rectifier, and a R-C filter network. The filtered d.c. voltage was applied to a bank of 17 cold-cathode tubes from which the stabilized output voltage was obtained.

On the spectrometer tables, the distance of each detector from a central axis was adjustable. One detector was fixed, while the other could be rotated about the central axis, the angular position being known
to within 0.25 degree.

A cathetometer was used for placing the point source on the axial line of the two detectors.
SOURCE PREPARATION

It has been pointed out (18, p. 146) that in any angular correlation measurement the nature of the radioactive source is very important. The theory, as developed by most of the investigators, assumes a point source. Moreover the source must be uniform and of thickness less than a critical value so that the observed correlation function is not distorted appreciably, by the self-scattering of gamma rays from the walls of the container. In addition, in order to maximize the ratio of true coincidence rate to chance coincidence rate and still obtain enough total coincidence in a reasonable time, the counters should subtend large solid angles at the point source and the strength of the source should be appropriate.

Consider a source of strength $N_0$ disintegrations per second. Assuming that the counter efficiencies for the two gamma ray energies are the same, (this was roughly so in the case of Sc$^{46}$ because the two gamma rays have energies 0.89-Mev and 1.12-Mev) the total coincidence output counting rate, $N_{\text{true}}$, is given by

$$N_{\text{true}} = N_0 w_1 w_2 W(\phi) e_1 e_2$$

where $w_1$, $w_2$, $e_1$ and $e_2$ are the solid angles and the
efficiencies, respectively for counters 1 and 2. \( W(\phi) \) is the correlation function at angle \( \phi \).

The chance coincidence rate, \( N_{ch} \), is given by

\[
N_{ch} = 2TN_1N_2 = 2TN_0w_1w_2e_1e_2
\]

where \( T \) is the resolving time of the circuit (43, p. 202). The factor 2 in this relation arises because the coincidence circuit operates if a pulse from counter 2 arrives within the time interval \( T \) of a pulse from counter 1, irrespective of which pulse occurs first.

Thus

\[
\frac{N_{true}}{N_{ch}} = \frac{W(\phi)}{2TN_0}
\]

Hence for a given \( 2T \), limited by the electronics used, \( N_0 \) must be kept as small as possible to get the true-to-chance ratio as large as possible. However if the source is too weak, we may not be able to get a useable total coincidence count rate.

The source material used was obtained from Oak Ridge National Laboratory in the form of ScCl$_3$ in 1N HCl solution with purity greater than 99%. Most of the HCl was evaporated by heating uniformly to increase specific activity. The sources were prepared by dropping a very small amount of concentrated solution into a 0.031 inch diameter hole in a lucite source holder. The hole was
sealed off by a cemented lucite cap to minimize evaporation and spilling. The thickness of the wall around the source was less than 0.063 inches which is much smaller than the critical thickness of 0.125 inches as calculated by Aeppli et al. (1, p. 357) for a lucite container. The Co$^{60}$ sources which were used to calibrate the apparatus were commercially calibrated sources of strengths 0.1 and 1.1 millicuries respectively sealed in stainless steel cylinders.
EXPERIMENTAL MEASUREMENTS

The response of the spectrometer was checked by calibrating it with a Co$^{60}$ source since the decay scheme and the directional correlation function of Ni$^{60}$ are well known (43, p. 553). This procedure affords a stringent test of the overall technique and the equipment. Ni$^{60}$ while decaying to its ground state by the emission of a two gamma-ray cascade after beta decay from Co$^{60}$ has a short intermediate state mean life ($8 \times 10^{-13}$ seconds) and hence the correlation is not destroyed. The ground state of Ni$^{60}$, like other even-even nuclei, has zero spin and even parity so that the selection rule $|j_1 - j_2| \leq L \leq j_2 + j_1$ requires that the second transition $j(L_2)0$ must be pure. Consequently only the first gamma ray may involve mixed multipolarity, which reduces considerably the number of possible interpretations of a measured correlation.

The pulses coming out of each of the two detectors were fed to the respective linear amplifiers in the two channels. The gains of the two amplifiers were adjusted so that the upper gamma-ray photo peak in channel two produced pulses of the same height as those from the low gamma-ray photo peak in channel one. The amplifier and the analyzer discriminators of each channel were
set at point 'V' (Figure 4). The window widths of the analyzers were set so as to straddle the peaks. The counting rate was kept low enough (about $10^3$ per second) to keep the counting loss negligible because of the finite dead time of the electronic equipment.

The resolving time $T$ of the fast-slow coincidence circuit was determined by the two incoherent source method in an auxiliary experiment, but under exactly the same conditions as those which applied during the correlation run. That is, each counter was made to look at a source identical to the actual source (whose correlation was to be determined) in the matter of counting rate and energy spectrum. Under such conditions, $T$, the resolving time of the coincidence circuit was given by

$$2T = \frac{N'_{\text{ch}}}{N'_1 N'_2}$$

where $N'_1$, $N'_2$, and $N'_{\text{ch}}$ are the slow singles in each channel and chance coincidence rates of this auxiliary experiment, respectively. The resolving time determinations were made in such a way that most of the correlation readings were not more than two hours away from such a determination. The resolving time was found to be of the order of $8 \times 10^{-8}$ sec. and it did not vary appreciably during any given run. In order to get
Figure 4. Pulse height spectrum of Ti$_{46}^4$ for channels 1 and 2.
sufficient statistics most of the resolving time determination readings were taken for at least one hour.

