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Directional Correlation Tests for
Violation of Time-Reversal Invariance in
Gamma Rays of $^{180}\text{Hf}^m$, ^{169}Tm , ^{175}Lu , and ^{184}W

by

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ABSTRACT

Directional Correlation Tests for Violation of Time-

Reversal Invariance in γ -Rays of $^{180}\text{Hf}^m$,
 ^{169}Tm , ^{175}Lu , and ^{184}W

by

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Experiments were performed to search for time-odd asymmetries in γ - γ directional correlations resulting from cascades $I_1 \xrightarrow{\gamma_1} I_2 \xrightarrow{\gamma_2} I_3$ in several radioactive sources. Searches were made for parity-even, time-odd terms of the form $(\vec{I}_1 \cdot \vec{k}_1 \times \vec{k}_2) (\vec{k}_1 \cdot \vec{k}_2)$, where $\vec{k}_1$ and $\vec{k}_2$ are the momentum vectors of the two γ -rays, for the "regular" multipole components of the following highly hindered transitions: 501-keV ($M2, E2, E3; ^{180}\text{Hf}^m$), 198- and 177-keV ($M1, E2; ^{169}\text{Tm}$), 283-keV ($E1, M2; ^{175}\text{Lu}$), and 922-keV ($E1, M2, E3; ^{184}\text{W}$), each of which served as the initial (γ_1) radiation in a cascade. A search was also conducted for a parity-odd, time-odd contribution of the form $(\vec{I}_1 \cdot \vec{k}_1 \times \vec{k}_2) (\vec{I}_1 \cdot \vec{k}_2)$ in the "irregular" (parity-violating) $E2$ component of the $^{180}\text{Hf}^m$ 501-keV γ -ray. Nuclear spin polarization was produced cryogenically at $T = 16\text{--}35$ mK, with

the sources introduced as impurities in magnetic hosts and external polarizing fields applied. Asymmetries were determined from comparison of coincidence rates with different spin directions, the γ -ray detectors remaining fixed. Limits on relative phase angles between the various multipole components of electromagnetic transition matrices were determined. Also, the time-odd components of the various transition matrices were evaluated as fractions of Weisskopf single-particle estimates, following arguments about the effects of nuclear-structure-related hindrances on time-odd components of nucleon-nucleon interactions which have previously been found valid in measurements of parity violation. These computations show components from 10^{-3} to 10^{-8} Weisskopf units, several orders more sensitive upper limits in several cases than have been established in previous TRIV experiments with γ -rays. In secondary experiments, the approach to microscopic magnetic saturation of the impurities Mn, Co, Ta, and Ir in polycrystalline iron at 20 mK were investigated by means of γ -ray angular distribution measurements, and it was found that applied external fields of 3 - 5 kOe were required to assure alignment of nuclear spins with applied magnetic field direction. Also, in another experiment, upper limits on the hyperfine nuclear magnetic fields at Zn, Zr, and Nb sites in the itinerant ferromagnet ZrZn_2 were determined by nuclear orientation.

hindrance factors in multipole components of nuclear gamma rays, as suggested by Steyert and Krane (Stey 73). Under assumptions about interactions outlined in Chapter V, more sensitive tests than previously obtained, by several orders of magnitude in some cases, have been achieved, casting strong doubt upon the electromagnetic or strong interaction as a source. In addition, all previous TRIV tests on γ -rays (as cited in Chapter V) have been performed on those with M1-E2 multipolarity. In the present experiments it has been possible to test γ -rays with M2-E2-E3, E1-M2, and E1-M2-E3 mixed multipolarities. As a final contribution to the literature, the difference technique wherein the nuclear spin is inverted (for the P-even, T-odd tests), or rotated through 90 degrees (for the P-odd, T-odd test in ^{180}Hf) appears to be unique.

Additional original experimental results (of secondary importance) obtained during the course of this program were (1) a determination of the degree of saturation of nuclear polarization as a function of externally applied field and (2) measurements of nuclear hyperfine fields in the interesting magnetic material ZrZn_2 . These two experiments are described in Chapter III, along with a general discussion of hyperfine magnetic fields and nuclear orientation.

In Chapter IV is contained a summary of the crucial aspects of the Krane, Steffen and Wheeler formalism for directional correlation of radiation from polarized nuclear levels, with a term-by-term description of the factors needed to understand the results of the present experiments.

The experiments performed on the isotopes $^{180}\text{Hf}^m$ (TRIV and simultaneous P- and T-violation) and on ^{169}Tm , ^{175}Lu , and ^{184}W (TRIV) are

described in detail in Chapter V. Also included are computations of multipole component relative phase angles and perturbation computations of hypothetical level mixing amplitudes.

In the Appendices will be found descriptions of $^3\text{He} - ^4\text{He}$ dilution refrigerator operating principles, a survey of ultralow temperature thermometry techniques, and a discussion of the minicomputer-based data acquisition system utilized in this experiment.

At the time of submission of this dissertation, the key results of all the experiments described above have been published or submitted for publication in the literature. (Explicit references are given in Chapters III and V.)

CHAPTER II

TIME-REVERSAL-INVARIANCE IN γ -DECAY

Fundamental Symmetries-Present Status

The discovery in 1957 of violation of parity conservation in the β -decay of ^{60}Co (Wu 57) and the apparent CP-violating asymmetry later found in the decay $K^0 \rightarrow 2\pi$ (Chr 64) have motivated both theorists and experimentalists to explore and question the validity of previously firmly held beliefs in the invariance of physical interactions with respect to charge inversion (C), parity inversion (P), and time reversal (T), and indeed to question the universal validity of the CPT-theorem itself; i.e. that the product of all three transformations does not affect a physical interaction.

Subsequent experiments have shown parity violation evidenced in nuclear radiation in isotopes such as ^{181}Ta , ^{175}Lu , ^{114}Cd , ^{41}K and ^{16}O (see for example the reviews of Fis 73, Gar 73). A strong (1.6%) parity violating contribution was measured in the 501-keV γ -radiation from ^{180}Hf by circular polarization measurements (Jen 70) and by Krane and Steyert in this laboratory by measurements of 0° - 180° asymmetries in the angular distribution of γ -radiation from a polarized nuclear level (Kra 71a). Also evidence for parity violation has been found by the detection of non-zero circular polarization of the emitted photon when thermal neutrons are captured by neutrons in the reaction $n+p \rightarrow d + \gamma$ (Lob 72).

On the other hand no further definite evidence has been seen for CP- or T-violation, although many experiments have been performed (as cited in Chapter V).

Time Reversal Invariance (TRI)-Significance

In the most elementary sense, TRI signifies that a physical equation of motion must remain invariant whenever time t is replaced by $-t$. In classical mechanics, clearly Newton's equations (i.e. $d^2p/dt^2 = F$) satisfy TRI since t appears in second order. One can easily visualize reversible two-body collisions which are equally valid with trajectories reversed in time.

Going to more complex classical situations, TRI as applied to statistical or thermodynamic descriptions of systems leads to conceptual difficulties. While it seems reasonable that the individual particle-particle interactions should obey TRI, yet we know that the entire system must obey, for example, the Second Law of Thermodynamics. Another way of saying this is that the probability that the entropy of an isolated statistical system is decreasing is minutely small. How this can be reconciled with T-invariance is a difficult problem, with no obvious resolution. A very interesting treatise on the problem was published by the philosopher of science Hans Reichenbach (Rei 55).

In quantum mechanics, the time reversal operator can be represented by an operator T with effect on the position, momentum, and spin variables of a particle thus:

$$T\vec{r}_i T^{-1} = \vec{r}_i = \vec{r}_i', \quad T\vec{p}_i T^{-1} = -\vec{p}_i = \vec{p}_i', \quad T\vec{S}_i T^{-1} = -\vec{S}_i = \vec{S}_i' \quad (\text{II-1})$$

where the primes indicate the transformed variables.

The time reversal operator T can be written as the product of a unitary operator U and a complex conjugating operator K , i.e. as an antiunitary operator which preserves the signs of the commutation relations of the form (x, p_x) , (s_x, s_y) , (Boh 69, ch. 1). The effect of such an operator on a matrix element can now be demonstrated:

$$\begin{aligned} \text{if } |A' \rangle &= T |A \rangle = UK|A \rangle, \\ |B' \rangle &= UK|B \rangle \end{aligned} \quad (\text{II-2})$$

then

$$\begin{aligned} \langle B|A \rangle &= \langle B|KU^{-1}UK|A \rangle = \langle KB|U^{-1}|A' \rangle \\ &= \langle B'|A' \rangle^* \end{aligned} \quad (\text{II-3})$$

The matrix elements are equal to the complex conjugate of one another.

In order to establish a relationship between TRIV and transitions between rotational states, one may note that even though T anticommutes with angular momentum, it is possible to combine T with a rotation operator R , representing a rotation of π radians about an axis in the x - y plane. R inverts the z -component of the angular momentum $\vec{I}$ so that RT commutes with $\vec{I}$. Thus eigenstates with quantum numbers IM can be constructed with unity eigenvalues, so that

$$RT|\alpha IM \rangle = |\alpha IM \rangle. \quad (\text{II-4})$$

where α represents other internal quantum numbers. Thus a matrix element of an operator X between two such states transforms as follows:

$$\begin{aligned} &\langle \alpha_2 I_2 M_2 | X | \alpha_1 I_1 M_1 \rangle \\ &= \langle \alpha_2 I_2 M_2 | RT X (RT)^{-1} | \alpha_1 I_1 M_1 \rangle^* \end{aligned} \quad (\text{II-5})$$

If an operator X is invariant under the RT transformation, then all the matrix elements of X are real. Correspondingly if X changes sign under the RT transformation, its matrix elements are purely imaginary. Two fundamental implications of this theorem will now be discussed.

First, consider the electric dipole moment of a particle. This moment obviously changes sign under the rotation R , but is invariant with respect to T . Hence the electric dipole moment is not invariant under the RT transformation and has purely imaginary matrix elements. In the late 1950's it was argued that a search for a possible non-zero electric dipole moment (EDM) of the neutron should be made to help resolve the question of whether time-reversal symmetry might be violated. An excellent review of the history and present status of neutron EDM measurements may be found in Gol 72. The present experimental limit of 10^{-23} e-cm is, unfortunately, compatible with predictions based on $TRIV$ in the electromagnetic, weak, and superweak interactions.

A second consequence of Eqn. II-5 is that, in nuclear level transitions, time-reversal invariance of electromagnetic coupling requires that the relative phases of transition matrices of components of various multipolarity in electromagnetic radiation be real. This was first pointed out by S. P. Lloyd (Llo 51). In Chapter IV it will be shown in detail how an angular correlation measurement from polarized nuclei can lead to a determination of actual values for these relative phase angles. A non-zero phase angle implies directly violation of time-reversal-symmetry. Also β -decay techniques can be used. In Chapter V there is a summary of the $TRIV$ experiments performed to date by nuclear radiation techniques.

First evidence for possible CP-violation

The qualifier "possible" will be used here due to some recent developments in the theory which will be mentioned later.

In the now-famous experiment of Christenson et al (Chr 64) a beam of neutral kaons (K^0) was produced by the reaction $\pi^- + p \rightarrow K^0 + \Lambda$ and the

decay products observed after a known time interval. Neutral kaons normally consist of an equal mixture of two components, K_S^0 and K_L^0 , which have life-times 0.86×10^{-10} and 5.17×10^{-8} sec, respectively, so that the K_S^0 component dies out very quickly by comparison. The K_S^0 component decays by several two meson modes (and $\pi^+ \pi^- \gamma$), but predominantly by the $\pi^+ \pi^-$ mode. The K_L^0 component decays by various three particle modes.

The kaon beam can be described as a sum of two wave functions

$$K_1^0 = (K^0 + \bar{K}^0)/2^{1/2}$$

and

$$K_2^0 = (\bar{K}^0 - K^0)/2^{1/2}$$

taking into account the degeneracy of K^0 and $\bar{K}^0$ with respect to virtual second order weak interactions (Lon 73). These two wave functions will have eigenvalues +1 and -1, respectively, with respect to CP, and they can be identified with the states K_S^0 and K_L^0 previously described.

In the Christenson experiment, the kaon beam was observed to show much larger percentage decay to $\pi^+ \pi^-$ than should be expected by the relative K_S^0 , K_L^0 lifetimes. The conclusion made was that some mechanism was transforming a CP = -1 system (described by K_2^0 above) to a CP = +1 system (described by K_1^0), thus violating CP-invariance.

The ratio of the amplitude of the forbidden decay $K_L^0 \rightarrow \pi^+ \pi^-$ to that of the allowed decay $K_S^0 \rightarrow \pi^+ \pi^-$ serves as a measure of the relative strength of the CP-violating interaction and is equal to $\approx 2 \times 10^{-3}$.

A number of possible origins for this CP-violating interaction appearing in the $K_L^0 \rightarrow 2\pi$ decay have been suggested, and have been summarized, for example, by Blin-Stoyle. (Bli 73a). These may be roughly classified into three groups according to the hypercharge (Y) selection

rules they satisfy. (1) $\Delta Y = 0$. Here the T-violation may arise from the so-called "milli-strong" interaction or else from the electromagnetic interaction itself, and effects of order 10^{-3} might be expected in nuclear electromagnetic transitions. Bernstein, Feinberg and Lee (Ber 65) hypothesized CP-violation in hadron-hadron interactions occurring in second order decay diagrams of the K^0 , and predicted strength comparable to that measured. (2) $\Delta Y = 1$. Here the T-violation is associated with weak forces, and occurs with amplitude 10^{-3} relative to the weak interaction and is hence referred to as "milli-weak". Many of the milliweak models of CP-violation contain a CP-violating $\Delta Y = 0$ component as well. Thus we might expect effects in γ -decay of order 10^{-8} or 10^{-9} . (3) $\Delta Y = 2$. Here some as yet unknown "superweak" interaction produces T-violation of relative amplitude 10^{-13} in γ -decay, but cancellations give rise to an anomalously large effect in K_L^0 -decay; an effect of such magnitude not being expected in any other case. Clearly it is desirable to attempt experiments at the 10^{-3} limit of the strong interaction in γ -decay in order that the possibility of T-violation of the type (1) above may be investigated.

In the light of continuing negative results for TRIV, there is reason to doubt the necessity of involving CP- or T-violation from the neutral kaon decay results. An analysis of the K^0 decay incorporating a wave function which includes a matrix with branch point singularities leads to a CP-preserving explanation of the observed K^0 decay phenomenon (Moh 72). Also, Kenny and Sachs (Ken 73) point out that T-violation in K^0 decay has not been established directly or indirectly, whether or not CPT is violated. Furthermore, if CP is violated in K^0 decay due to a "superweak" interaction, it is not necessarily true that

CPT is invariant in that interaction.

It is of interest to remark here on experiments planned jointly by Los Alamos and UCLA, headed by Roy P. Haddock, to test TRI at the IAMPF accelerator. One experiment (IAMPF proposal number 90) will test reciprocity in the reactions $\pi^- + {}^3\text{He} \rightarrow \gamma + \text{T}$ and $\pi^+ + \text{T} \rightarrow {}^3\text{He} + \gamma$, for a rather direct test of time-reversal invariance. In another experiment (IAMPF proposal number 58) a polarized proton target will be used and cross-sections measured in the reaction $\pi^- + \text{p} \rightarrow \gamma + \text{n}$. Determining the resulting multipole amplitudes is to lead to a quantitative analysis of TRI.

Nuclear γ -Decay Experiments

In order to obtain a meaningful interpretation of the results of a γ -decay T experiment, one must know the extent to which the T-conserving interactions affect the laboratory observables. Here it seems advisable to choose a case for study in which the γ -transition is greatly retarded. In this case it may be possible to obtain a relative enhancement of a T-violating component. This possibility has been recently discussed by Steyert and Krane (Stey 73). The use of retarded transitions has proved effective in studies of P-violation (Jen 70, Lip 71) and has been suggested by Clement and Heller (Cle 71, Cle 73) to be of use in studies of T-violation in γ -decay and by Barroso and Blin-Stoyle (Bar 73) in β -decay. The γ -rays studied in this program all were chosen for their high Weisskopf hindrance values. Other experimental criteria, such as small anticipated perturbation due to precession of the intermediate nuclear state in a γ - γ cascade and

sufficient polarizability in a magnetic medium at the temperatures available, are discussed in Chapter V.