In the case of the Co$^{60}$ test determinations, commercially calibrated sources of strengths 0.1 and 1.1 millicuries were used. The triple coincidence and slow singles rates were determined with the movable detector set alternately at 180° and 90°. About 15,000 true coincidence counts were accumulated at each of the two angles and the anisotropy, A, in the case of the Co$^{60}$ source was found to be 0.160 ± 0.027. This is in agreement with the published results (43, p. 553) even though some of the conditions for angular correlation work (such as having point source) were not very well fulfilled.

Sc$^{46}$ sources were prepared according to the procedure outlined in the section on source preparation. By trial, a source strength of about 40 microcuries was found to be optimum. This gave a true-to-chance coincidence ratio of about four. The method followed for taking data was the same, except for a few modifications, as for the calibration with the Co$^{60}$ sources. Because of the frequent de-mounting of one detector for the resolving time determinations, a cathometer was used to set its position reproducibly. The source detector distance was 7.0 cm throughout the experiment.
Readings were taken for a period of one-half hour each. The background effect even in the presence of some other sources in the room was found to be negligible because of the spectrometer's built-in energy selection. The triple coincidence and the slow singles rates were determined at ten angles between $90^\circ$ and $180^\circ$ in ten-degree increments. To correct for any asymmetry in the spectrometer table, coincidences were taken for angles of $90^\circ$ to $180^\circ$ and $180^\circ$ to $270^\circ$ between the axes of the detectors and the results of the measurements relative to symmetric angles with respect to $180^\circ$ were combined. The number of true coincidence counts accumulated at each angle was between 60,000 and 85,000 with the exception of $90^\circ$ (or $270^\circ$) where 637,500 were accumulated. The analysis of the data is outlined in the appendix and the results are given in Figure 5 and Table A-2.
Figure 5. Experimental correlation function in Ti$^{46}$ and corrected theoretical curve for 4(2)2(2)0
CONCLUSIONS

The results of gamma-gamma directional correlation measurements in Ti\textsuperscript{46} are summarized in Figure 5 and Table A-2. In conjunction with the theory, these results make it possible to strengthen the spin and parity assignments of the first two excited states of this nuclide. Ti\textsuperscript{46} like other even-even nuclei has even parity and zero spin in its ground state. This information and the selection rule \(|j_1 - j_2| \leq L \leq j_2 + j_1\) require that the 0.885-Mev gamma ray must be pure (unmixed multipolarity). As outlined above, various experiments are consistent with an assignment \(2^+\) for the first excited state. This, in turn, fixes the transition to the ground state to be pure quadrupole (i.e. \(L = 2\)). So the combined transition from the second excited state can be represented as \(j_1(L_1)2(2)^0\).

In order to try to assign a value for \(j_1\), and \(L_1\), all spin sequences commensurate with the selection rule \(|j_1 - j| \leq L_1 \leq j_1 + j\) were considered and they are listed in Table 1. The values of the coefficients, \(a_2\) and \(a_4\), listed in the table were calculated for each transition by using the tables of Biedenharn and Rose (6, p. 746).

From the study of the 0.36-Mev beta spectrum (21, p. 185) the log ft value for the decay is 6.2 which
Table 1. Possible Spin Sequences

<table>
<thead>
<tr>
<th>Cascade</th>
<th>$F_2(L_{1/2}l_1j_1)$</th>
<th>$F_4(L_{1/2}l_1j_1)$</th>
<th>$a_{2\text{theo}}^2$</th>
<th>$a_{4\text{theo}}^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$j_1(j_2)j_2$</td>
<td></td>
<td></td>
<td>$F_2(202)$</td>
<td>$F_4(202)$</td>
</tr>
<tr>
<td>$f_{0}(L_{1/2}l_1j_1)$</td>
<td></td>
<td></td>
<td>$F_2(202)$</td>
<td>$F_4(202)$</td>
</tr>
<tr>
<td>Experimental least sq.</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$3(1)2(2)0$</td>
<td>$+0.1195$</td>
<td>$+0.0714$</td>
<td>$0.1049^+$</td>
<td>$0.0057^+$</td>
</tr>
<tr>
<td>$3(2)2(2)0$</td>
<td>$+0.3415$</td>
<td>$+0.2041$</td>
<td>$0.0019^-$</td>
<td>$0.0030^-$</td>
</tr>
<tr>
<td>$3(3)2(2)0$</td>
<td>$+0.3287$</td>
<td>$+0.1964$</td>
<td>$0.0019^-$</td>
<td>$0.0030^-$</td>
</tr>
<tr>
<td>$3(4)2(2)0$</td>
<td>$+0.0726$</td>
<td>$-0.3436$</td>
<td>$0.3415^+$</td>
<td>$0.0019^-$</td>
</tr>
<tr>
<td>$3(5)2(2)0$</td>
<td>$0.3287$</td>
<td>$+0.0952$</td>
<td>$0.3287^+$</td>
<td>$0.0952^+$</td>
</tr>
<tr>
<td>$3(6)2(2)0$</td>
<td>$+0.0726$</td>
<td>$-0.3436$</td>
<td>$0.3415^+$</td>
<td>$0.0019^-$</td>
</tr>
<tr>
<td>$3(7)2(2)0$</td>
<td>$0.3287$</td>
<td>$+0.0952$</td>
<td>$0.3287^+$</td>
<td>$0.0952^+$</td>
</tr>
<tr>
<td>$4(2)2(2)0$</td>
<td>$-0.1707$</td>
<td>$-0.2041$</td>
<td>$-0.1707$</td>
<td>$-0.2041$</td>
</tr>
<tr>
<td>$4(3)2(2)0$</td>
<td>$+0.0297$</td>
<td>$-0.2678$</td>
<td>$0.0297$</td>
<td>$-0.2678$</td>
</tr>
<tr>
<td>$4(4)2(2)0$</td>
<td>$+0.2811$</td>
<td>$-0.3005$</td>
<td>$0.2811$</td>
<td>$-0.3005$</td>
</tr>
<tr>
<td>$4(5)2(2)0$</td>
<td>$0.4288$</td>
<td>$+0.0317$</td>
<td>$0.4288^+$</td>
<td>$0.0317^+$</td>
</tr>
<tr>
<td>$4(6)2(2)0$</td>
<td>$+0.2811$</td>
<td>$-0.3005$</td>
<td>$0.2811^+$</td>
<td>$0.3005^+$</td>
</tr>
<tr>
<td>$4(7)2(2)0$</td>
<td>$0.4288$</td>
<td>$+0.0317$</td>
<td>$0.4288^+$</td>
<td>$0.0317^+$</td>
</tr>
<tr>
<td>$5(3)2(2)0$</td>
<td>$-0.1707$</td>
<td>$0.1020$</td>
<td>$-0.1707$</td>
<td>$0.1020$</td>
</tr>
<tr>
<td>$5(4)2(2)0$</td>
<td>$+0.4482$</td>
<td>$-0.2678$</td>
<td>$0.4482^+$</td>
<td>$0.0317^+$</td>
</tr>
<tr>
<td>$5(5)2(2)0$</td>
<td>$+0.4288$</td>
<td>$-0.2563$</td>
<td>$0.4288^+$</td>
<td>$0.2563^+$</td>
</tr>
<tr>
<td>$5(6)2(2)0$</td>
<td>$0.4482$</td>
<td>$-0.0317$</td>
<td>$0.4482^+$</td>
<td>$-0.0317^+$</td>
</tr>
<tr>
<td>$5(7)2(2)0$</td>
<td>$+0.4288$</td>
<td>$-0.2563$</td>
<td>$0.4288^+$</td>
<td>$-0.2563^+$</td>
</tr>
<tr>
<td>$F_2(202) = -0.5976; F_4(202) = -1.069; F_0(L_{1/2}l_1j_1) = 1; a_0 = 1.$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

is consistent with either a first forbidden transition or an allowed transition. However, since both the shell model and the evidence from the internal conversion coefficient as pointed out above require that the second excited state must have even parity, we can accept it as established that the 0.36-Mev beta decay is allowed. This transition should involve $\Delta j = 0, \pm 1, \text{(no)}$
according to the Gamow-Teller selection rule. This result and the assignment of the ground state of $^{46}$Sc as $4^+$ limit the possible spin values of the 2.004-Mev state of $^{46}$Ti to 3, 4, and 5. So the different spin sequences listed in Table 1 are the result of $3, 4, 5 \ (L_\perp)2(2)0$ and the selection rule $|j_1 - j_2| \leq L \leq j_1 + j_2$. The values of the coefficients $a_2$ and $a_4$ found experimentally check quite well with the theoretical values for the $4(2)2(2)0$ transition. The coefficients for the transitions involving $L_\perp = 3$ or higher values were not calculated as the transition probability is almost zero for such transitions in competition with the transitions involving lower multipole order. Hence if the first gamma radiation following the beta decay is pure, it must be a quadrupole and the cascade is $4(2)2(2)0$. The experimental points are compared with theoretical directional correlation function for a $4(2)2(2)0$ type of cascade modified for the solid angle correction (appendix) because of the finite size of the detectors (Figure 5).

However the possibility still exists that suitable mixing of radiations in the first gamma radiation would produce a mixed directional correlation function with coefficients that would agree with the experimental least square values. Biedenharn and Rose (6, p. 747)
have outlined a method for calculating the mixture of two transitions \( j_1(L_1)j(L_2)j_2 \) and \( j_1(L'_1)j(L_2)j \) where \( L'_1 = L_1 + 1 \). For cascades in which such a mixture exists, the correlation, \( W(\theta)_\text{mix} \), can be expressed

\[
W(\theta)_\text{mix} = W_1(\theta) + S^2W_{II}(\theta) + 2S_{III}(\theta)
\]

\[
= \sum_v a_v P_v(\cos\theta)
\]