Possible tests for T-violation, P-violation, and simultaneous violation of T and P have been summarized by Boehm (Boe 68). For a γ - γ directional correlation with an initially polarized nuclear level $\vec{I}$, T-violation may be tested by looking for terms of the form

$$(\vec{I} \cdot \vec{k} \times \vec{k}') \quad (\vec{k} \cdot \vec{k}')$$

where $\vec{k}$ and $\vec{k}'$ are the momentum vectors of the first and second γ -rays. This term contains five quantities which change sign under a time reversal, and hence is odd with respect to time. But it contains only four terms (in $\vec{k}$ and $\vec{k}'$) which undergo sign changes under parity inversion (spins are parity invariant), and hence is even with respect to P. Basically, the tests made in this program tested for changes in the directional correlation function when $\vec{I}$ was reversed, effectively changing the sign of the T-odd term above.

In the case of ^{180}Hf , a TRIV test was made on the parity-violating M2 component of the 501 keV γ -ray by means of a search for P-odd, T-odd terms of the form

$$(\vec{I} \cdot \vec{k} \times \vec{k}') \quad (\vec{I} \cdot \vec{k}')$$

which have three parity-odd terms (in $\vec{k}$ and $\vec{k}'$) and again five time-odd terms.

Further details of the mathematics and experimental methods involved in these nuclear γ -decay TRIV tests are given in Chapters IV and V.

CHAPTER III

HYPERFINE MAGNETIC FIELDS ON IMPURITY NUCLEI IN MAGNETIC SUBSTANCES

Origins and Magnitudes

The study of hyperfine fields and corresponding magnetic dipole and electric quadrupole moment interactions with nuclei are of vast importance to several areas of physics — solid state, atomic, and, in the present case, nuclear physics. This introductory discussion will be restricted to the magnetic hyperfine fields acting on non-magnetic impurities in crystalline ferromagnetic transition metals and in ferromagnetic and paramagnetic alloys.

Theories for the sources of the magnetic fields are discussed in several review publications (e.g. Fre 65, Dan 68, Stea 73). Here we shall touch only upon a few more basic ideas of the theory.

Historically, the interaction involved was treated as a coupling between the magnetic dipole moment of a nucleus and the orbital and intrinsic spin moments of a single electron. From Dirac relativistic theory, Fermi derived an explicit equation which we shall quote here mainly for historical interest. The interaction Hamiltonian H_D is given by (Fre 65)

$$H_D = -gg_I\mu_o\mu_n \left\{ \frac{8\pi}{3} \delta(\vec{r}) \vec{S} \cdot \vec{I} + \frac{(\vec{L} - \vec{S}) \cdot \vec{I}}{r^3} + \frac{3(\vec{S} \cdot \vec{r})(\vec{I} \cdot \vec{r})}{r^5} \right\} = -\vec{\mu}_I \cdot \vec{H}, \quad (\text{III-1})$$

where $\vec{L}$ and $\vec{S}$ are the orbital and intrinsic electron spin operators, and $\vec{I}$ is the nuclear-spin operator. The first term (non-zero for s-electrons only), is called the Fermi contact term, an extensively used terminology in the literature.

This basic equation has been corrected for various effects such as high-Z nuclei, relativistic effects, and other subtleties in subsequent work by several workers. This one-electron theory has served as the basis for analysis of more physically realistic complex situations, involving interactions with many electrons and in various thermodynamic phases (solid, gas, liquid).

Consider an atom of atomic number Z in dilute concentration in a ferromagnetic metal (Fe, Co, Ni). The magnetic hyperfine field at the nucleus of that atom can be considered to be from three sources: (1) from polarization magnetization H_{cp}^Z of the inner core (1s, 2s, and 3s) electrons by the magnetic electrons in the metal, (2) from polarization H_{ce}^Z of the 4s conduction electrons by the magnetic moments of the ferromagnetic metal atoms (since d-electrons have essentially zero probability of being at the nucleus, hyperfine field measurements can serve as a measure of the s- or d- nature of the conduction electrons in certain cases, as described in the last experiment of this Chapter), (3) from polarization H_v^Z of the valence electrons of the impurity atom which may overlap to some degree with the ferromagnetic metal atoms, depending on the relative atomic volumes (i.e. upon Z).

Thus the total magnetic hyperfine field may be written as the sum of the three contributions discussed,

$$H_z = H_{cp}^z + H_{ce}^z + H_v^z . \quad (III-2)$$

These terms each fluctuate with atomic number. The major contributions come from the latter two, which tend to be of opposite sign (Stea 73). For example, the signs of the hyperfine fields on impurities in iron are highly dependent upon the valence of the impurity, being negative for valencies less than four and positive for higher valencies (Dan 68).

Solid state theorists have constructed very detailed explanations for these magnetic fields in terms of computed spin densities, charge-screening, etc., further discussion of which would be beyond the scope of this treatise.

The magnitudes of such fields tend to be in the range from tens of kOe to MOe, and tabulations are available (Stea 73, Kos 71).

Measurements of Hyperfine Fields

The most important experimental techniques for determining the magnitudes and signs of magnetic hyperfine fields acting upon impurities in magnetic materials are: (1) nuclear magnetic resonance (NMR), wherein an absorption resonance occurs at energy ΔE corresponding to hyperfine splitting energies between magnetic substates when an applied magnetic field is modulated at an r-f frequency ω ; i.e. at resonance, $E = \hbar\omega = g \mu_N H$, where g is the magnetic g-factor of the particular nuclear state being acted upon, and μ_N is the nuclear magneton. Thus H , the effective hyperfine field, may be determined; (2) electron spin resonance (EPR); (3) measurements of nuclear specific heats; (4) Mössbauer resonance measurements; (5) measurements of

interactions of polarized neutrons with spin-polarized nuclei; (6) measurement of angular distributions of γ -radiations from oriented nuclei; (7) perturbed angular (directional) correlation measurements on γ -rays from radioactive impurity nuclei.

In this present work we have utilized methods (6) and (7), both of which are appropriate to the nuclear orientation methods used during this research program. In the following section are reviewed in more detail the mechanisms and mathematical formalisms involved in analysis of γ -ray angular distribution measurements from oriented or polarized nuclei, and later an explicit example of such measurements in ZrZn_2 is given. In Chapter V an example of the PAC technique used to determine unknown electromagnetic mixing ratios (for the 922-keV γ -ray from ^{184}W), assuming a known hyperfine field, is presented.

Nuclear Orientation

Here we shall give a general phenomenological description of the orientation of impurity nucleus spins in ferromagnetic materials, assuming the presence of large internal nuclear hyperfine fields (as described in the preceding section).

The hyperfine field direction, if aligned along some definite axis by the alignment of the overall magnetization of a sample of ferromagnetic material (usually polycrystalline), defines a quantization z -axis for the nuclear spin. Thus the projection of nuclear spin on that axis has only the values $m\hbar$, where the magnetic quantum number m is an integer restricted by $|m| \leq I$, with $2I + 1$ values. For each value of m there is a corresponding energy equal to $-\vec{\mu} \cdot \vec{H}$ or

$-g\mu_N H$, so that at an ambient temperature T , the relative populations over a set of nuclei are given by the Maxwell-Boltzmann distribution; i.e. $P(m) \propto \exp(g\mu_N H / kT)$, where k is the Boltzmann constant. At high temperatures, i.e. when $kT \gg |g\mu_N H|$, clearly very little difference in population among spin substates exists. However when H is large or T is sufficiently low to reverse the preceding inequality, then the orientations with $m = \pm I$ (depending on the sign of H) are most heavily populated by a significant amount.

It is convenient to define the hyperfine splitting energy in terms of temperature units. The difference in magnetic field interaction energy between two spin substates is given by

$$\Delta E = gI\mu_N H / I = g\mu_N H \quad (\text{III-3})$$

and we define

$$\Delta_T = (\Delta E / k), \quad (\text{III-4})$$

which becomes, with insertion of the values $\mu_N = 5.05 \times 10^{-24}$ ergs/gauss and $k = 1.38 \times 10^{-16}$ (esu), with H in units of MOe,

$$\Delta_T (\text{mK}) = 36.6 g H (\text{MOe}), \quad (\text{III-5})$$

where, of course, $g = (\mu / \mu_N I)$ is the nuclear g -factor. With the definition of Eqn. III-5, the hyperfine splitting energy can easily be compared with the lowest temperature available experimentally, to see if nuclear polarization will be significant.

The orientation parameters B_K used in Chapter IV for quantitative description of nuclear orientation are defined by the explicit

relation (Ste 71)

$$B_K(I) = (2I + 1)^{1/2} (2K + 1)^{1/2} \frac{\sum_{m=-I}^{+I} (-1)^{I+m} \begin{pmatrix} I & I & K \\ -m & m & 0 \end{pmatrix} e^{m\Delta/T}}{\sum_{m=-I}^{+I} e^{m\Delta/T}} \quad (\text{III-6})$$

These parameters have been tabulated as a function of nuclear spin I and of the ratio Δ_T/T (Kra 71b, 73a).

It is important to note that if the quantization axis is taken to be in the $+z$ -direction, then odd K B_K values are all negative (Kra 73b). Also all B_K 's are zero when $K > 2I$.

In the TRIV experiments of this work those terms in the correlation even with respect to parity inversion and odd with respect to time-reversal have only odd K -values, so that in those cases typically only B_1 and B_3 are of any significance, while in the one experiment (with ^{180}Hf) involving P-odd, T-odd terms, only B_2 contributes to that part of the directional correlation function.

It is important to know how real this description of nuclear spin directions actually may be. Some effort in this research program was devoted to investigations of possible nonalignment of the mean direction of spins with the axis of an externally applied magnetizing field.

Approach to Magnetic Saturation for Impurities in Ferromagnetic Polycrystalline Materials

Presented in this section will be a summary of the literature and basic phenomenology of magnetic saturation in cases for which a nucleus to be polarized is dissolved in dilute concentration into a polycrystalline ferromagnetic material. Some present experimental results on both macroscopic and microscopic saturation will also be presented.

Macroscopic saturation

Excellent cornerstone reviews of ferromagnetism are the books of Bozorth (Boz 59) and Kneller (Kne 62) as well as the review by Stoner (Sto 50).

The problem which was addressed as a preliminary to the main experiments of this work was the better understanding of the saturation behavior of hyperfine fields at impurity nucleus sites. Many works have surveyed the magnitudes and sources of such fields for various impurities in Fe, Co, and Ni (Kos 71, Stea 73, Win 71, Kaw 74, Ara 69, Bab 70).

Much work has been done on description of the macroscopic saturation curves for pure polycrystalline nickel and iron (Sto 50, Bro 40, Boz 59). Some of the earliest work in deriving quantitative expressions for macroscopic saturation curves was done by Néel (Sto 50). The historic approach has been to parametrize the magnetization in the form of a McLaurin series of inverse powers of applied magnetic field H , i.e.

$$M = M_S \left(1 - \frac{a}{H} - \frac{b}{H^2} - \dots\right) \quad (\text{III-7})$$

where M_S is the magnetization at saturation of a piece of iron or nickel, following the relation $B = H + 4\pi M$ for the magnetic field B in the object. Experimentally, the $1/H$ dependence has been found negligible, with the second order term accounting for all but a few percent of the H -dependence over a wide range of magnetic induction values. A very satisfactory explicit expression is that given by Stoner (Sto 50) for the magnetization as follows:

$$M = M_S (1 - b/H^2), \quad (\text{III-8})$$

$$\text{where } b = c \times \frac{8}{105} \left(\frac{K_1}{M_S} \right)^2$$

$$\text{and } c = \frac{1}{2} \left[1 + \frac{\alpha}{2(1+\alpha)} + \frac{\alpha^2}{4(1+\alpha)^{3/2}} \ln \left\{ \frac{(1+\alpha)^{(1/2)+1}}{(1+\alpha)^{(1/2)-1}} \right\} \right]$$

$$\alpha = H/4\pi M_S. \quad (\text{III-9})$$

The function c is a very slowly varying function, with $\lim_{\alpha \rightarrow 0} c = 1/2$ and for the range of H from 6 kOe to 22 kOe, $c = 0.7$. This function takes into account the magnetic interaction between domains.

The model which is used to understand saturation behavior is that in which the sample is made up of a large number of magnetic domains of random size distribution and with more or less random distribution of magnetization vector directions and magnitudes. As H is increased, the individual magnetization vectors gradually rotate into the direction of H until, at saturation, they are all colinear.

K_1 is one of a set of three parameters (K_0 , K_1 , K_2) in common use which are defined in terms of the magnetic anisotropy coefficients W_{ijk} ($i, j, k, = 1, 2, 3$) for a particular ferromagnetic crystalline material. The anisotropy coefficients serve as a measure of the anisotropy in magnetic energy density along different axes in the crystalline material. Specifically, one can let $W_{ijk} = \int_0^{I_s} H dI$ be the work required to magnetize a specimen in the $\{ijk\}$ direction. Then the definitions $K_0 = W_{100}$, $K_1 = 4(W_{110} - W_{100})$ and $K_2 = 27(W_{111} - W_{100}) - 36(W_{110} - W_{100})$ are made. K_1 can thus be determined by determining the difference in area under magnetization curves in the $\{110\}$ and $\{100\}$ directions (Boz 59, p. 563). For iron, $K_1 = (57 \pm 5) \times 10^4$ erg/cm³ (Kne 62, p. 190). We note here that Stoner uses K_4 and K_6 for what most other authors call K_1 and K_2 .

The effects of non-ferromagnetic impurities on the macroscopic saturation curve have been experimentally systematized by Néel (Sto 50, p. 143) as follows: If $M_m = (1 - v) \times M_s$, where v is the volume concentration of impurities, then, with $\alpha = H/4\pi M_m$, and M_m the predicted magnetization at saturation, Néel found the following empirical formula:

$$1 - \frac{M}{M_m} = \frac{v}{2(1-v)} \left\{ \frac{2+3\alpha}{4(1+\alpha)^{1/2}} \left[\frac{(1+\alpha)^{1/2} + 1}{(1+\alpha)^{1/2} - 1} \right] - \frac{3}{2} \right\} \quad (\text{III-10})$$

As shall be shown later in a specific example, the effect of small concentrations of impurities in iron on the shape of the macroscopic saturation is very slight.

A final aspect requiring consideration in predicting the macroscopic saturation behavior is the effect of sample geometry via demagnetization. The external magnetic field lines about the sample oppose the applied magnetic induction lines, thus decreasing the effective applied induction. Qualitatively, it is relatively easy to see that a long thin needle-shaped oriented along the direction of $\vec{H}$ results in the least amount of demagnetization. The contribution to demagnetization has been computed and tabulated as a function of sample geometry for both prolate and oblate ellipsoids by Bozorth (Bo 59, p. 846). A correction factor $N/4\pi$ is applied thus:

$$H = H_0 - \frac{N}{4\pi} (B - H) = H_0 - NM, \quad (\text{III-11})$$

to deduce an effective magnetic induction H . Obviously the correction must be iterative, since an initial value for magnetization M must be estimated. For our work, only one iteration sufficed, however.

Applying the demagnetization correction to Eqn. III-8, we have

$$M = M_S [1 - b/(H_0 - NM)^2] \quad (\text{III-12})$$

With just one iteration, one starts with a first estimate of magnetization, M_0 , then simply reevaluates with M by replacing H with $H_0 - NM_0$.

In order to visualize the effect of sample geometry (within the limits of our cryogenic apparatus), saturation curves for three different sample geometries were computed: (a) a "long" needle with no demagnetization, (b) an oblate ellipsoid with the ratio of long axis to short axis equal to 83.3, yielding a demagnetization correction factor $N/4\pi = 0.09$, and (c) an oblate ellipsoid with (long axis/short axis) = 27.7, and $N/4\pi = 0.027$.

Geometries (b) and (c) represent the cases of samples approximately 0.6 cm square x 0.08 mm thick and 1.8 cm square x 0.08 mm thick, respectively. These represented reasonable sample sizes for our apparatus.