where \( W_1 \) and \( W_{II} \) are the directional correlation functions for the pure \( 2L_1 \)-pole and pure \( 2L_1 + 1 \)-pole radiation in the otherwise mixed transition; \( W_{III} \) is an interference function and \( S^2 \) is the ratio of the intensities of the \( 2L_1 + 1 \)-pole to that of the \( 2L_1 \)-pole. The coefficients, \( a_2 \) and \( a_4 \), and anisotropy, \( A \), are quadratic functions of \( S \). \( a_2 \) being the best known quantity from the least squares analysis, \( W(\theta)_\text{mix} \) was calculated for various mixed transitions e.g., \( 4(2)4(2)0 \) and \( 4(3)2(2)0 \); \( 3(1)2(2)0 \) and \( 3(2)2(2)0 \); and \( 3(2)2(2)0 \) and \( 3(3)2(2)0 \). The mixed \( a_2 \) coefficients were equated to the solid-angle-corrected least squares \( a_2 \). When solutions existed, two values of \( S \) were obtained which were used to calculate the corresponding mixed \( a_4 \) coefficients for comparison with the solid angle corrected least squares \( a_4 \). Only three possible cases were considered as the other had vanishingly small transition probabilities. Table 2 lists the results of this
Table 2. Mixed Transition Correlation Coefficients for $a_{2 1 s}^2 = 0.1049 \pm 0.0020$.

<table>
<thead>
<tr>
<th>Mixed Cascades</th>
<th>$\delta$</th>
<th>$a_{4 \text{mix}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>3(1)2(2)0 and 3(2)2(2)0</td>
<td>-0.2500</td>
<td>-0.0048</td>
</tr>
<tr>
<td>3(2)2(2)0 and 3(3)2(2)0</td>
<td>imaginary</td>
<td>a$_{4 1 s}$ = 0.0057 $^\dagger$</td>
</tr>
<tr>
<td>4(2)2(2)0 and 4(3)2(2)0</td>
<td>-0.00483</td>
<td>+0.00084</td>
</tr>
<tr>
<td>4(3)2(2)0</td>
<td>-1.6158</td>
<td>-0.0341</td>
</tr>
</tbody>
</table>

The choice was finally limited to only two possibilities which are 4(2)2(2)0 and 4(3)2(2)0; 3(1)2(2)0 and 3(2)2(2)0; all other cases yielded $a_4$'s which were clearly inconsistent with the experimental results. Figure 6 shows the variation of the correlation function for the pure transition for these four cases. For 4(2)2(2)0 and 4(3)2(2)0 mixing the value $\delta = -0.00483$ gives a value for $a_4$ consistent with the experimental results. To conserve parity the mixing in this case must be between E2 and M3. From this value of $\delta$ we get 99.995% E2 mixed with 0.005% M3. The value of $\delta = -0.2500$ obtained for 3(1)2(2)0 and 3(2)2(2)0 mixing gives a predicted $a_{4 \text{mix}}$ which has a sign different from the $a_{4 1 s}$.
Figure 6. Corrected theoretical curves of correlation function for some possible transitions.
However, $a_{4^{1.5}}$ is not sufficiently well known to be able to call this sign difference a clear inconsistency. Using this value of $\delta$ to calculate the mixing yields dipole radiation to be 94.12% mixed with 5.88% of quadrupole radiation. To conserve parity this should be 94.12%M1 mixed with 5.88%E2.

These results indicate that there is possibly a choice to be made between the mixture of E2 (99.995%) and M3 (0.005%) and the mixture of M1 (94.12%) and E2 (5.88%). The dependence of transition probability for gamma ray emission on multipolarity and the electromagnetic nature of the transition does not seem to rule out the possibility for the mixture of M1 and E2. Both of these lead to a prediction of a mean life for the second excited state of $\sim 10^{-11}$ sec. which is not in disagreement with the measured value which is less than $3 \times 10^{-11}$ sec. (3).

A consideration based on the notion of parity favored and parity unfavored transitions does not rule out this possibility either. However the experimentally determined values of internal conversion coefficients agree with the theoretically calculated values only if we assume that the transition is predominantly E2. In the case of a predominantly M1 transition the theoretically calculated values (38) are almost double the experimentally determined values. Hence the transition under discussion is most likely an almost pure E2
transition mixed with at most $0.005 \pm 0.002\% M_3$.

It may be pointed out that the mixing ratio can also be determined from a comparison of the experimental value of the anisotropy with the anisotropy calculated from $a_2^{\text{mix}}$ and $a_4^{\text{mix}}$ which is given by

$$A_{\text{exp}} = A_{\text{mix}} = \frac{1 + a_2 + a_4}{1 - \frac{a_2}{2} + \frac{3a_4}{8}} - 1 = f(\delta)$$

For the $4(2)2(2)0-4(3)2(2)0$ mixture, the quadratic equation in $\delta$ gave two values, $-1.5237$ and $-0.0047$, which gave the values of $a_4^{\text{mix}}$ to be $0.0633$ and $0.0085$ respectively. The mixing ratio calculated this way is completely consistent with that calculated from the least squares fit. An almost pure E2 transition for the 1.12-Mev gamma ray requires an assignment of $j_1 = 4$ and even parity for the 2.004-Mev state of Ti$^{46}$. This assignment of $4^+$ to this state is in agreement with the prediction of the shell model of nuclei (43, p. 423) and of the unified model of nuclei (43, p. 473). The unified model predicts that, for an even-even nucleus in which the ground state has $K = \Lambda = 0$, the lowest rotational levels have spin values $j_1 = 0, 2, 4....$ and even parity. The directional correlation function is extremely sensitive to $\delta$ and hence can be used very fruitfully to determine the mixing ratio of mixed multipolarity gamma-ray transitions in those cases where it is applicable to experimentally obtained results.
BIBLIOGRAPHY


The dependence of the triple coincidence counting rate on the angular position of the movable detector corresponds to the theoretical correlation function only under the assumption of point source and point detectors with no scattering and other disturbing factors such as the earth's magnetic field. Since the effect to be sought is small, it is imperative to perform the experiment with great care, and corrections should be made for any deviations from ideal conditions. As discussed in the section on source preparation, the sources were made to be essentially point sources, and the thickness of the walls of the lucite source holder was chosen to be much less than the critical thickness so as to minimize scattering. The geometrical corrections applied because of the solid angle subtended by the detectors at the point source were used from the tables by Stanford and Rivers (45). These theoretical calculations were checked experimentally, and they applied quite well for the scintillation spectrometer used for this experiment. A mu metal shield was put on the photomultiplier of the movable detector to reduce the effect of extraneous fields such as earth's magnetic field.
The analysis of the data was carried out according to the method suggested by Rose (40). The number of true coincidence counts \( n_{\text{true}}(\theta) \) of the \( i \# \) run at the angle \( \theta \) was determined by subtracting the chance coincidence counts \( 2Tn_{1i}(\theta)n_{2i}(\theta) \) from the triple coincidence counts \( n_{\text{triple}}(\theta) \), where \( n_{1i}(\theta) \) and \( n_{2i}(\theta) \) are the slow singles of channels one and two, respectively and \( T \) is the resolving time of the coincidence circuit. The number of true coincidence counts \( n_{\text{true}}(\theta) = \) time \( x \) rate of true coincidences) was divided by the product of singles in channels one and two in order to eliminate, to a first approximation, the effects of the slow electronic drift of the apparatus with consequent variation of the efficiencies of the detectors. This gave the true coincidence ratio

\[
N_i(\theta) = \frac{n_{\text{true}}(\theta)}{n_{1i}(\theta)n_{2i}(\theta)} = \frac{n_{\text{triple}}(\theta) - 2Tn_{1i}(\theta)n_{2i}(\theta)}{n_{1i}(\theta)n_{2i}(\theta)}
\]

The standard deviation connected in a function \( F(x_i) \) due to uncertainties in the variables, \( x_i \), is given by

\[
S_{F(x_i)}^2 = \sum_i \left( \frac{\partial F}{\partial x_i} \right)^2 S_{x_i}^2
\]

Consequently standard deviation \( s_{n_i}(\theta) \) associated with \( n_i(\theta) \) is
\[ s^2_i = \frac{1}{N_i(90^\circ)} \frac{n_{\text{triple}}(\theta)}{n_{11}(\theta)n_{21}(\theta)} \left[ \frac{(n_{11}(\theta)n_{21}(\theta))}{(n_{11}'n_{21}')^2} \right]^2 \left( \frac{t'}{t} \right)^2 \times n_{\text{ich}}^2 \]

where \( n_{11}', n_{21}' \) and \( n_{\text{ich}}' \) are the singles in channels one and two and \( n_{\text{ich}}' \) the chance coincidence counts, respectively, in time \( t' \) in the auxiliary two-independent-source experiment to determine the resolving time of the circuit.

The data taken at any particular angle \( \theta \) was divided into subgroups so that the readings were steady in any particular subgroup. The average coincidence ratio \( \overline{N_i}(90^\circ) \) was found for each subgroup. The true coincidence ratio \( N_i(\theta) \) at angle \( \theta \) in each subgroup was divided by the average coincidence ratio \( \overline{N_i}(90^\circ) \) at \( 90^\circ \) for that particular subgroup to give a normalized ratio \( N_i(\theta)/\overline{N_i}(90^\circ) \). The standard deviation \( s_i \) associated with this normal ratio can be written as

\[ s^2_i = \frac{1}{2} \frac{s^2_{N_i(\theta)}}{N_i(90^\circ)} + \frac{N_i(\theta)}{N_i(90^\circ)} \frac{s^2_{N_i(90^\circ)}}{2} \left( \frac{t'}{t} \right)^2 \times n_{\text{ich}}^2 \]

The average weighted normalized ratio may be written...
\[
\frac{N_i(\theta)}{N_i(90^\circ)} = \frac{\sum_i w_i N_i(\theta)/N_i(90^\circ)}{\sum_i w_i}
\]

where the weight \( w_i \) associated with the \( i \)th run is the ratio \( b/S_i^2 \), \( b \) is an arbitrarily chosen constant to assign weights and \( S_i \) is the standard deviation associated with the normalized ratio \( N_i(\theta)/N_i(90^\circ) \). The standard deviation \( s(\theta) \) of the average weighted normalized ratio is given by

\[
s^2(\theta) = \frac{\sum_i w_i d_i^2}{(m-1) \sum_i w_i}
\]