For pure polycrystalline iron at 0 C, the saturation magnetization M_S is equal to 1735.2 Oe (Sto 59) and increases by about 2% as temperature decreases to 0 K (Boz 59, p. 431). Since our samples are investigated at temperatures in the mK region, we take for our computations $M_S(0 \text{ K}) = 1770 \text{ Oe}$. A fixed value of 0.6 was taken for the function c defined in Eqn. III-9. Proceeding to compute values for M/M_S with one iteration in the geometries (b) and (c), the three curves of Fig. III-1 were obtained. It is clearly seen that for H below 2 kOe, sample geometry becomes very important and also that in

Fig. III-1. Theoretically predicted macroscopic magnetization saturation curves for pure polycrystalline iron where the sample, which is subjected to an applied magnetic field H , is in the shape of an oblate ellipsoid with the ratio of long axis to short axis equal to (a) ∞ (an infinitely long needle), (b) 83.3, and (c) 27.7. M is the sample magnetization, with saturation value M_s . The effect of demagnetization due to source geometry is graphically clear.

the range of geometries considered, an applied H of only 2 kOe is sufficient to insure macroscopic saturation.

Using Eqn. III-10, the effect of 1% volume concentration of non-magnetic impurities in iron is found to have less than 1% effect over the entire range of these saturation curves.

An experimental measurement of the macroscopic saturation curve was made on an iron sample approximating geometry (b) by the a.c. hysteresis induction method. A coil apparatus for this purpose was constructed as follows. Two coils of 500 and 750 turns of #42 copper wire were wound in opposite directions, one onto a plastic form over the sample in close proximity (to maximize the enclosed flux), the other 1-2 cm away along the same axis. This assembly was attached to the end of a threaded bakelite rod 0.5 in in diameter which in turn was screwed inside an inside threaded 1 in o.d. bakelite tube. On the outside and at one end of the tube was wound a coil 3.5 cm long and 0.6 cm in thickness, consisting of 3200 turns of #34 copper wire. The exterior solenoid had a measured coil constant of 890 Oe/amp on the axis at the center of the coil and 520 Oe/amp at the ends of the coil. The larger interior coil was located off center while the 500 turn coil was located at the center of the solenoid. A ramp generator supplied linear current ramps with peak value 1.5 -2.0 amperes at a repetition rate of 10 hz. The voltage drop across a 10 watt, 1 ohm resistor provided the horizontal sweep signal (proportional to H) on a Tektronix 535A oscilloscope with type 0 operational amplifier plug-in unit. The operational amplifier was adjusted to perform as an inte-

grating amplifier. The vertical signal was supplied by the difference in induced emf between the two small oppositely wound coils described above.

In order to balance the system and to cancel out non-magnetization contributions, the sample was removed and the position of the inner rod with two coils mounted was adjusted by means of rotation until the induced signals from the two coils exactly cancelled, giving no vertical deflection. The sample was then inserted into its pickup coil (500 turn) so that the observed saturation curve on the oscilloscope was due only to the magnetization of the sample with no direct contribution from H_0 . This procedure was followed with the coil-sample assembly immersed in liquid nitrogen, to approximate the low temperature environment expected in nuclear polarization experiments.

Microscopic saturation

From observations on macroscopic saturation such as discussed in the previous section, workers in the past have quite generally assumed that applied fields of 1.5 - 2.0 kOe were sufficient when performing nuclear orientation and perturbed-angular-correlation experiments, as well as any types of experiments making use of nuclear hyperfine magnetic fields, and have assumed that the hyperfine fields saturate in the same way that the sample macroscopically saturates. However, unexpected discrepancies have been noted in nuclear g-factors deduced from PAC measurements (Sio 72), nuclear alignment measurements (Kra 74a), in nuclear γ -ray anisotropy thermometry (Sit 71) (and its use to measure properties of ^3He down to 4 mK) (Joh 68), and in comparisons of Mössbauer and γ -ray anisotropy thermometry. These peculiari-

ties are primarily due to the failure of the achievement of magnetic saturation in the vicinity of the impurity atom.

Investigations into possible sources for such discrepancies have been carried out, primarily by a group at the Weizmann Institute of Science, Rehovot, Israel (Ben 67, 71, Aha 69, 70a, 70b, 71).

The nucleus of an impurity either dissolved or implanted in a ferromagnetic material will, as discussed earlier in this chapter, be subjected to magnetic hyperfine fields of up to 10 MDe. If the ferromagnetic host is magnetically saturated by an externally applied magnetic field H , it might be assumed that the hyperfine fields are aligned in the H -direction. However, as discussed by Aharoni (Aha 69, 70a, 70b), there is a tendency for impurities to orient along certain crystalline axes due to magnetostrictive, magnetocrystalline, exchange and magnetostatic forces. This effect has been observed by Ben-Zvi et al (Ben 67) with Coulomb-excited ^{186}W and ^{148}Nd impurities recoil-implanted into polycrystalline iron and nickel foils.

Experiments performed

In order to determine what magnitudes of externally applied magnetic fields were required to be sure that microscopic saturation was achieved in typical cases of impurities in iron, the γ -ray measurements described below were carried out.

A superconducting coil pair was wound onto a pure copper form designed to fit over the Dewar tail and provide high intensity horizontal magnetic fields. The coils were 0.8 cm in thickness at the outer edge, 0.64 cm thick at the inner radii. The inner radius was 0.5 cm, while the outer radius tapered from 2.1 cm on the sides toward the sam-

ple to 1.65 cm on the sides away from the sample. Each coil had 4520 turns of type T48B Nb-Sn superconducting wire (Supercon, Inc., 9 Eric Drive, Natick, Mass.) with core diameter 0.003 in, copper sheath diameter 0.004 in, and formvar insulation outer diameter 0.005 in. The spacing between the inner surfaces of the two axially placed coils was 1.7 cm, giving a pseudo-Helmholtz configuration. The computed (and measured) coil constant on the axis mid-way between the coils was 1.54 kOe/ampere. The measurement was made with a Bell hall effect device. The field was computed to be uniform to within 5% over the spatial extent of the samples used in this experiment.

The inset of Fig. III-2 shows the samples (S) which were cooled to $T = 25$ mK by the dilution refrigerator (R) in the field (H) produced by the superconducting coils. Currents up to 10 amperes were used to drive the coils, thus providing external fields up to 15.4 kOe. The γ -ray counting rates along the axis of the applied field were measured with two 40 cm^3 coaxial Ge(Li) detectors (D) as H was varied from 0 to 15.4 kOe. The sample (S), as previously discussed, consisted of three iron foils, about 0.1 mm thick, about 2 mm wide, and 6 mm long, side-by-side to form a mosaic 0.1 mm thick. The long axes of the three foils were aligned in the $\vec{H}$ -direction. Of the three 99.99% Fe foils, one had ^{57}Co and ^{54}Mn diffused into it at 1200 C in a hydrogen atmosphere for thermometry purposes (see Appendix B). Another iron foil contained 0.25 at% Os, the third foil 0.07 at% natural Ta. The Fe (Os) and Fe(Ta) foils were irradiated with thermal neutrons to form radioactive ^{191}Os and ^{182}Ta , respectively, and annealed at 900 C in H_2 . The ^{191}Os activity is the parent of $^{191}\text{Ir}^m$ (half-life 4.9 μsec)

Fig. III-2. The normalized orientation parameter $C(H)$, as defined in Eqn. III-14, vs. applied field H . For ^{54}Mn , $E_\gamma = 835$ keV; for ^{57}Co , $E_\gamma = 122$ and 136 keV, for ^{182}Ta , $E_\gamma = 179$ and 264 keV; for ^{191}Ir , $E_\gamma = 129$ keV. The curves indicate approximate fits to the data. Also shown are: the expected relative magnetic saturation curve (macroscopic) for the iron sample (Eqn. III-12) and a schematic of the experimental arrangement.

and, since the spin-lattice relaxation of $^{191}\text{Ir}^m$ is about 0.1 sec (much less than the half-life), the measured γ -ray anisotropies for this isotope are characteristic of the $^{191}\text{Ir}^m$ polarization. Metallographic examination of the three foils after the experiments showed no precipitation of impurities. These examinations, done by optical magnification of slices of the composite sample, were performed by Charles Javorsky of IASL group CMB-6.

Experiments carried out at $T = 1$ K (where there was no significant nuclear alignment) showed that source motion due to $\vec{H}$ was effectively eliminated by mechanical stabilization of the apparatus. This procedure consisted of measuring count rate vs. magnetic field over the full range of H available and determining no significant relationship between the two quantities.

Results

We recall that the angular distribution of γ -radiation, normalized to the isotropic high-temperature limit, is given by

$$W(\theta) = 1 + B_2 A_2 P_2(\cos \theta) \quad (\text{III-13})$$

(The contribution of the P_4 term can be neglected to an accuracy of better than 0.5%) Here θ is the angle between the direction of $\vec{H}$ and the detector axis. (In the present case, the detector axes are parallel to the applied field.) For simplicity all non-orientation dependencies are included in the parameter A_2 . In Fig. III-2 is plotted the parameter

$$C(H) = B_2(H)/B_2(15.4 \text{ kG}) . \quad (\text{III-14})$$

Since the normalized $C(H)$ should be the same for two γ -rays of a given isotope, in some cases we have averaged $C(H)$ for two γ -rays of a particular isotope. The values of E_γ are listed in the caption to Fig. III-2. A small correction to $C(H)$ was made to account for the fact that the nucleus "feels" the sum of hyperfine and applied fields, the two fields being in approximately opposite directions for the nuclei studied.

Also shown in Fig. III-2 is the theoretical magnetic saturation curve for nearly pure polycrystalline iron as was shown for this and other geometries in Fig. III-1.

Interpretation of results

In order to interpret the results we use the simplified model of Ben 67, where the nuclei are assumed subject to a constant hyperfine field lying not along $\vec{H}$, but rather along the generating vector of a cone, making a mean polar angle of θ with $\vec{H}$. The angle θ is a spatial average of the spectrum of angles predicted by Aharoni (Aha 69, 70a, 70b). Under this assumption, along with the observation from Fig. III-2 that the 15.4 kOe applied field essentially saturates $C(H)$, we can write

$$B_2(H)A_2P_2(\theta = 0) = B_2(15.4 \text{ kOe}) A_2P_2(\theta = 0), \quad (\text{III-15})$$

or,

$$C(H) = P_2(\cos \theta). \quad (\text{III-16})$$

We expect that $C(H)$ approaches unity for large H , and that the sign of H must not be significant. Hence, to lowest order,

$$C(H) \approx 1 - b/H^2, \quad (\text{III-17})$$

where b is a constant. Also, $P_2(\cos \theta) = 1 - (3/2)\sin^2\theta$, and thus

$$\sin \theta \propto 1/H. \quad (\text{III-18})$$

In Fig. III-3, $\sin \theta$ is plotted against $1/H$, and we note an approximately linear behavior in the region $H = 1-15$ kOe.

Aharoni (Aha 69), in considering only magnetostrictive effects, also predicts $\sin \theta \propto 1/H$. In a later publication, Aharoni (Aha 70) considers also magnetocrystalline, exchange, and magnetostatic forces for W and Nd, and concludes a much weaker predicted dependence of $\sin \theta$ upon $1/H$. The present results indicate a somewhat stronger relationship between θ and H than is predicted by the latter publication. Elementary considerations would also suggest a $1/H$ behavior, in contrast to Aharoni's latter calculations. If on the average there is a small crystalline field on the impurity atomic electrons of strength $F_{\perp}$ perpendicular to $\vec{H}$ and $F_{\parallel}$ parallel to $\vec{H}$ then $\sin \theta \approx \tan \theta = F_{\perp}/(F_{\parallel} + H) \propto 1/H$ for large H .

It is noted that the angle θ is smaller in Mn than in the other samples tested. (Qualitative measurements on ^{125}Sb , ^{60}Co , and ^{193}Os in Fe show results similar to Co, Ta, and Ir in the $H \approx 1$ kOe region.) Both Co and Mn are similar in size and electronic configuration to Fe. However, transition elements lying to the right of Fe in the periodic table are found to cause a widespread perturbation when introduced into the Fe lattice (out to $10 \text{ \AA}$, involving ≈ 200 atoms). Moreover, Co is very magnetostrictive in Fe

Fig. III-3. The value of $\sin \theta$, determined from Eqn. III-16, vs. $1/H$, for ^{191}Ir , ^{57}Co , ^{182}Ta , and ^{54}Mn , based on measurements of the γ -rays listed for Fig. III-2. The apparent linear behavior in the region $H = 1-15$ kOe, discussed in the text, is illustrated by approximately fit straight lines.

(Boz 59, p. 380), so that the observed cone angle for Co is larger than for Mn. Recent experiments (Ara 69) show that Mn and Ta dilutely alloyed into iron have none other than a dilution effect on the macroscopic saturation of the iron, as discussed earlier in this chapter. However, the large nuclear radius of Ta relative to iron results in a local strain and thus the large observed cone angle θ .

Alignment of nuclei at a non-zero cone angle with respect to H is important for interpretations of a number of measurements. For small θ (large H) the sizes of errors which result from ignoring θ can be estimated. We shall discuss here the effect of H on perturbed-angular-correlation (PAC) measurements and on determinations of nuclear orientation parameters.

As shown by Ben-Zvi, et al, (Ben 67) in an inhomogeneous field varying from 0.4 to 1.5 kOe the resulting error in previous determinations of hyperfine couplings is about 20-30% and the cone angle is calculated to be about 30° for ^{186}W in Ni and Fe. Examination of the calculations of Ben-Zvi, et al, indicate that this underestimation should decrease as $\theta^2 \propto 1/H^2$. A 20% underestimation of the hyperfine interaction was observed for ^{106}Pd in Fe for $H = 1$ kOe (Joh 68). Deutch and Nielsen (Deu 69, Nie 67) have also seen low-H anomalies in PAC measurements which our present results suggest are attributable to a lack of impurity magnetic saturation.

As another specific example, integral PAC measurements of the magnetic moments of the $I = 5/2$ levels in ^{191}Ir and ^{193}Ir , with Ir diffused into iron as a source, and with an applied field $H = 1.2$

kOe have yielded $\mu = (0.42 \pm 0.05)\mu_N$ and $(0.48 \pm 0.08)\mu_N$ (Avi 70), respectively; whereas another measurement utilizing Coulomb excitation and recoil-into-gas, not involving a ferromagnet host, yielded the higher values $\mu = (0.58 \pm 0.10)\mu_N$ and $(0.73 \pm 0.13)\mu_N$, respectively (Ber 72). (For further PAC examples, see Sio 72 and Kra 72a).

For $H = 1$ kOe, measurements ignoring θ would result in underestimates of the orientation parameter B_2 by about 15%. Again, the magnitude of this error decreases like $1/H^2$, since $1 - P_{2,4}(\cos \theta) \approx \theta^2$ for small θ , and $\theta^2 \propto 1/H^2$. Corresponding errors would be observed in determinations of nuclear moments and in those particular decay parameters which depend directly on the value of B_K (i.e., U_K and A_K).

The cone angle effect can also help to explain discrepancies in γ -ray and Mössbauer low-temperature thermometry measurements. Thermometry interpretations, as well as other results of this section are reported in a paper published by the author with collaborators (Kra 73c).

In conclusion, for the materials studied, $\theta = 20^\circ/H(\text{kOe})$, to within 20%. For Mn, θ appears to be near the lower extreme of the 20% spread, while values of θ for other nuclei [Co, Sb, Ta, Ir, Os, W (Ben 67), and Pd (Owe 69)] are near the upper extreme of this estimate. Estimates and corrections of previous PAC, γ -anisotropy and thermometry errors are feasible; future experimenters should utilize externally applied polarizing fields greater than 3 to 5 kOe in order to reduce the cone angle θ to a negligible

value.

We are gratified to see recognition of the cone angle problem in hyperfine field measurements (e.g. Kaw 74) since the publication of this work (Kra 73c) mentioned above.

Nuclear Orientation Example -- Hyperfine Fields in ZrZn_2

During the early part of this research program, the interesting magnetic properties of ZrZn_2 came to attention as a worthwhile preliminary investigation. The results which follow were completed and published early in the program (Mur 73).

The intermetallic ZrZn_2 is a weak Stoner type itinerant ferromagnet, with a net moment of about $0.16 \mu_B$ per molecule (Kna 71). The magnetic properties of this material have been ascribed to a narrow d-electron conduction band of about 0.05 eV half-width with a high density of states at the Fermi level. The magnetism is thought to reside mainly at the Zr sites, and the extent of s-d hybridization has been thought to be small.

Since hyperfine magnetic fields are very sensitive to s-electron character in polarized electrons (Campb 69), measurements of hyperfine fields were made as a means of detecting any significant s-d hybridization. It has been found possible to study the Zr site using NMR at temperatures well above the Curie temperature, 35 K (Yam 65), but as the temperature is lowered to 80 K, the resonance signal broadens and disappears. Furthermore, while broad lines can be studied by Mössbauer techniques, Zr and Zn are not suited to the

Mössbauer approach. Since one must resort to the reliable but less sensitive methods of nuclear orientation in order to search for s-d hybridization at the Zr and Zn sites.

A 3 mm cube of polycrystalline $ZrZn_2$ (part of the same batch used in Smi 71) was irradiated with thermal neutrons to produce radioactive $^{65,69}Zn$ and $^{95,97}Zr$, and, from beta-decay of the zirconium isotopes, also $^{95,97}Nb$. The sample was soldered with indium, together with an iron foil in which a small amount of ^{54}Mn and ^{60}Co radioisotopes had been dissolved, to the copper probe of the dilution refrigerator. The sample was cooled to a steady temperature of 18 mK, the temperature being continuously monitored by the anisotropy of γ -rays from the ^{54}Mn and ^{60}Co (see Appendix B).

Following a well-established technique, two 40 cc Ge(Li) detectors were placed at a 90° angle with respect to the sample, in the horizontal plane and, by means of two perpendicular pairs of superconducting coils, a 2-1/2 kOe field applied to the sample was rotated through 90° alternately between data runs, first pointing along the axis of one detector, then towards the other detector. Differences in γ -counting rates are related to nuclear alignment through the standard relationship $W(\theta) = 1 + \sum_{K=2,4,\dots} Q_K B_K U_K A_K P_K (\cos \theta)$, $K = 2, 4, \dots$, where θ is the observation angle with respect to the applied field direction; Q_K, B_K, U_K, A_K, P_K are defined in Chapters III and IV. In this case, a good approximation is obtained by including only the $K = 2$ term.

Within experimental error limits, nuclear alignments of Zn, Zr and Nb were found to be nil, resulting in upper limits on possi-

ble hyperfine magnetic fields at those sites. The data are summarized in Table III-1.

From the data, it is concluded that the magnitudes of hyperfine fields at Zn, Zr, and Nb sites must be less than about 25, 60, and 30 kOe, respectively. For Zr, this result is compatible with the measurements of Ogawa and Sakamoto (Oga 67), which yielded a value -55 kOe, and also with the measurements of Yamadaya and Asanuma (Yam 65) — -38 kOe.

Campbell (Campb 69) has shown that the magnetic moment of s conduction electrons gives rise to hyperfine fields $H_s = 3.5, 2.0,$ and $2.3 \text{ MOe}/\mu_B$ in Zn, Zr, and Nb, respectively, while d-electron effects are about an order of magnitude less.

Neglecting possible mutual cancellation of d and s contributions yields s-electron magnetism $\mu_s (\mu_B) \propto H/H_s$, with upper limits 0.007, 0.03, and 0.013 μ_B for Zn, Zr, and Nb. On the other hand, if the hyperfine contribution from d-type electrons is large, then cancellation by about a 10% s-contribution would be occurring. In either case we can conclude that only very little s-d hybridization of the conduction electrons exists in ZrZn_2 .

Table III-1. γ -Ray asymmetry data, $ZrZn_2$ experiment

Aligned nucleus	γ -energy (keV)	Measured $B_2 U_2 A_2 Q_2$ (units of 10^{-4})	Calculated ^(a) $U_2 A_2$	Δ (mK)	H (kOe)
$^{69}_{30}Zn_{39}$	439	3.3 ± 9.3	-1.61	< 0.18	$< 25^{(b)}$
$^{95}_{40}Zr_{55}$	756	-7.7 ± 12	0.25 ± 0.11	< 0.9	< 60
$^{95}_{41}Nb_{55}$	765	25 ± 27	$0.23 \pm 0.065^{(c)}$	< 1.8	< 35
$^{97}_{41}Nb_{56}$	658	13 ± 15.5	$0.23 \pm 0.065^{(d)}$	< 1.08	$< 22^{(e)}$

(b) Assuming nuclear magnetic moment of $9/2^+$ level in $^{69}_{30}Zn$ given by $|\mu| \approx 1\mu_N$ (deduced from nuclear systematics and measured moments of spin $9/2^+$ levels in the even Z, odd N isotopes $^{69,71,73}_{32}Ge$).

(c) Taking E2/M1 mixing ratio $\delta = -0.06$ to $+0.08$ (Mes 72).

(d) Assuming δ same as for 765 line.

(e) Assuming nuclear magnetic moment of $^{97}_{41}Nb$ spin $9/2$ ground state $|\mu| \approx 6\mu_N$ (deduced from measured values for spin $9/2$ states in the odd Z, even N isotopes $^{93,95}_{41}Nb$ and $^{99}_{43}Tc$).

CHAPTER IV

DIRECTIONAL CORRELATION OF γ - γ RADIATION CASCADES FROM ORIENTED NUCLEI

Since the formalism used in the present work is rather new and since it differs in important respects from the correlation formalism long in general use by nuclear physicists and physical chemists, we shall review concisely the salient points of both the "old" and "new" formalisms.

The "Old" Formalism

Here we refer to the formalism of Frauenfelder and Steffen (Fra 65) which, until a few years ago, was the universally accepted standard for handling γ - γ correlations. The advent of correlations from nuclei polarized at ultralow temperatures required new mathematics to account for the degree of polarization. Also, the Rose-Biedenharn sign convention for electromagnetic mixing ratios in the case of mixed transitions has been replaced in the standard reference tables (such as the Nuclear Data Sheets) by the Krane and Steffen convention (Kra 70).

To review the earlier convention, we consider the case of a cascade of unmixed γ -radiations on initial nuclear level of spin I_i to an intermediate level of spin I (giving off γ -radiation of multipolarity L_1) and finally a transition to the final level of spin I_f (giving off a γ -ray of

multipolarity L_2). In the unpolarized (high-temperature) state, all magnetic substances with magnetic quantum numbers m_i ($|m_i| \leq I_i$) are considered to be equally populated. The quantization axis is taken to coincide with the direction of the first γ -ray. The probabilities $P(m)$ of population of the magnetic substates of quantum number m ($|m| \leq I$) for the intermediate state are given by

$$P(m) \propto \sum_{m_i} G(m, m_i) F_{L_1}^{M_1}(\theta=0), \quad (\text{IV-1})$$

where $G(m, m_i)$ is the relative transition probability for a transition from state (I_i, m_i) to state (I, m) , and $F_{L_1}^{M_1}(\theta=0)$ is the directional distribution function for the (L_1, M_1) radiation component along the initial direction, with $M_1 = m_i - m$. Thus the direction correlation function for the L_2 radiation vs. the L_1 radiation is given by summing over intermediate and final level substates as

$$W(\theta) \propto \sum_{m, m_f} P(m) G(m, m_f) F_{L_2}^{M_2}(\theta). \quad (\text{IV-2})$$

To make this more specific, one notes that the transition probabilities can be written in terms of Clebsch-Gordon coefficients thus:

$$G(m, m_f) = |\langle I_f m_f L_2 M_2 | I \rangle|^2 \quad (\text{IV-3})$$

which combined with a limit on M_1 to values ± 1 along the $\theta=0$ direction gives

$$W(\theta) \propto \sum_{m_f, m_i} |\langle I_f m_f L_1 \pm 1 | I_i m_i \rangle|^2 F_{L_1}^{\pm 1} \langle I_f m_f L_2 M_2 | I m \rangle F_{L_2}^{M_2}(\theta), \quad (\text{IV-4})$$

noting that a general expression for this directional distribution can be written as

$$\begin{aligned}
 F_L^M(\theta) = & \frac{1}{2} \left[1 - \frac{M(M+1)}{L(L+1)} \right] |Y_{L,M} + 1|^2 \\
 & + \frac{1}{2} \left[1 - \frac{M(M-1)}{L(L+1)} \right] |Y_{L,M} - 1|^2 \\
 & + \frac{M^2}{L(L+1)} |Y_{L,M}|^2
 \end{aligned}
 \tag{IV-5}$$

It can be shown in a very elegant manner (Fra 65) that for the case of an unperturbed - cascade of two pure (unmixed) radiations as described above, the correlation function reduces to the form

$$W(\theta) = \sum_K \frac{A_K}{K} P_K(\cos \theta), \tag{IV-6}$$

with the following properties.

- (a) For parity conserving transitions (the usual case till recently) where only directions or linear polarizations are measured, only even values of K are included in the summation.
- (b) K is limited by $0 < K < \text{Min}(2I, 2L_1, 2L_2)$.
- (c) If the multipolarity of the second γ -ray (L_2) is the lowest allowed by selection rules, and that transition unmixed, then the directional correlation between yet another succeeding γ -ray of the same multipolarity (L_2) and the initial γ -ray will be identical to the previous correlation function.

In the case described, the function A_{KK} can be factored into two parts, each depending on only one of the transitions, i.e.

$$A_K = F_K (L_1 L_1 I_i I) F_K (L_2 L_2 I_f I) \quad (IV-7)$$

where

$$F_K = (-1)^{I_i + I - 1} (2L + 1) | (2I + 1)(2K + 1) |^{1/2} \begin{pmatrix} L & L & K \\ 1 & -1 & 0 \end{pmatrix} \left\{ \begin{matrix} L & L & K \\ I & I & I_i \end{matrix} \right\} \quad (IV-8)$$

are functions which are tabulated or easily computed by looking up the 3-j and 6-j symbols.

For mixed transitions the electromagnetic mixing ratio is defined by ratios of reduced matrix elements, i.e.

$$\delta_1(\gamma) = \frac{\langle I || L_1^1 \Pi_1^1 || I_i \rangle}{\langle I || L_1^1 \Pi_1^1 || I_i \rangle} \quad (IV-9)$$

where the reduced matrix elements, originally defined (by means of the Wigner-Eckart theorem) in terms of the interaction hamiltonian H between an initial state I_i and state I as

$$\langle I_i || L \Pi || I \rangle = \frac{\langle \text{Im} L \Pi || H || I_i M_i \rangle}{\langle \text{Im} L \Pi || I_i M_i \rangle} \quad (IV-10)$$

From a fundamental theorem of quantum mechanics, if the transition can be described by a hamiltonian operator H which commutes with the time reversal operator T , then the reduced matrix elements defined above may be chosen to be real, so that

$$\langle I || L || I_1 \rangle^* = \langle I || L || I_1 \rangle \quad (\text{IV-11})$$

and the relative phases of electromagnetic mixing ratios are then always 0 or π (Llo 51). Thus we say that the mixing ratio is real if time reversal invariance (TRI) is obeyed.

In the doubly mixed case (two components of multipolarity L_1 and L'_1 in the first radiation; L_2 and L'_2 in the second) the correlation coefficients A_{KK} are given by

$$A_{KK} = A_K(L_1 L'_1 I_1 I) A_K(L_2 L'_2 I_f I) \quad (\text{IV-12})$$

where

$$A_K(L_1 L'_1 I_1 I) = \frac{F_K(L_1 L_1 I_1 I) + 2\delta_1 F_K(L_1 L'_1 I_1 I) + (\delta_1)^2 F_K(L_1 L'_1 I_1 I)}{1 + (\delta_1)^2} \quad (\text{IV-13})$$

with an analogous expression for $A_K(L_2 L'_2 I_f I)$. Note that in this earlier convention, δ_1 must be modified from its normally tabulated value by a minus sign [actually $(-1)^{L'+L}$] due to the ordering of the oriented state in the matrix elements for the Rose-Biedenharn convention. In the more recent Krane & Steffen convention (Kra 70), $A_K(L_1 L'_1 I_1 I)$ is replaced by an orientation parameter B_K and the mixing ratio is not changed in sign; instead in B_K the factor $(-1)^{L'+L}$ is introduced into the middle term of the numerator.

The F-functions are given by the form

$$F_K(LL'I_iI) = (-1)^{I_i+I}^{-1} [(2L+1)(2L'+1)(2I+1)(2K+1)]^{1/2} \\ \times \begin{pmatrix} L & L' & K \\ 1 & -1 & 0 \end{pmatrix} \begin{Bmatrix} L & L' & K \\ I & I & I_i \end{Bmatrix} \quad (\text{IV-14})$$

We note here that, for ordinary (non-spin-polarization) γ - γ correlations, this formalism is quite practical and useful, with the following precautions:

- (a) When looking up the mixing ratio for an electromagnetic transition, one must take care as to which sign convention has been utilized (most newer papers and tables are in the Krane-Steffen convention).
- (b) The F-functions have been conveniently tabulated (e.g., Kra 71b), but are usually listed with the spin orders reversed from the previous convention (i.e. $F(L_1, L_2, I_2, I_1)$) so that care must be used in reading the tables.

This discussion has purposely been brief so that heavier emphasis may be placed on the newer convention which is more suitable to our present purposes.

The Krane, Steffen and Wheeler DCO Convention

The following will be a precis of the most significant portions of the "K,S,&W" notation, presented in more complete detail in the references (Kra 70, 71b, 73a, 73b, 73d)

An initial nuclear level of spin I_i is considered to be oriented

or polarized with respect to some orientation axis, due to the phenomena discussed earlier in Chapter III. This level decays to a level of lower energy with spin I_2 giving off radiation X_1 , and finally there is a subsequent decay to the final level of spin I_3 with emission of radiation X_2 . We thus write the sequence as $I_1 \xrightarrow{X_1} I_2 \xrightarrow{X_2} I_3$ in general. A correlation between radiations X_1 and X_2 is desired. For this purpose three angles θ_1 , θ_2 , and ϕ (see Fig. IV-1) are chosen to describe the directions of the momentum vectors $\vec{k}_1$ and $\vec{k}_2$ of the two radiations. The basic axis of the system is the orientation axis of the initial state I_1 . This is defined as the z-axis. (Normally this will correspond to the direction of some known laboratory magnetic field). θ_1 is the angle from $\vec{k}_1$ to the z-axis, θ_2 is the angle from the z-axis to $\vec{k}_2$, and ϕ is the angle between the projection of $\vec{k}_1$, in the plane the z-axis and the equivalent projection of $\vec{k}_2$, as shown in Fig. IV-1. We have utilized this convention consistently throughout the remainder of this work.

The experimental directional correlation function describing the coincidence rate as a function of geometry, where both radiations are observed, is written as the summation

$$\begin{aligned}
 W(\theta_1, \theta_2, \phi) = & \sum_{\lambda_1 \lambda_2} B_{\lambda_1} (I_1) A_{\lambda_1}^{\lambda_2 \lambda_1} (X_1) A_{\lambda_2} (X_2) \\
 & \times H_{\lambda_1 \lambda_2} (\theta_1, \theta_2, \phi) Q_{\lambda_1} (X_1) Q_{\lambda_2} (X_2) . \quad (IV-15)
 \end{aligned}$$

Fig. IV-1. The angles in a directional correlation of two successive radiations, X_1 and X_2 , emitted from an axial symmetric oriented source S (from Kra 73b).

The indices λ_1 and λ_2 are the tensor ranks of the statistical tensors describing the nuclear states I_1 and I_2 . If for example, the initial state is unoriented then $\lambda_1 = 0$. $\lambda_1 = 1$ corresponds to orientation, $\lambda_1 = 2$ to polarization of the initial state. The index λ is the tensor rank of the radiation field and its observation efficiency tensor; i.e. $\lambda = 0$ corresponds to non-observation of the field in a particular direction. Odd values of λ_2 or λ correspond to cases where radiation X_2 or X_1 is a γ -ray which either violates parity selection rules or is circularly polarized.