where \( m \) is the number of pieces of data and \( d_i \) the difference of the \( i \)th normalized ratio \( N_i(\theta)/N_i(90^\circ) \) from the average weighted normalized ratio. The new standard deviation \( s(\theta) \) of the weighted normalized ratio was calculated by bringing all the normalized ratios \( N(\theta)/N(90^\circ) \), which had values of \( 'd' \) greater than \( 3 \times s(\theta) \), to such a value of \( N(\theta)/N_i(90^\circ) \) that \( 'd' \) was equal to \( 3 \times s(\theta) \). This did not change the value of the average weighted normalized ratio appreciably but \( s(\theta) \) gave a letter determination of the standard deviation associated with \( N_i(\theta)/N_i(90^\circ) \). The procedure carried out so far gave \( N_i(\theta)/N_i(90^\circ) + s(\theta) \). The same procedure was followed at each angle to get a set of ten such ratios which were used for least squares analysis.
of the data. These ratios are listed in Table 1-A. Certain weight was associated with each of these ratios for least square analysis and it was given by $b'_1 \frac{1}{S^2}$

where $b'_1$ is another arbitrarily chosen constant and $S$ is the standard deviation associated with $\frac{N_i(\theta)}{N_i(90^\circ)}$.

Table 1-A. Average Weighted normalized ratios.

<table>
<thead>
<tr>
<th>Angle ((\theta))</th>
<th>$\frac{N_i(\theta)}{N_i(90^\circ)} \pm \frac{S(\theta)}{N_i(90^\circ)}$</th>
<th>Normalized $\frac{N_i(\theta)}{N_i(90^\circ)}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>90°</td>
<td>0.9980 $\pm$ 0.0024</td>
<td>1.0000 $\pm$ 0.0024</td>
</tr>
<tr>
<td>100°</td>
<td>0.9999 $\pm$ 0.0022</td>
<td>1.0019 $\pm$ 0.0022</td>
</tr>
<tr>
<td>110°</td>
<td>1.0102 $\pm$ 0.0028</td>
<td>1.0102 $\pm$ 0.0028</td>
</tr>
<tr>
<td>120°</td>
<td>1.0361 $\pm$ 0.0059</td>
<td>1.0361 $\pm$ 0.0059</td>
</tr>
<tr>
<td>130°</td>
<td>1.0580 $\pm$ 0.0024</td>
<td>1.0580 $\pm$ 0.0024</td>
</tr>
<tr>
<td>140°</td>
<td>1.0850 $\pm$ 0.0023</td>
<td>1.0850 $\pm$ 0.0023</td>
</tr>
<tr>
<td>150°</td>
<td>1.1056 $\pm$ 0.0029</td>
<td>1.1056 $\pm$ 0.0029</td>
</tr>
<tr>
<td>160°</td>
<td>1.1266 $\pm$ 0.0028</td>
<td>1.1266 $\pm$ 0.0028</td>
</tr>
<tr>
<td>170°</td>
<td>1.1429 $\pm$ 0.0039</td>
<td>1.1429 $\pm$ 0.0039</td>
</tr>
<tr>
<td>180°</td>
<td>1.1544 $\pm$ 0.0029</td>
<td>1.1544 $\pm$ 0.0029</td>
</tr>
</tbody>
</table>

Least Squares Analysis of the Data

The determination of the coefficients describing $W(\theta)_{\text{exp}}$ was carried out using a least-squares analysis. A series of Legendre polynomials were used to represent $\frac{N_i(\theta)}{N_i(90^\circ)}$ as closely as possible (30, p. 613):
\[ w(\theta)_{\text{exp}} = \frac{N_i(\theta)}{N_i(90)} = \sum_j a_j p_j(\cos \theta) = \sum_j a_j A_{ij} \]

The coefficients \( a_j \), which were the most probable values for the given data, were determined by minimizing the following function:

\[
\sum_i w_i \left( \frac{N_i(\theta)}{N_i(90)} - \sum_j a_j A_{ij} \right)^2
\]

This yielded the normal equations

\[
\sum_i w_i \left( \frac{N_i(\theta)}{N_i(90)} - \sum_j a_j A_{ij} \right) A_{ik} = 0
\]

One now defines a symmetric matrix

\[ C = \tilde{A} W A \]

with elements

\[ C_{jk} = \sum_i w_i A_{ij} A_{ik} = C_{kj} \]

where \( W \) is a diagonal matrix with elements \( w_i \), \( A \) is a matrix of Legendre polynomials and \( A \) is its transposed matrix. Now let

\[ B = \tilde{A} W N \]

where \( N \) is the vector with components \( \frac{N_i(\theta)}{N_i(90)} \). Then it follows that \( C a = B \), and by matrix multiplication

\[ a = C^{-1} B \], or in detail

\[ a_j = \sum_k C_{jk}^{-1} B_k \]
Since the coefficients are a function of $\frac{N_1(\theta)/N_1(90)}{\theta}$, any variation in this quantity will cause a corresponding variation in the coefficients. From the form of $a_j$ one observes that

$$a_j = \sum_{ik} c_{kj}^{-1}A_{ik}w_i \frac{N_1(\theta)}{N_1(90)}$$

and

$$s_{aj} = \sum_{ik} c_{kj}^{-1}A_{ik}w_i \overline{s_i}$$

where $\overline{s_i}$ is the error in $\frac{N_1(\theta)/N_1(90)}{\theta}$. Likewise,

$$s^2_{aj} = \sum_{ikl} c_{kj}^{-1}c_{lj}^{-1}A_{ik}A_{il}w_i \overline{s_i}^2$$

$$= \sum_{ikl} c_{kj}^{-1}c_{lj}^{-1}A_{ik}A_{il}w_i \frac{b'_1}{i^2} \overline{s_i}^2$$

$$= b'_1 \sum_{kl} c_{kj}^{-1}c_{lj}^{-1} \sum_i w_i A_{ik}A_{il}$$

$$= b'_1 \sum_{kl} c_{kj}^{-1}c_{lj}^{-1} c_{lk}$$

Now set $k = j$ and sum over $l$ and $k$ to obtain

$$s^2_{aj} = b'_1 c_{jj}^{-1}$$
The standard error in the coefficients was given by the diagonal elements of $C^{-1}$ multiplied by $b_{1,1}$.