Limits on the integral summation indices are: $0 \leq \lambda_1 \leq 2I_1$, $\lambda \leq L + L'$, and $\lambda_2 + \lambda_1 \geq \lambda \geq |\lambda_2 - \lambda_1|$. A tabulation of the implications of the oddness and evenness of λ_1 , λ , λ_2 and the sum $\lambda_1 + \lambda + \lambda_2$ is presented in Table IV-1 (from Kra 73d). We note from Table IV-1 that terms in $W(\theta_1, \theta_2, \phi)$ corresponding to T-violation in X_1 are separable, as are those corresponding to P-violation in X_1 . This useful property will be discussed further below.

The orientation parameters $B_{\lambda 1}(I_1)$ for the initial state were discussed earlier in Chapter III.

The generalized angular distribution coefficients $A_{\lambda}^{\lambda_2 \lambda_1}(X_1)$ for the first radiation are described in general for various multipolarity components $L, L' \dots$

$$A_{\lambda}^{\lambda_2 \lambda_1}(X_1) = \frac{\sum_{LL'} S_{\lambda}^{\lambda_2 \lambda_1}(LL'; X_1) F_{\lambda}^{\lambda_2 \lambda_1}(LL'; I_2 I_1)^{<I_2> <I_1> | X_1(L) | | I_1 > I_2 | | X_1(L') | | I_1 > *}}{\sum_L S_0(LL; X_1) | <I_2> | X_1(L) | | I_1 > |^2} \quad (IV-16)$$

Table IV-1. Restrictions on indices $\lambda_1, \lambda, \lambda_2$ (from Kra 73d)

Index	Allowed Range	Odd	Even
λ_1	$\lambda_1 \leq 2I_1$	Initial state polarized	Initial state aligned
λ	$\lambda \leq L_1 + L_1'$	Radiation X_1 is β , P-violating or circularly polarized γ	Directional distribution of P-nonviolating radiation X_1
λ_2	$\begin{cases} \lambda_2 \leq L_2 + L_2' \\ \lambda_2 \leq 2I_2 \end{cases}$	Radiation X_2 is P-violating or circularly polarized γ	Directional distribution of P-nonviolating radiation X_2
$\lambda_1 + \lambda + \lambda_2$	$\begin{cases} \lambda_1 + \lambda_2 \geq \lambda \\ \lambda_1 - \lambda_2 \leq \lambda \end{cases}$	T-violation	T-nonviolation

where the sum is carried out over the allowed multipolarities L, L' such that $\lambda < L + L'$. The b_λ are the particle parameters (unity for gamma radiation), and the $F_\lambda^{\lambda_2 \lambda_1}$ are generalized F-coefficients, defined by

$$F_\lambda^{\lambda_2 \lambda_1}(LL'I_2I_1) = (-1)^{L' + \lambda_1 + \lambda_2 + [\hat{I}_1 \hat{I}_2 \hat{L} \hat{L}' \hat{\lambda}_1 \hat{\lambda}_2]}^{1/2} \times \begin{pmatrix} L & L' & \lambda \\ 1 & -1 & 0 \end{pmatrix} \begin{Bmatrix} I_2 & L & I_1 \\ I_2 & L' & I_1 \\ \lambda_2 & \lambda & \lambda_1 \end{Bmatrix}, \quad (\text{IV-17})$$

where $\hat{x} = 2x + 1$.

An important property is that

$$F_\lambda^{\lambda_2 \lambda_1}(LL'I_2I_1) = (-1)^{\lambda_1 + \lambda + \lambda_2} F_\lambda^{\lambda_2 \lambda_1}(L'LI_2I_1) \quad (\text{IV-18})$$

The brackets () and { } indicate Wigner 3-j and 9-j symbols, respectively. The indices λ_2, λ_1 and λ must form a closed vector triangle; that is, $\lambda_2 + \lambda_1 > \lambda > |\lambda_2 - \lambda_1|$. The coefficient S is a function of λ and of the X_1 radiation multipolarities; i.e.

$$S = \frac{1}{2} \left\{ 1 + (-1)^{\Lambda_\pi + \Lambda_{\pi'} + L + L' + \lambda} \right\} \quad (\text{IV-19})$$

where

$$\Lambda_\pi = \begin{cases} 0 & \text{for electric multipole component} \\ 1 & \text{for magnetic multipole component.} \end{cases} \quad (\text{IV-20})$$

(Kra 74c).

The various types of $A_\lambda^{\lambda_2 \lambda_1}$ terms will now be enumerated and discussed briefly, for both doubly and triply mixed initial γ -rays.

P-even, T-even terms

For these terms, $\lambda_1 + \lambda + \lambda_2$ is even, and, with no parity violation, λ_1 and λ_2 are also even, hence λ is also even and the factor S is equal to 1 for E1-M2 or M1-E2 mixtures and also for all cross-terms involved in an E1-M2-E3 mixture. Thus, for a doubly mixed γ_1 with multipolarities L_1 and L'_1 ,

$$A_{\lambda}^{\lambda_2 \lambda_1} = \left[F_{\lambda}^{\lambda_2 \lambda_1} (L_1 L_1 I_2 I_1) \langle L \rangle \langle L \rangle^* + F_{\lambda}^{\lambda_2 \lambda_1} (L_1 L_1 I_2 I_1) \langle L \rangle \langle L' \rangle^* \right. \\ \left. + F_{\lambda}^{\lambda_2 \lambda_1} (L'_1 L_1 I_2 I_1) \langle L' \rangle \langle L \rangle^* + F_{\lambda}^{\lambda_2 \lambda_1} (L'_1 L_1 I_2 I_1) \langle L' \rangle \langle L' \rangle^* \right] \\ / (|\langle L \rangle|^2 + |\langle L' \rangle|^2) , \quad (IV-21)$$

where $|L'| = |L| = |I_2| |L| |I_1|$ is the square root of the transition rate of the L-component of radiation.

If L' is out of phase by angle η with respect to L , then $\langle L \rangle \langle L' \rangle^* = |L| |L'| e^{-i\eta}$ and $\langle L' \rangle \langle L \rangle^* = |L| |L'| e^{i\eta}$ which combined with the property of Eqn. IV-18 reduces Eqn. IV-16 to

$$A_{\lambda}^{\lambda_2 \lambda_1} = \frac{F_{\lambda}^{\lambda_2 \lambda_1} (L_1 L_1 I_2 I_1) |L|^2 + 2 F_{\lambda}^{\lambda_2 \lambda_1} \text{Re} \delta_1 + |\delta_1|^2 F_{\lambda}^{\lambda_2 \lambda_1} (L_1 L'_1 I_2 I_1)}{1 + |\delta_1|^2} \quad (IV-22)$$

This is the analogue of Eqn. IV-13 in the "old" notation for ordinary angular correlations. We note in the middle term of the numerator that $\text{Re} \delta_1$ appears rather than just δ_1 . In the absence of TRIV, i.e., if

$\sin \eta = 0$, then $\text{Re}\delta_1 = \delta_1$. However, in the more general case we cannot make such an assumption. (The values of mixing ratios obtained from electron conversion measurements, etc., are actually $\text{Re}\delta_1$).

For a triply mixed initial radiation, with radiation components L_1, L_1', L_1'' with $L_1' = L_1 + 1$, $L_1'' = L_1 + 2$, and with mixing ratios $\delta = \frac{\langle L_1' \rangle}{\langle L_1 \rangle}$, $\bar{\delta} = \frac{\langle L_1'' \rangle}{\langle L_1 \rangle}$ and phase angles η and $\bar{\eta}$ associated with δ and $\bar{\delta}$, Equation IV-22 becomes, for the P-even, T even terms,

$$\begin{aligned} A_{\lambda}^{\lambda_2 \lambda_1}(\delta_1, \bar{\delta}) = & \left[F_{\lambda}^{\lambda_2 \lambda_1}(L_1 L_1 I_2 I_1) + 2\text{Re}\delta F_{\lambda}^{\lambda_2 \lambda_1}(L_1 L_1' I_2 I_1) \right. \\ & + |\delta|^2 F_{\lambda}^{\lambda_2 \lambda_1}(L_1' L_1' I_2 I_1) + 2\text{Re}\bar{\delta} F_{\lambda}^{\lambda_2 \lambda_1}(L_1 L_1'' I_2 I_1) \\ & + 2|\delta||\bar{\delta}| F_{\lambda}^{\lambda_2 \lambda_1}(L_1' L_1'' I_2 I_1) [\cos(\bar{\eta} + \eta)] \\ & \left. + |\bar{\delta}|^2 F_{\lambda}^{\lambda_2 \lambda_1}(L_1'' L_1'' I_2 I_1) \right] / (|\delta|^2 + |\bar{\delta}|^2) . \quad (\text{IV-23}) \end{aligned}$$

P-odd, T-even terms

For these terms λ_2 is even, but both λ_1 and λ must be odd. For a doubly mixed plus parity-violating γ -ray (L_1, L_1' and L_1^P) such that L_1^P violates parity then coefficient $S = 0$ for cross terms involving L and L_P . This is of interest to the analysis of $E1-M2(E2)$ transitions such as the 501-keV γ -ray from $^{180}\text{Hf}^m$. In such a case, equation IV-16 becomes

$$A_{\lambda}^{\lambda_2 \lambda_1} = \frac{2\text{Re}\epsilon}{1 + |\delta_1|^2 + |\epsilon|^2} \left\{ F_{\lambda}^{\lambda_2 \lambda_1} (L_1 L_1^P I_2 I_1) + |\delta_1| F_{\lambda}^{\lambda_2 \lambda_1} (L_1' L_1^P I_2 I_1) \right\} \quad (\text{IV-24})$$

where $\epsilon = \frac{\langle L_1^P \rangle}{\langle L_1 \rangle}$ is the mixing ratio for the parity-violating component.

P-even, T-odd terms

For these terms, λ must be odd, hence in a doubly mixed γ -ray,

$S = 0$ for all terms except the two cross-terms, hence

$$A_{\lambda}^{\lambda_2 \lambda_1} = \frac{F_{\lambda}^{\lambda_2 \lambda_1} (L_1 L_1' I_2 I_1) \langle L \rangle \langle L' \rangle^* + F_{\lambda}^{\lambda_2 \lambda_1} (L_1' L_1 I_2 I_1) \langle L' \rangle \langle L \rangle^*}{|L|^2 + |L'|^2} \quad (\text{IV-25})$$

In all T-odd cases, the property of F-functions in Equation IV-18 requires that the two F-functions in the numerator of Equation IV-25 be of opposite sign. Hence in doubly-mixed P-even, T-odd terms

$$A_{\lambda}^{\lambda_2 \lambda_1} = \frac{-2i F_{\lambda}^{\lambda_2 \lambda_1} (L_1 L_1' I_2 I_1) |L| |L'| \sin \eta}{|L|^2 + |L'|^2} \quad (\text{IV-26})$$

and we see that the phase $\sin \eta$ factors out of such terms, giving a clue as to how the phase may be determined.

In order to analyze the TRIV experiment for the triply-mixed (E1-M2-E3) 922 keV γ -ray from ^{184}W , which is described experimentally in Chapter V, we derive the P-even T-odd terms here explicitly.

In order to do this, we note that $S = +1$ for the $(L, L+1)$, $(L, L+2)$ and $(L+1, L+2)$ terms, since λ must be even (for no P-violation), λ_2 even,

and thus λ_1 odd (to maintain the oddness of the sum $\lambda_1 + \lambda + \lambda_2$). The angular distribution may then be written as $W = W_0 + W_T + W'$, where W_0 and W_T contain the T-conserving and T-violating terms (or, equivalently, the $\lambda_1 = \text{even}$ and $\lambda_1 = \text{odd}$ terms), respectively. An additional term W' has been included to account for possible polarization-sensitive T-nonviolating effects discussed below. By periodically reversing the direction of nuclear polarization we compute the asymmetry A , defined by

$$A = \frac{W(\uparrow) - W(\downarrow)}{W(\uparrow) + W(\downarrow)} . \quad (\text{IV-27})$$

$W(\uparrow)$ and $W(\downarrow)$ indicate the measured correlation with nuclear spin up and down, respectively. Since the λ_1 -odd terms in W_T change sign under this reversal while the λ_1 -even terms in W_0 do not, this may be written, assuming $W_T \ll W_0$, as

$$A \approx \frac{W_T + W'}{W_0} . \quad (\text{IV-28})$$

After correcting for precession by subtracting out a computed W' , the phase angle may be found from recognizing that the imaginary part of W_T must be multiplied by $\sin \eta$, via Equation IV-26, so that

$$\sin \eta \approx \frac{A_{\text{TRIV}}}{W_T/W_0} . \quad (\text{IV-29})$$

From Equation IV-25, we have for these terms

$$\begin{aligned}
 A_{\lambda}^{\lambda} 2^{\lambda} 1 = & [F(L_1 \ L_1+1 \ I_2 \ I_1) \langle L \rangle \langle L+1 \rangle^* \\
 & + F(L_1+1 \ L \ I_2 \ I_1) \langle L \rangle^* \langle L+1 \rangle \\
 & + F(L_1 \ L_1+2 \ I_2 \ I_1) \langle L \rangle \langle L+2 \rangle^* \\
 & + F(L_1+2 \ L_1 \ I_2 \ I_1) \langle L+2 \rangle^* \langle L \rangle \\
 & + F(L_1+1 \ L_1+2 \ I_2 \ I_1) \langle L_1+1 \ L_1+2 \rangle^* \\
 & + F(L_1+2 \ L_1+1 \ I_2 \ I_1) \langle L_1+2 \rangle \langle L_1+1 \rangle^*] \\
 & / (|L|^2 + |L+1|^2 + |L+2|^2) . \quad (IV-30)
 \end{aligned}$$

Again using the property of Equation IV-18, and assuming $\langle L+1 \rangle = |L_1| e^{i\eta}$, $\langle L+2 \rangle = |L+2| e^{i\bar{\eta}}$, we have after simplification and with the definitions $|\delta| = \frac{|L+1|}{|L|}$ and $|\bar{\delta}| = \frac{|L+2|}{|L|}$, the following result:

$$\begin{aligned}
 A_{\lambda}^{\lambda} 2^{\lambda} 1(\delta, \bar{\delta}) \Big|_{\substack{\text{T-odd} \\ \text{P-even}}} = & -2 \left[|\delta_1| F_{\lambda}^{\lambda} 2^{\lambda} 1(L_1 \ L_1+1 \ I_2 \ I_1) \sin \eta \right. \\
 & + |\delta_2| F_{\lambda}^{\lambda} 2^{\lambda} 1(L_1 \ L_1+2 \ I_2 \ I_1) \sin \bar{\eta} \\
 & + |\delta_1| |\delta_2| F_{\lambda}^{\lambda} 2^{\lambda} 1(L_1+1 \ L_1+2 \ I_2 \ I_1) (\sin \eta \cos \bar{\eta} - \\
 & \left. \sin \bar{\eta} \cos \eta) \right] / (|\delta_1|^2 + |\delta_2|^2) . \quad (IV-31)
 \end{aligned}$$

In the analysis of the ^{184}W TRIV experiment it was necessary to consider possible solutions involving two variables, η and $\bar{\eta}$, in one equation. The simplest possibilities are those for which either $\sin \eta = \eta$ or $\sin \bar{\eta} = 0$. The results (allowing for $\sin \eta = 0 \Rightarrow \eta = 0$ or π) are

Case I: $\sin \eta = 0$

$$A_{\lambda}^{\lambda_2 \lambda_1} \Big|_{\substack{\text{P-even} \\ \text{T-odd} \\ \sin \eta = 0}} = -2i|\bar{\delta}| \left[\frac{F(L_1 \ L_1+2 \ I_2 \ I_1) - \delta_1 F(L_1+1 \ L_1+2 \ I_2 \ I_1)}{1+|\delta_1|^2 + |\delta_2|^2} \right] \sin \bar{\eta} \quad (\text{IV-32})$$

and

Case II: $\sin \bar{\eta} = 0$;

$$A_{\lambda}^{\lambda_2 \lambda_1} \Big|_{\substack{\text{P-even} \\ \text{T-odd} \\ \sin \bar{\eta} = 0}} = 2i|\delta| \left[\frac{F_{\lambda}^{\lambda_2 \lambda_1}(L_1 \ L_1+1 \ I_2 \ I_1) + \delta_2 F_{\lambda}^{\lambda_2 \lambda_1}(L_1+1 \ L_1+2 \ I_2 \ I_1)}{1+|\delta_1|^2 + |\delta_2|^2} \right] \times \sin \eta \quad (\text{IV-33})$$

At this point we note that in Section VIII for ^{184}W , with $L = 1$, $L' = 2$, $L'' = 3$, we have let $\eta = \eta_{12}$ and $\bar{\eta} = \eta_{13}$.

To continue this general discussion of the "KSW" notation, the next terms in Equation IV-15 for $W(\theta_1, \theta_2, \phi)$ are the $A_{\lambda_2}(x_2)$, which are the ordinary angular distribution coefficients for the second radiation, as would be used to describe the directional correlations from unoriented sources (e.g. Fra 65). In the most general form they are written as

$$A_{\lambda}(X) = \left[\sum_{LL'} b_{\lambda}(LL'; X) F_{\lambda}(LL'I_3I_2) \langle I_3 || X_L || I_2 \rangle \langle I_3 || X_L || I_2 \rangle^* \right] \\ \times \left[\sum_L b_0(LL; X) \langle I_3 || X_L || I_2 \rangle \right]^{-1} \quad (IV-34)$$

where all the quantities have been defined following Eqn. IV-16.

The directional distribution coefficient for a mixed multipole πL and $\pi'L'$ gamma transition is

$$A_{\lambda}(\gamma) = [F_{\lambda}(LL'I_3I_2) + 2\delta F_{\lambda}(LL'I_3I_2) + \delta^2 F_{\lambda}(L'L'I_3I_2)] / (1 + \delta^2), \quad (IV-35)$$

as previously discussed.

P-odd, T-odd terms

These are the terms for which λ_1 and λ_2 are even, but λ is odd.

In this case Equation IV-16 becomes for doubly mixed cases with an additional parity violating component L_1^P , and where L and L' are in phase, with η the phase of $\langle L_1^P \rangle$ with respect to $\langle L_1 \rangle$

$$A_{\lambda}^{\lambda_2 \lambda_1} = \left[F_{\lambda}^{\lambda_2 \lambda_1}(L_1 L_1^P I_2 I_1) |L_1| |L_1^P| (\cos \eta - i \sin \eta) \right. \\ + F_{\lambda}^{\lambda_2 \lambda_1}(L_1 L_1 I_2 I_1) |L_1| |L_1^P| (\cos \eta + i \sin \eta) \\ + F_{\lambda}^{\lambda_2 \lambda_1}(L_1' L_1^P I_2 I_1) |L_1'| |L_1^P| (\cos \eta - i \sin \eta) \\ \left. + F_{\lambda}^{\lambda_2 \lambda_1}(L_1' L_1 I_2 I_1) |L_1'| |L_1^P| (\cos \eta + i \sin \eta) \right] \\ / (|L_1|^2 + |L_1'|^2 + |L_1^P|^2), \quad (IV-36)$$

which reduces to

$$A_{\lambda}^{\lambda_2 \lambda_1} = -2i \left[\frac{F_{\lambda}^{\lambda_2 \lambda_1 (L_1 L_1^P I_2 I_1)} + |\delta| F_{\lambda}^{\lambda_2 \lambda_1 (L_1 L_1^P I_2 I_1)}}{+|\delta|^2 + |\epsilon|^2} \right] \text{Im} \epsilon \quad (\text{IV-37})$$

We note from the preceding equation that if the values of an experimental parity-violating component mixing ratio is assumed to be the real part of ϵ , then the P-odd, T-odd terms are proportional to $|\epsilon| \cos \eta \times \tan \eta$, so that the sum of these terms in that case would provide a measure of $\tan \eta$, not $\sin \eta$, provided the effect of $|\epsilon|^2$ on the denominators were assumed small.

The $H_{\lambda \lambda_2 \lambda_1}$ function (geometric dependence)

Finally, the last factor in Equation IV-15 is the function $H_{\lambda \lambda_2 \lambda_1}(\theta_1 \theta_2 \phi)$ which includes the geometric dependence. This function is defined in Kra 73b and examples are given there for various geometries. We shall give here only the most general expression, as later presented in Kra 73d to include T-odd and T-even terms:

$$\begin{aligned} H_{\lambda_1 \lambda \lambda_2}(\theta_1 \theta_2 \phi) &= \sum_{q \geq 0} \begin{Bmatrix} 2 & \delta_{q0} \\ -2i \end{Bmatrix} \langle \lambda_1 \ 0 \ \lambda_q | \lambda_2 q \rangle (-1)^{\lambda} \\ &\times \left(\frac{2\lambda + 1}{2\lambda_2 + 1} \right)^{1/2} \left[\frac{(\lambda - q)! (\lambda_2 - q)!}{(\lambda + q)! (\lambda_2 + q)!} \right]^{1/2} \\ &\times P_{\lambda}^q(\cos \theta_1) P_{\lambda_2}^q(\cos \theta_2) \begin{Bmatrix} \cos q\phi \\ \sin q\phi \end{Bmatrix}. \end{aligned} \quad (\text{IV-38})$$

Of the two pairs of quantities in brackets, the upper member refers to cases in which $\lambda_1 + \lambda + \lambda_2$ is even, while the lower member is for odd values of the sum; the latter case is the one of interest for studies of time-reversal invariance. The P_λ^q are the associated Legendre polynomials.

The geometry used in each of the TRIV experiments of this dissertation was the NIN2 geometry of Krane, Steffen and Wheeler (Kra 73b) for which the two γ -rays are detected in the plane perpendicular to the polarization axis.

Detector geometry factors

The factors $Q_\lambda(X_1)$ and $Q_\lambda(X_2)$ are the detector geometry efficiency factors for the two γ -rays which are less than 1 in magnitude due to the shape and finite extent of typical detectors. From considerations of efficiency tensors, (Ros 53) one defines an integral for a cylindrical detector with axis along the direction of a gamma ray emanating from a point source, where β is the angle between the axis and a line from the source to a volume element in the detector by

$$J_K(\gamma) = \int P_K(\cos \beta) \left(1 - e^{-\tau(\gamma)\chi(\beta)}\right) \sin \beta d\beta \quad (\text{IV-39})$$

where $\tau(\gamma)$ is the linear absorption coefficient for the material from which the detector is constructed and $\chi(\beta)$ is the distance along the line from the source starting from the front face measured to a particular volume element. This integration is carried out over the path lengths

$\chi(\beta)$ throughout the active volume of the detector. The absorption coefficients $\tau(\gamma)$ are energy-dependent.

The attenuation coefficient $Q_K(\gamma)$ must generally be obtained via numerical integrations. Many publications are available tabulating $Q_K(\gamma)$ versus peak energy and source distance for various configurations of both NaI(Tl) detectors (Yat 65, Fer 65) and for coaxial drifted Ge(Li) detectors (Camp 69, Aun 73, Avi 72, Kra 72b). For applications not requiring high precision, interpolation from existing tables is usually sufficient.

CHAPTER V

TRIV EXPERIMENTS PERFORMED

Introduction

Searches for asymmetries indicative of violation of time-reversal invariance in nucleon-nucleon interactions were conducted by γ - γ directional correlation techniques with γ -rays of the isotopes ^{180}Hf , ^{175}Lu , ^{169}Tm , and ^{184}W . The magnitudes of phase angles between electromagnetic multipole components were in each case estimated, except in the case of the 922 keV radiation of ^{184}W , which is triply mixed, leading to only a relation between two phase angles.

The T-odd P-even, T-odd P-odd, etc. terms in the Krane, Steffen and Wheeler directional correlation function have been discussed in detail in Chapter IV, along with indications of how the phase angles in question are related to those various terms. We have shown how determinations of the relative phase angle between electromagnetic components gives an indication of TRIV. In addition, in the following discussions of specific experiments, it will be demonstrated how large hindrances in electromagnetic transition rates may lead to higher sensitivities in determinations of TRIV components of transition matrices otherwise not practically attainable.

Where feasible, first-order perturbation calculations based on recently presented arguments of Steyert and Krane (Stey 73) have been made to estimate magnitudes of possible Hamiltonians mixing states which could lead to the measured TRIV values.

Details of techniques and analysis are presented in the separate descriptions of the experiments for each set of isotopes.

Experimental Techniques

As described more fully in the following sections, polarizations of initial nuclear levels were obtained cryogenically, by taking advantage of large hyperfine magnetic fields obtained at impurity nuclei dissolved into magnetic materials (as described in general in Chapter III) coupling with the nuclear magnetic dipole moments. The ultralow temperatures required for appreciable polarization of initial nuclear spin states were obtained by the use of a ^3He - ^4He dilution refrigerator, the operating principles of which are discussed in Appendix A. Thermometry methods are discussed in Appendix B. The configuration and construction of the portion of the cryogenic apparatus used to hold and magnetically align the source is described in some detail in the discussion of the ^{180}Hf experiments which follows, as are discussed the functions of a detector and electronics system used to acquire the γ -ray counting rates. A somewhat more detailed discussion of the computer data acquisition system may be found in Appendix C.

TRIV Tests on Components of the 501-keV γ -Ray of $^{180\text{m}}\text{Hf}$

This section describes the first reported tests of T-violation and simultaneous violation of P- and T-symmetry in nuclear γ -decay using the enhancement concept. These studies were undertaken on the 501-keV transition of ^{180}Hf , in which the enhancement consideration gives rise to the largest observable case of irregular parity multi-

poles yet observed (Jen 70, Lip 71, Kra 71a). Two reports of the T-violation experimental work have been published recently (Mur 73, Kra 74b).

The $^{180}\text{Hf}^m$ decay scheme

The decay scheme of $^{180}\text{Hf}^m$ is shown in Fig. V-1 (Led 67). The 5.5-h isomer is assigned as an intrinsic excitation having 8 units of spin projection on the nuclear symmetry axis ($K = 8$); the large value of the magnetic moment of this state ($\mu = 8.6 \pm 1.0 \mu$ [Kor 71]) identifies it as a 2-proton configuration. The isomer decays to the 8^+ and 6^+ rotational states of the ground-state band ($K = 0$) with total branching intensities 86% and 14%, respectively. The transition of interest for the present work is the 501-keV γ -ray, which is of mixed M2 + E3 multipolarity. The amplitude mixing ratio E3/M2 has been measured to be +5.3 (Kra 71a, Bod 61). The K-selection rule for γ -transitions ($L > \Delta K$) results in the strong retardation of the 501-keV γ -ray multipoles relative to single-particle Weisskopf estimates; the M2 and E3 transition probabilities have hindrances relative to Weisskopf estimates of $H_W(M2) = 1.3 \times 10^{14}$ and $H_W(E3) = 2.0 \times 10^9$ (the γ -ray matrix elements are reduced relative to Weisskopf estimates by the respective square roots of these numbers).

The unusually large hindrances associated with the 501-keV transition (or rather with its "regular" P- and T-conserving multipoles) may aid the observation of "irregular" P- and T-violating multipoles in the radiation field if they are not also similarly hindered. Previous studies have indeed confirmed the presence of a P-irregular E2 multipole with an E2/M2 amplitude mixing ratio of -0.04 (Jen 70, Lip 71, Kra

Fig. V-1. Simplified decay scheme of $^{180}\text{Hf}^m$.

71a). Although this E2 multipole produces quite a substantial laboratory effect (a 1.6% 0°-180° asymmetry in the decay of polarized $^{180}\text{Hf}^m$), the E2 multipole is also rather strongly hindered, with $H_w(E2) = 2 \times 10^9$. However, here most of the hindrance is associated with the weakness (relative strength about 10^{-6} or 10^{-7}) of the parity violating interaction. Thus, nuclear structure effects provide only an additional hindrance of 2×10^2 or 2×10^3 in this case. It is likely that the weak interaction force responsible for the P-irregular multipoles is similar in character to that of the normal strong nuclear interaction and, thus, it is not too surprising that the irregular γ -transition is hindered by intrinsic nuclear structure effects. However, T-violating interactions are probably not at all similar to the regular strong or weak interaction operators and thus it is extremely unlikely that the T-violating transition intensities will be similarly hindered. Thus ^{180}Hf may provide a test of T-violation even more sensitive than that of P-violation.

In the present work we observe the angular correlation from the polarized initial level at 1142 keV between the mixed E3 + M2 501-keV γ -ray and either the subsequent pure E2 332- or 215-keV γ -rays. Since the sequence of transitions depopulating the 6^+ level constitutes a basic sequence, the 332- and 215-keV transitions have identical angular correlations (neglecting perturbations) in coincidence with the 501-keV transition.

Experimental details

Sample preparation. As in the measurements of parity nonconservation in the γ -decay of $^{180}\text{Hf}^m$ (Kra 71a), the Hf was arc-melted with Zr and Fe to form the cubic ferromagnet $(\text{Hf}_{0.05}\text{Zr}_{0.95})\text{Fe}_2$. Crystal bar Hf enriched in ^{179}Hf was used (87% ^{179}Hf , 8.5% ^{180}Hf , 3.3% ^{178}Hf , 1% ^{177}Hf , 0.2% ^{176}Hf) in order to minimize contamination with ^{181}Hf decay gamma rays. In ZrFe_2 , Hf experiences a magnetic hyperfine field of -200 ± 20 kOe, resulting in an energy difference $\Delta = \mu H/I$ between the adjacent magnetic hyperfine levels (corresponding to different values of I_z) of the 8^- state of -7.9 ± 0.5 mK. At dilution refrigerator temperatures of 20 to 35 mK, polarizations approaching unity are achieved. The nuclei align in a direction which is opposite to that of the $(\text{Hf}, \text{Zr})\text{Fe}_2$ polarization. The $(\text{Hf}, \text{Zr})\text{Fe}_2$ polarization is in turn parallel to the direction of the modest magnetic field applied to polarize the ferromagnet (in these experiments, a generous kOe was used). Thus, the nuclear polarization is opposite to the applied magnetic field.

For ease of polarization, and in order to minimize γ -ray absorption, the samples (after annealing at 950°C for 16 h) were cut into 7 mm x 1 mm x 1 mm needles and were activated in the Los Alamos Omega West Reactor, receiving total integrated fluxes of 4×10^{15} neutrons/cm². They were mounted and polarized vertically in the cryostat, while the detectors observed γ -rays emitted normal to the long vertical sample axis.

Cryogenic apparatus. Figure V-2 shows the cryogenic assembly.

The sample is soldered with high purity indium onto a copper fin, the upper end of which is threaded. This is inserted through the bottom of the refrigerator vacuum can and screws into a split copper receptacle which is in turn indium-soldered to the cold finger coming down from the mixing chamber of the ^3He - ^4He dilution refrigerator. Adequate thermal contact is provided from the sample to the refrigerator (1) through the indium solder which is driven normal by the high applied field (2), through the copper-to-copper screw joint which is very tight at low temperatures because of contraction in the nylon collar, and finally (3) through the cold finger. The cold finger is fabricated from 15 0.04-cm diam high purity annealed copper wires, spot-welded to the mixing chamber which contains a sintered copper disk for thermal contact to the mixture. Electrical resistance at liquid N_2 temperature across this screw joint was measured as $7\ \mu\Omega$. This design is predicated on requirements of good thermal contact, minimization of eddy current heating due to the periodic magnetic field reversals, and convenience of sample changing. The time involved in inserting the sample, soldering the 1/2 in. cap on the bottom of the refrigerator vacuum bulb, replacing the He and N_2 dewars, and cooling to 35 mK is about 6 h. The operating temperature of the refrigerator mixing chamber in the absence of any heat load is 14 mK.