**Solid Angle Correction**

In order to compare the experimentally determined values of the coefficients $a_2$ and $a_4$ one has to account for the correction because of the finite size of the detectors. The analysis of such a correction was carried out by Rose (40). The theoretical values of $a_2$ and $a_4$ calculated on the basis of point source and point detectors can be modified to give the modified theoretical correlation function $W(\theta)$ as

$$ W(\theta) = \sum_{\nu} a_{\nu} P_{\nu}(\cos \theta) \frac{J_{\nu}(1)}{J_{\nu}(1)} \times \frac{J_{\nu}(2)}{J_{\nu}(2)} $$

The ratios $\frac{J_{\nu}(1)}{J_{\nu}(1)}$ where $\nu = 0, 2, 4$, have been calculated (45). Table A-2 shows the calculations of the $a_j$, their errors, $W(\theta)_{\text{exp.}}$ and $A_{\text{exp.}}$. 
Table A-2. Calculations Sheet for the Least-Squares Analysis of the Experimental Coefficients of Ti$_{46}$.

<table>
<thead>
<tr>
<th>$\theta$</th>
<th>$\frac{N_1(\theta)}{N_1(90^\circ)}$</th>
<th>$W_1(1 \cdot sq)$</th>
<th>$b'_1 = 5.76 \times 10^{-6}$</th>
<th>$A_{12}$</th>
<th>$A_{14}$</th>
<th>$A_{12}^2$</th>
<th>$A_{14}^2$</th>
<th>$A_{12}A_{14}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>90°</td>
<td>1.0000</td>
<td>1.0000</td>
<td>-0.5000</td>
<td>+0.3750</td>
<td>+0.2500</td>
<td>+0.14063</td>
<td>-0.18750</td>
<td></td>
</tr>
<tr>
<td>100°</td>
<td>1.0019</td>
<td>1.1901</td>
<td>-0.4548</td>
<td>+0.2659</td>
<td>+0.2068</td>
<td>+0.07070</td>
<td>-0.12093</td>
<td></td>
</tr>
<tr>
<td>110°</td>
<td>1.0122</td>
<td>0.7347</td>
<td>-0.3245</td>
<td>-0.0038</td>
<td>+0.1053</td>
<td>+0.000014</td>
<td>+0.00123</td>
<td></td>
</tr>
<tr>
<td>120°</td>
<td>1.0382</td>
<td>0.1655</td>
<td>-0.1250</td>
<td>-0.2891</td>
<td>+0.0156</td>
<td>+0.08358</td>
<td>+0.03614</td>
<td></td>
</tr>
<tr>
<td>130°</td>
<td>1.0601</td>
<td>1.1000</td>
<td>+0.1198</td>
<td>-0.4275</td>
<td>+0.0144</td>
<td>+0.18276</td>
<td>-0.05121</td>
<td></td>
</tr>
<tr>
<td>140°</td>
<td>1.0872</td>
<td>1.0888</td>
<td>+0.3802</td>
<td>-0.3190</td>
<td>+0.1446</td>
<td>+0.10176</td>
<td>-0.12128</td>
<td></td>
</tr>
<tr>
<td>150°</td>
<td>1.1078</td>
<td>0.6849</td>
<td>+0.6250</td>
<td>+0.0234</td>
<td>+0.3906</td>
<td>+0.00055</td>
<td>+0.01463</td>
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</tr>
<tr>
<td>160°</td>
<td>1.1289</td>
<td>0.7247</td>
<td>+0.8245</td>
<td>+0.4750</td>
<td>+0.6798</td>
<td>+0.22563</td>
<td>+0.39164</td>
<td></td>
</tr>
<tr>
<td>170°</td>
<td>1.1452</td>
<td>0.3787</td>
<td>+0.9548</td>
<td>+0.8532</td>
<td>+0.9116</td>
<td>+0.72795</td>
<td>+0.81464</td>
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</tr>
<tr>
<td>180°</td>
<td>1.1567</td>
<td>0.7347</td>
<td>+1.0000</td>
<td>+1.0000</td>
<td>+1.0000</td>
<td>+1.00000</td>
<td>+1.00000</td>
<td></td>
</tr>
<tr>
<td>$W_{1A_{12}}$</td>
<td>$W_{1A_{14}}$</td>
<td>$W_{1A_{12}^2}$</td>
<td>$W_{1A_{14}^2}$</td>
<td>$W_{1A_{12}A_{14}}$</td>
<td>$\frac{N_1(\theta)}{N_1(90^o)}$</td>
<td>$W_{1A_{12}}$</td>
<td>$\frac{N_1(\theta)}{N_1(90^o)}$</td>
<td>$W_{1A_{14}}$</td>
</tr>
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<td>----------------</td>
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<td>----------------</td>
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<td>----------------</td>
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<td>-0.5000</td>
<td>+0.3750</td>
<td>+0.2500</td>
<td>+0.14063</td>
<td>-0.18750</td>
<td>1.00000</td>
<td>-0.50000</td>
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<td>+0.2461</td>
<td>+0.08414</td>
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<td>1.19236</td>
<td>-0.54229</td>
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<td>-0.0028</td>
<td>+0.0774</td>
<td>+0.00091</td>
<td>+0.00090</td>
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<td>-0.21432</td>
<td>-0.00283</td>
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<tr>
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<td>-0.0478</td>
<td>+0.0006</td>
<td>+0.01383</td>
<td>+0.00598</td>
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<td>-0.02148</td>
<td>-0.04967</td>
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<tr>
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<td>+0.0144</td>
<td>+0.18276</td>
<td>-0.05121</td>
<td>1.06010</td>
<td>+0.12700</td>
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</tr>
<tr>
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<td>+0.1574</td>
<td>+0.11080</td>
<td>-0.13205</td>
<td>1.18374</td>
<td>+0.45005</td>
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<td>+0.00038</td>
<td>+0.01002</td>
<td>0.75873</td>
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<td>+0.7347</td>
<td>0.84983</td>
<td>+0.84983</td>
<td>+0.84983</td>
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</tr>
</tbody>
</table>
Table A-2 continued

\[
\sum_{i} w_{i} = + 7.7121 \quad \sum_{i} w_{i} A_{12} A_{14} = + 0.8332
\]

\[
\sum_{i} w_{i} A_{12} = + 1.3636 \quad \sum_{i} w_{i} \frac{N_{1}(\theta)}{N_{1}(90^\circ)} = + 8.22633
\]

\[
\sum_{i} w_{i} A_{12}^{2} = + 2.5947 \quad \sum_{i} w_{i} A_{12} \frac{N_{1}(\theta)}{N_{1}(90^\circ)} = + 1.6980
\]

\[
\sum_{i} w_{i} A_{14} = + 1.2888 \quad \sum_{i} w_{i} A_{14} \frac{N_{1}(\theta)}{N_{1}(90^\circ)} = + 1.44288
\]

\[
\sum_{i} w_{i} A_{14}^{2} = + 1.7087
\]
Table A-2 continued

\[
| c | =
| \sum_{i} w_i | \sum_{i} w_i A_{i12} | \sum_{i} w_i A_{i14} |
| \sum_{i} w_i A_{i12} | \sum_{i} w_i^2 A_{i12} | \sum_{i} w_i A_{i12} A_{i14} |
| \sum_{i} w_i A_{i14} | \sum_{i} w_i A_{i12} A_{i14} | \sum_{i} w_i A_{i14} |
\]

= 24.280

\[
\begin{bmatrix}
a_0 \\
a_2 \\
a_k
\end{bmatrix}
= 
\begin{bmatrix}
+15.4012 \times 10^{-2} & -5.1738 \times 10^{-2} & -9.0931 \times 10^{-2} \\
-5.1738 \times 10^{-2} & +47.4329 \times 10^{-2} & -19.2269 \times 10^{-2} \\
-9.0931 \times 10^{-2} & -19.2269 \times 10^{-2} & +74.7548 \times 10^{-2}
\end{bmatrix}
\begin{bmatrix}
h_{a_0} \\
h_{a_2} \\
h_{a_k}
\end{bmatrix}
\]

\[
a_v' = \text{normalized with } a_0 = 1.0000
\]

\[
a_0 = +1.0480 \\
a_2 = +0.1018 \\
a_k = +0.0044
\]

\[
a_0 = +1.0000 \\
a_2 = +0.0971 \\
a_k = +0.0042
\]
Table A-2 continued

\[ s^2 a_o = b'_1 c^{-1} j_j = (5.76 \times 10^{-6}) (15.4012 \times 12^{-2}) \]
\[ = 88.71 \times 10^{-8} \]
\[ s^2 a_o = 0.00094 \]

\[ s^2 a_2 = (5.76 \times 47.4329 \times 10^{-8})^{1/2} = 0.00165 \]

\[ s^2 a_4 = (5.76 \times 74.7548 \times 10^{-8})^{1/2} = 0.0021 \]

Errors in the \( a_v \) normalized and compounded

\[ s^2 a_o = 0.00099; \quad s^2 a_2 = 0.0020; \quad s^2 a_3 = 0.0030 \]

Solid angle correction for \( a_v \)

\[ a'_2 = \frac{J_0(1) \cdot J_0(2)}{J_2(1) \cdot J_2(2)} a_2 = \frac{0.0971}{0.9622 \times 0.9618} = 0.049 \]

\[ a'_4 = \frac{J_0(1) \cdot J_0(2)}{J_4(1) \cdot J_4(2)} a_4 = \frac{0.0042}{0.8588 \times 0.8586} = 0.0057 \]

\[ W(\theta)_{\text{exp}} = 1 + (0.1049 + 0.002)P_2(\cos \theta) + (0.0057 + 0.003)P_4(\cos \theta) \]

\[ A_{\text{exp}} = \frac{W(180^\circ) - W(90^\circ)}{W(90^\circ)} = 0.1710 \pm 0.0038 \]