Also schematically shown in Fig. V-2 are the vertical superconducting magnets which were used to provide the vertical polarizing field on the sample. They were 2 cm in diam with 1.8 cm separating

Fig. V-2. Schematic representation of interior of cryostat, showing source mounting arrangement, polarizing coils, magnetic shielding, and a portion of the cryogenics. Shown are vertical (a) and horizontal (b) cross-sections.

the two coils of 1000 turns each. The field at the sample was 0.54 kOe per ampere. The horizontal coils, 2 cm in diameter and 2.5 cm apart had 500 turns each and provided 0.1 kOe per ampere. Both sets of coils were fabricated from 0.005 in diam Suprcon type T48B Formvar insulated copper-clad NbTi wire (Suprcon, Inc., Natick, Mass.). The current leads from the magnet power supply were 20 mil diam copper-clad NbTi wire.

To reverse the nuclear polarization, the magnetic field is rotated by gradually turning on the horizontal coils while the vertical coils are gradually being turned off. When the vertical coil current is zero, the leads are reversed via a double pole-double throw relay, and the vertical coils are again gradually energized while the 90° coils are gradually turned off. Each 90° rotation takes 20 sec. Because the magnetic field is rotated rather than reversed, magnetic hysteresis energy dissipation is much smaller. Also the nuclei are able to follow the slowly rotating field, and no nuclear relaxation is required before the nuclei assume their new opposite polarization (nuclear spin lattice relaxation times at these temperatures are typically many seconds). Many of these cryogenic considerations are discussed in further detail in Kra 71c.

Electronics. Data are accumulated in and the experiment is controlled by the Nova (Data General Corp., Southboro, Mass., 01722) mini-computer-based data acquisition system (Han 74) which is shown in Fig. V-3. With the aid of the block diagram in Fig. V-3 and the coincidence and singles pulse height spectra shown in Fig. V-4, we may review the

Fig. V-3. Block diagram of data acquisition system.

Fig. V-4. Typical $^{180}\text{Hf}^m$ singles and coincidence (w/501-keV) γ -ray spectra from 7.6 x 7.6 cm NaI(Tl) detectors.

types of data which are accumulated by the system. The detector pulses are digitized by the ADC's and through the data channel the appropriate memory location in the computer is incremented. However, in order to keep the ADC dead time small ($\sim 10\%$) only one in ten singles pulses is stored (the ADC's are operated such as to require a logic pulse into the coincidence input to digitize the pulse). But if the other detector registers a pulse from the particular peak of interest (501-keV) in coincidence (resolving time $2\tau = 0.8 \mu\text{sec}$) with this pulse, the pulse is always digitized and it is stored in a different memory location to form the coincidence spectrum.

The computer records the number of seconds that the ADC has been alive for the preset real-time counting period. It also records the number of times that the "particular peak" (501-keV) timing SCA's have been triggered. At the end of the preset counting period, and while the nuclear polarization is being rotated, the computer calculates the required counting rates. Knowing the approximate position of a peak of interest, it will calculate the centroid to the nearest 1/10 channel and add up the counts in a preset number of channels above and below that center. It will also add 1/2 the number of counts in a preset number of channels further away from the center (this avoids a sharp cutoff, and the arbitrariness of giving one channel full weight and the one next to it zero weight). The total number of counts calculated in this way is then divided by the live time to form the counting rate. Backgrounds in a particular region of the spectrum are also integrated. For four of the peaks the computer calculates at the end of a series for field-up and for field-down runs the exact average center of the peak to the nearest 1/100

of a channel. Any systematic gain changes with field direction would be readily apparent from these numbers. With the magnetic field directed upward, for example, data are accumulated for a preset time, typically 5 to 20 min, following which the computer turns off the ADC's and the monitor scalers, sends out a magnet power supply motor control command to gradually turn on the horizontal magnets (magnet power supply A in Fig. V-3), outputs the requested data on the tape, reverses the polarity switch, and turns supply A off and Supply B (vertical coils) back on, so that current now flows in the opposite direction. This process continues automatically until manually stopped. After the third such computer period, the computer compares the results of a particular counting period, for example, the second period when the field was pointed downward, with the average counting rate of the previous and following counting period when the field was directed upward. It outputs, on paper tape, the difference and the square of this difference. As the data series progresses, the differences and their squares for the field-down Δ_D (and similarly for the field-up) periods are summed as they are accumulated. When the series is manually terminated after n such Δ_D 's have been obtained, the computer calculates and outputs the raw asymmetry A_r , the average difference in a particular counting rate for field up or down divided by twice the mean counting rate C .

$$A_r = \frac{\sum \Delta_D}{2nC} \quad (V-1)$$

The computer also prints out the experimental uncertainty in A_r , δ_r .

$$\delta_r = \frac{\sqrt{3/2}}{\sqrt{Cn}} \quad (V-2)$$

The $\sqrt{3/2}$ comes from the fact that each Δ_D is the difference between one number and the average of two other numbers with the same uncertainty which are counted twice in the sequence. The computer also puts out the normalized χ^2 for the data

$$\chi^2 = \sum (\Delta_D - \bar{\Delta}_D)^2 \tau^2 / [3/2 C \tau (n - 1)]. \quad (V-3)$$

The τ^2 in the numerator makes the numerator the sum of the squares of the fluctuations in the counts, and the denominator is the expected squared fluctuation in each Δ_D times $n - 1$ (required to make the expected $\chi^2 = 1$). Details of the computer interface system are discussed further in Appendix C.

Detector configuration. For this experiment two 7.6 cm x 7.6 cm NaI(Tl) detectors mounted on the dewar were used to provide high photo peak efficiency, ϵ , for the coincidence experiments. The upper limit on the source strength and counting rate was imposed by detector pulse pileup considerations rather than by cryogenic or coincidence true-to-accidental considerations.

The 135° interdetector angle was chosen so as to maximize the magnitude of the scalar $(\vec{i} \cdot \vec{k} \times \vec{k}')(\vec{k} \cdot \vec{k}')$ (at 45° the detectors are too close together to fit conveniently, and detector-to-detector γ -scattering could be a problem). The geometry is shown schematically in Fig. V-5. The detector distance is a compromise between the requirements for a high-coincidence counting rate (which goes like product of the two detector efficiencies) and the need to keep the solid angle correction factor Q_2 small (the T-odd effect is decreased

Fig. V-5. Simplified schematic of experimental geometry used for the tests for P-even, T-odd TRIV asymmetries.

by the product of the two detector Q_2 's). When the source was strong, the detector distance was 7-9 cm, ϵ was about 0.03, and Q_2 was about 0.85 for γ 's in the range of interest. As the source decayed and the detectors were moved to within 4.2 cm from the source, ϵ grew to about 0.08 and Q_2 fell to about 0.7.

The superconducting coils provide a 5 kOe field at the sample, and to prevent this fringing field from causing a systematic gain shift, sensitive to field direction, which would provide a false asymmetry in the counting rate of the 501-keV γ -ray in the timing SCA, extensive shielding was required. First, the coils were surrounded by a soft-iron cylindrical shell 8 cm long, 5 cm in diameter, with 3 mm walls. The ends of the cylinder were almost closed as well (see Fig. V-2). Even with shielding wrapped around the outside of the dewar, and with standard magnetic shields on the detectors, field-sensitive gain shifts and SCA counting rate changes were observed. This problem was solved by the addition of a thin (0.005 cm) lead foil enclosure around the iron cylinder. This superconducting shield, soldered together with superconducting indium, prevented the escape of any stray field which would penetrate the iron except at the 2 cm diam openings at the ends where the refrigerator sample tube entered. No stray-fields in excess of 0.01 Oe were measured outside the dewar. With the lead shielding in place, no gain or count rate changes correlated with field direction were observed to precisions of 1/5000 and 1/3000 respectively.

Evaluation of experimental method

This TRIV test is designed to be a "difference" experiment, look-

ing for differences in counting rates with the nuclear spins pointed up compared to when they are pointed down. Obviously this simple method eliminates some sources of error, but can cause some problems as well. The prime advantage of this method is that the experiment looks for counting rate differences with no changes in the source or detector positions, and no changes in the electronics. The difference technique not only searched for small asymmetries upon field reversal, but also accumulated simultaneous gated coincidence spectra accepting the 501-keV γ -ray in either detector 1 or detector 2 (see Fig. V-5). This symmetric detector method is equivalent to observing the correlation at ϕ and $2\pi-\phi$, and referring to Eqn. IV-38 we see that $W_O(\phi) = W_O(2\pi-\phi)$, while $W_T(\phi) = -W_T(2\pi-\phi)$. A similar transformation $\phi \rightarrow 2\pi-\phi$ results from reversing the field direction (dashed line in Fig. V-5). The source and detectors are rigidly mounted so as not to be moved by magnetic forces (which should not change with field reversal in any event, since the force is proportional to H^2 on non-permanent magnets). Elaborate shielding previously described prevents a magnetic field influence on the experiment. Effects of drifts in the electronics are averaged out by frequent field direction changes (every 5 min at high count rates, every 20 min at low count rates). Most sources of systematic error can be examined with the source at or above 1 K where there would be no nuclear polarization and therefore no T-odd effect. Tests for systematic error in the higher counting rate parity violation tests carried out with this equipment show no problems above the 10^{-4} level.

There are, however, some cryogenic problems associated with this difference experiment technique. Each field rotation puts a few ergs

of heat into the sample, primarily through magnetic hysteresis heating, and tests have shown that a continuous rotation of 0.05 Hz causes a 5 mK increase in source temperature. A field reversal instead of rotation would be intolerable. The twisted high purity (99.999%) copper wires in the refrigerator system can conduct 0.6 erg/sec for $\Delta T = 1$ mK (there are 15 wires, 0.04 cm in diameter).

Another aspect of the rotating field method is the problem of nuclear phase lags between the field rotation and the nuclear rotation; this is found to be 1 sec for ^{60}Co in Fe compared to 1 min for the nuclear spin lattice relaxation times. Thus, the nuclei are found to follow the nuclear rotation very nearly adiabatically (Kra 71c).

Results

The raw and corrected experimentally measured asymmetries for various singles and coincidence peaks are listed in Tables V-1 and V-2, after being averaged appropriately for both analyzer systems. Several experimental corrections were made to the 215 keV and 332 keV coincidence peaks as follows, in decreasing order of importance:

- (1) The effect of a small systematic residual asymmetry, as measured in the 501-keV singles peak and in the single-channel-analyzers set for that γ -ray, was removed by an additive factor determined to be $(1.4 \pm 0.9) \times 10^{-4}$ for the warm data and $(1.7 \pm 0.5) \times 10^{-4}$ for the cold. However, as discussed above, these factors were first multiplied by a factor of 1.4 to allow for a possible parity-violating contribution from misalignment of the source and other errors. We choose a factor less than the value 1.8 previously discussed, since previously the residual effect had been assumed to be purely from misalignment,

Table V-1. Some typical raw measured asymmetries
for the ^{180}Hf TRIV experiment

E_{γ} (keV)	Singles Peak Asymmetries ($\times 10^4$)				Coin(w/501) Asymmetries ($\times 10^4$)		
	215	332	444	501	215	332	444
$T = 20-35 \text{ mK}$	0.9 ± 0.5	0.3 ± 0.6	0.6 ± 0.6	0.1 ± 0.9	7.6 ± 4.3	0.0 ± 4.8	0.9 ± 2.3
$T > 2 \text{ K}$	0.9 ± 0.8	-0.6 ± 0.9	1.5 ± 1.0	2.6 ± 1.7	4.7 ± 5.9	-1.9 ± 6.3	2.5 ± 5.9

Table V-2. Corrected experimental asymmetries
for the ^{180}Hf TRIV experiment

E_{γ} (keV)	Coincidence Peak (w/501-keV) Asymmetries ($\times 10^4$)	
	215	332
$T = 20 - 35 \text{ mK}$	$+12.0 \pm 5.5$	$+2.8 \pm 6.1$
$T > 2 \text{ K}$	$+ 9.1 \pm 7.6$	$+0.9 \pm 7.9$

whereas we feel that misalignment is unlikely to be the sole contributor to the residual asymmetry. (2) A 15% upward correction was made to compensate for a representative true-to-chance coincidence ratio of 6.5. This ratio was found by statistically weighing periodically monitored true-to-chance measurements taken during the experiment. (3) The asymmetry in a background region lying just above the 501-keV peak in the coincidence spectrum was measured and an appropriate correction, less than 10% upward for each of the asymmetries of interest, was performed.

The 215-, 440-, and 332-keV singles peak asymmetries served as monitors of systematic problems.

After these systematic corrections were performed, the warm and cold asymmetries used in the remaining discussion were as listed in Table V-2.

As discussed previously, a non-negligible but uncertain amount of precession effect is expected to appear in the 501-215 keV asymmetry. In order to remove most of this effect, the warm asymmetry was subtracted from the cold to obtain the resultant value $(2.9 \pm 9.4) \times 10^{-4}$. This value was then averaged with the low temperature asymmetry for the 501-332-keV correlation to obtain the overall TRIV asymmetry

$A = W_T/W_O$ from Eqn. IV-29

$$A = (+2.8 \pm 5.1) \times 10^{-4}. \quad (V-4)$$

With $W_O = 1.297$ and with $W_T = +0.0076 \sin \eta$ resulting from a computer calculation of the directional correlation further, using a program written by K. S. Krane, the phase is given by

$$\sin \eta = +0.048 \pm 0.087.$$

The significance of this result will be discussed further in the last section of this chapter.

The phase angle η which characterizes the T-violation is given by (assuming $\sin \eta$ to be small)

$$\sin \eta = \frac{1}{|\delta|} \operatorname{Im}(\delta) \approx \frac{\operatorname{Im}\langle L+1 \rangle}{|\langle L+1 \rangle|} - \frac{\operatorname{Im}\langle L \rangle}{|\langle L \rangle|} \quad (V-5)$$

In order to obtain meaningful relative comparisons of the matrix elements it is necessary to normalize with respect to density of final state effects (essentially energy dependence), effects of the radiation field at the origin (L-dependence), and nuclear size effects (A-dependence). This can be most conveniently done by expressing the matrix elements in Weisskopf units, denoted by the subscript W. We then obtain, choosing $\langle L+1 \rangle$ to be real,

$$|\operatorname{Im}\langle L \rangle_W| = |\langle L \rangle \sin \eta| / \langle L \rangle_W = |\sin \eta| [K_W(L)]^{-1/2} \quad (V-6)$$

where K_W denotes the Weisskopf hindrance of the transition probability. Thus $|\operatorname{Im}\langle M2 \rangle_W| = (4.2 \pm 7.5) \times 10^{-9}$. A similar expression may be obtained for $\operatorname{Im}\langle L+1 \rangle_W$, assuming $\langle L \rangle$ to be real. We obtain $|\operatorname{Im}\langle E3 \rangle_W| = (1.1 \pm 1.9) \times 10^{-6}$. The results for the out-of-phase components for this and previous experiments are compared in the concluding section of this chapter.

Significance with respect to T-odd potential

The question remains as to whether this small upper limit of the T-odd γ -ray matrix element results from a hindrance of the T-odd operators in a manner similar to that of the T-even operators, or

whether it results from the intrinsically small strength of the T-odd Hamiltonian. As discussed in the previous section, nuclear structure effects probably do not strongly hinder the P-odd matrix element. Since the P-odd operators are likely to be of a two-body nature, as are the P-even operators, only a modest net enhancement is obtained. However, the T-odd nucleon-nucleon interactions may be of a three-body character (Cle 71, Cle 73), and at very least are momentum-dependent, so that transition operators associated with such interactions are unlikely to be hindered in a manner similar to those associated with the T-even interactions. Similarly, if T-violation arises in the electromagnetic field itself (as opposed to in the nuclear Hamiltonian), the T-odd transition operators will be two-body, while normal transition operators are one-body. Again the T-odd matrix elements may not be hindered as much as are the T-even matrix elements. We therefore conclude that, in the absence of detailed calculations of T-odd matrix elements, the hindrance concept is expected to be at least as effective in the T-odd case as in the P-odd case, and that the above-deduced out-of-phase matrix elements are probably not strongly hindered by nuclear-structure effects, but rather represent a measure of the strength of the T-odd potential.

A first-order perturbation estimation of a possible TRIV mixing Hamiltonian component, producing TRIV due to mixing of an out-of-phase $K = 2$ octupole level $| \tilde{8}^- \rangle$ with the 8^- [8] 1142-keV level has been presented by Krane (Kra 74b), resulting in

$$\langle \tilde{8}^- | H_{\text{odd}} | 8^- \rangle \approx 0.4 \pm 0.7 \text{ eV.}$$

A test of simultaneous P- and T-
violation in the E2/M2 mixing ratio

Parity violation associated with the interaction of nucleons within the nucleus is well established. The largest known such effect occurs in $^{180}\text{Hf}^m$ where one sees a 1.7% parity violating forward-backward asymmetry of γ -rays emitted from a polarized nucleus (Kra 71a) as well as a 0.2% circular polarization of γ -rays emitted by an unpolarized nucleus (Fis 73, Gar 73).

This unusual phenomenon occurs in the case of the 501-keV γ -transition of the 5.5h metastable state of $^{180}\text{Hf}^m$ through the mixing of a parity violating $\tilde{\text{E2}}$ contribution into the parity conserving M2-E3 mixed γ -radiation. Time reversal invariance requires (Llo 51) that the relative phase angles of all three components must be either 0 or π ; that is, in Figure V-6 there must be no imaginary out-of-phase component of any of the three multipolarities relative to the others. Previous work (Mur 73) has shown that the relative M2-E3 phase angle, η , (see Fig. V-6 is given by $\sin \eta = +0.048 \pm 0.087$. It has also been shown (Kra 71b) that the real part of the anomalously large $\tilde{\text{E2}}$ component is -0.038 ± 0.004 of the M2 component. However, no measurement exists of either the magnitude of the anomalous $\tilde{\text{E2}}$ component or the imaginary out-of-phase $\tilde{\text{E2}}$ component.

Because of the unusually large symmetry breaking in this γ -ray and the fact that no test of simultaneous parity and time-reversal invariance violation (P-T) had previously been done in nuclear-electromagnetic transitions such an experiment is attractive. In addition, using the theory of Zachariasen and Zweig (Zac 65), Szymanski (Szy 68) has shown that a very sizeable P-T violation relative to the P-violation is possible. Since the nature of such symmetry-breaking forces

Fig. V-6. Phase angle relationships among the $M2$, $\tilde{E}2$, and $E3$ components of the 501-keV γ -ray from $^{180}\text{Hf}^m$.

is unclear (Cab 64), such an experiment could shed light on them.

On the other hand there are several reasons, both experimental and theoretical, to expect that the imaginary part of the $E2$ transition should be unmeasurably small. First, time reversal violation in the parity conserving parts of nuclear γ -ray transitions has not yet been seen. Second, the very small upper limit of 10^{-23} e-cm has been placed on the (P-T violating) neutron electric dipole moment (Dre 73). Third, since the electromagnetic and strong interaction are thought to conserve parity, simultaneous P-T violation is expected only at the level of milli-weak theories. Here, the amount of simultaneous P-T violation is the product of the P and T violations, each of which is extremely small or zero (Bli 73a) (except for P in this peculiar transition), although in certain models it may be larger.

Methods

Simultaneous P-T violation in the 501-keV γ transition would appear (Szy 68) in lowest order as a non-zero value for the scalar $\langle (\vec{I} \cdot \vec{k}_1 \times \vec{k}_2) (\vec{I} \cdot \vec{k}_2) \rangle$ if photon polarization is not observed, where I is the initial nuclear polarization, $\vec{k}_1$ is the momentum of the 501-keV γ -ray and $\vec{k}_2$ is the momentum of either the 332-keV or the 215-keV γ -rays which are emitted in succession following the 501-keV transition (they essentially serve as an analyzer of the final $6+$ state left after the 501-keV transition). Figure V-7 shows the geometrical arrangement which maximizes this scalar quantity. Detectors 1 (D1) and 2 (D2) in themselves constitute a complete experiment, looking for differences in the coincidence counting rates depending on whether $\vec{I}$ is along $\vec{I}$ or $\vec{I}'$ or $\vec{I}''$, (i.e., with angle $\theta_2 = -\pi/4$,

Fig. V-7. Geometry and detector configuration for measuring the P-odd, T-odd(out of phase) component of the E2 transition matrix in the 501-keV radiation from $^{180}\text{Hf}^m$. The detectors are in the horizontal plane, I , I' , and I'' lie in the vertical y - z plane. Two simultaneous experiments were carried out by measuring coincidences between detectors 1&2 as well as 2&3 as discussed in the text.

$-\pi/4$, or $+\pi/4$ respectively in the convention of Krane, Steffen and Wheeler (Kra 73b). However, the difference experiment can be freed of some systematic errors if the effect in $D_2 - D_1$ is compared with $D_2 - D_3$. A true P-T violation would, for example, cause the $D_2 - D_1$ rate to increase and the $D_2 - D_3$ rate to decrease while many systematic problems would affect both $D_2 - D_1$ and $D_2 - D_3$ in the same way.

In order to achieve nuclear polarization, hafnium enriched to 87% ^{179}Hf was arc-melted in ferromagnetic ZrFe_2 to form $(\text{Zr}_{0.95}\text{Hf}_{0.05})\text{Fe}_2$. The sample was cut into disc-shaped pieces which were irradiated with thermal neutrons. Individual radioactive samples 0.5 mm thick by 6 mm diameter were attached to a $^3\text{He} - ^4\text{He}$ dilution refrigerator with the disk lying in the y-z vertical plane (See Fig. V-2), and cooled to 16-30 mK. Two pairs of horizontal and vertical superconducting Helmholtz coils produced a resultant external field of 4kOe along $-\hat{i}$, $-\hat{i}'$, or $-\hat{i}''$ to polarize the ferromagnetic host and thus the $^{180}\text{Hf}^m$ [the hyperfine splitting of $^{180}\text{Hf}^m$ in ZrFe_2 is -7.9 ± 0.5 mK (Kra 71a)]. The results reported are from ten separate sources, each providing about 20 hours of useful data before becoming too weak. Temperatures were determined from measurements of the asymmetry of the 332-keV γ -ray from Hf and, in some cases, of the 835-keV γ -ray from an auxiliary source consisting of ^{54}Mn dissolved in an iron foil which was indium-soldered together with the Hf source (see Appendix B).

Three 7.6 cm x 7.6 cm NaI(Tl) detectors were typically positioned at initial distances of 7 cm from the source, and moved in to 4.25 cm as the source decayed. The average source activity at the beginning of "cold" data accumulation was 2.5 μCi , giving coincidence rates of 12/sec.

For detector 2 a full singles and two coincidence spectra were recorded; the coincidence spectrum was gated ($2\tau = 0.25 \mu\text{sec}$) by a γ_1 pulse in the other detectors. The data were accumulated and recorded by a minicomputer-based data acquisition system (Han 74). After each counting interval of 10 minutes the magnetic field was rotated through 90° slowly by reversing the field in either the horizontal or vertical coils. This cycle was continuously repeated for several hours.

Details concerning magnetic shielding design and effectiveness as well as the electronics were described earlier in this chapter.

The correlation may be represented by the expression $W = W_0 + W_{PT}$, where W_0 , the correlation function of the two γ -rays in the absence of P-T violation, depends only on the alignment ($B_2, B_4, \dots$) of the state I and not on the polarization ($B_1, B_3, \dots$); W_{PT} , the P-T violating term (see Equation IV-37), also does not depend on polarization and is maximized for the presently employed geometry $\vec{k} \perp \vec{I}$, $\vec{k} \perp \vec{k}'$, and $(\vec{I}, \vec{k}') = \pm \pi/4$ or $-3\pi/4$.

By rotating the direction of nuclear polarization by $\pi/2$ we search for coincidence asymmetries of the form $A = [W(\theta_2^a) - W(\theta_2^b)] / [W(\theta_2^b)] \approx W_{PT}/W_0$, assuming $W_{PT} \ll W_0$. $W(\theta_2^a)$ and $W(\theta_2^b)$ indicate the measured correlation with θ_2^a and θ_2^b equal to $-3\pi/4$ and $-\pi/4$ (or $\pi/4$ and $-\pi/4$), respectively, with $\theta_1 = \phi = \pi/2$.

For the $\gamma_1 - \gamma_2$ (and also $\gamma_1 - \gamma_3$) correlation, we obtain $W_0(T = 20 \text{ mK}) = 1.123$ for our geometry and

$$\frac{W_{PT}}{W_0} = -0.00164 B_2 Q_2 (\gamma_1) Q_2 (\gamma_2) \frac{\Im \frac{E_2}{E_2}}{\Re \frac{E_2}{E_2}} \quad (V-7)$$

Results

The second moment of the alignment, B_2 , averaged 0.68 for the cold experiments while the average $Q_2(\gamma_1) Q_2(\gamma_2)$ was about 0.6. Each data series of several hours was corrected individually for B_2 , Q_2 's, true-to-chance ratio (10:1), SCA (gating rate) asymmetry, background asymmetry (peak: Compton background generally 10:1). The results are summarized in Table V-3.

Because of the disconcerting singles asymmetry, a_s , seen in some of the data of Table V-3, tests were made using the 113-208 keV cascade of ^{177}Hf (daughter of ^{177}Lu) in ZrFe_2 for source temperatures, $4\text{K} \leq T \leq 77\text{K}$. The non-zero value of a_s is associated with magnetic field induced source motion - a problem which does not occur in a T or P experiment where the field is simply reversed (forces on a soft ferromagnet like ZrFe_2 do not change when a magnetic field is reversed).

Since the quantity of interest is the $D_2 - D_1$ asymmetry minus the $D_2 - D_3$ asymmetry, the singles asymmetry in D_2 should not influence the results. In fact, for ^{177}Hf with $|a_s(208 \text{ keV})| = (241 \pm 3) \times 10^{-4}$, $A/Q_2(\gamma_1)Q_2(\gamma_2)$ corrected in the same way as α in Table V-3 was $(6.9 \pm 12) \times 10^{-4}$. After mechanical source stabilization which reduced $|a_s(208 \text{ keV})|$ to $(1.2 \pm 1.4) \times 10^{-4}$, $A/Q_2(\gamma_1)Q_2(\gamma_2)$ corrected as before was $(4.0 \pm 8.3) \times 10^{-4}$. The warm data taken with $^{180}\text{Hf}^m$ in ZrFe_2 sources shows similar results, but with larger statistical uncertainties.

The average P-T violating γ -ray asymmetry from all the $^{180}\text{Hf}^m$ results is

$$\bar{\alpha} = +(11.3 \pm 9.4) \times 10^{-4}$$

Table V-3. Summary of P-T results on ^{180}Hf

NORMALIZED SINGLES ASYMMETRY						NORMALIZED COINCIDENCE ASYMMETRY		
$a_s = A/Q_2(\gamma)$					$a_b^{(a)} = \frac{A}{Q_2(\gamma_1)Q_2(\text{BG})}$	$\gamma_1 = 501 \text{ keV}$ $a = A/B_2Q_2(\gamma_1)Q_2(\gamma_2)$		
θ_2^a	θ_2^b	$a_s(\text{D2})$		$a_s(\text{SCA-D3})$ $-a_s(\text{SCA-D1})$		$a(\text{D3-D2}) - a(\text{D1-D2})$		Corrected Avg. Coincidence Asymmetry,
		215 keV	332 keV	501 keV		$\gamma_2 = 215 \text{ keV}$	$\gamma_2 = 332 \text{ keV}$	
$-3\pi/4$	$-\pi/4$	$+21.6 \pm 9.4$	$+89 \pm 16$	$+1.9 \pm 1.6$	$+24 \pm 32$	$+23 \pm 9.2$	$+14 \pm 11$	$+17.4 \pm 10.1$
$\frac{\pi}{4}$	$-\frac{\pi}{4}$	-5.4 ± 19	$+44 \pm 21$	-3.4 ± 1.1	-123 ± 63	-17 ± 23	-86 ± 29	-26.2 ± 25.3
Result $a = +11.3 \pm 9.4$								

(a)

BG - Background region, 400-450 keV

which from Eqn. V-7 gives

$$\frac{\text{Im } \overset{\sim}{E2}}{\text{Re } \overset{\sim}{E2}} = -0.69 \pm 0.57$$

This establishes an upper limit for $\text{Im } \overset{\sim}{E2}$ of $-0.04/5.3$, or 1% of the transition amplitude.

Hence, in spite of the anomalous breaking of spatial symmetry in this γ -transition, no definite deviation from the requirement of temporal symmetry is seen in either M2, E3, or $\overset{\sim}{E2}$ components.

These results have been accepted for publication (Mur 74a).

γ -Rays of ^{169}Tm and ^{175}Lu

General

In the preceding experiment it was found, by performing a search for asymmetries in γ - γ correlations in $^{180}\text{Hf}^m$ (provided certain assumptions were made about the source of a time-reversal-invariance violating (TRIV) potential being three-body or otherwise different from the normal potential giving rise to electromagnetic transitions) that TRIV in the highly hindered 501-keV γ -ray of that isotope could be ruled out to a sensitivity of 10^{-6} and 10^{-9} in the E3 and M2 components, respectively.

The possibility of enhancement of a TRIV test due to hindrance (Stey 73) led to a similar test on the 198-110-keV and 177-131-keV γ - γ cascades in ^{169}Tm and on the 283-114-keV cascade in ^{175}Lu . ^{175}Lu is a β -decay daughter of ^{175}Yb while ^{169}Tm results from electron capture in ^{169}Yb . The Weisskopf hindrances of the 198-, 177-, and 283-keV radiation components are as high as 10^6 . Figures V-8 and V-9 show

Fig. v-8. Simplified decay scheme of ^{169}Yb , showing the cascades $I_1 \xrightarrow{\gamma_1} I_2 \xrightarrow{\gamma_2} I_3$ and $I_1' \xrightarrow{\gamma_1'} I_2' \xrightarrow{\gamma_2'} I_3'$ from ^{169}Tm studied.

Fig. V-9. Simplified decay scheme of ^{175}Yb , showing the cascade $I_1 \xrightarrow{\gamma_1} I_2 \xrightarrow{\gamma_2} I_3$ from ^{175}Lu studied.

the relevant simplified decay schemes.

A difference experiment on directional correlations of the γ - γ cascades mentioned above was performed to test again for the presence of a time violating term of the form $(\vec{k}_1 \cdot \vec{k}_2)(\vec{I}_1 \cdot \vec{k}_1 \times \vec{k}_2)$, as described earlier.

This appears to be the first TRIV test on an $E1 + M2$ transition (the 283-keV γ -ray). Also, it should be emphasized that the difference method chosen to measure the asymmetries appears to be unique in the literature.

The electromagnetic phase results obtained are interpreted in terms of Weisskopf estimate normalizations of the TRIV components of the γ_1 radiations in each case. Also, for the two γ - γ cascades in ^{169}Tm , we have interpreted the TRIV results in terms of possible mixing of the $7/2^+$ $[1/2]$ level at 139-keV into the I_1 spin state, which is the $7/2^+$ $[7/2]$ level at 316-keV, in order to estimate the resultant expectation value of a TRIV component of the interaction Hamiltonian.

As in the ^{180}Hf TRIV experiment, the angles $\theta_1 = \theta_2 = \pi/2$, $\phi = 3\pi/4$ were chosen so as to maximize the leading TRIV term in the directional correlation function.

The polarization was again vertical, produced by the same pair of superconducting coils with reversible current source, while the correlation was performed in the horizontal plane.

The results of the experiment have been accepted for publication (Mur 74b).

Methods

In order to achieve polarization of initial states (the 32 day half-life ground state of ^{169}Yb and the 101 hour half-life ground state of ^{175}Yb), advantage was taken of the large hyperfine magnetic field experienced by Yb nuclei when dilutely alloyed into gold. This field has an effective value $H_{\text{hf}} = 1.77 \text{ MOe}$ up to 1.86 MOe as the external field on the sample is varied from 1 kOe to 10 kOe (Spa 71, 73). With our applied field of 5 kOe, we have taken $H_{\text{hf}} = +1.8 \text{ MOe}$ as a reasonable value. Hence, significant polarization of the $^{169,175}\text{Yb}$ ground states (which have magnetic moments $-0.63 \pm 0.09 \mu_N$ and $+0.40 \pm 0.05 \mu_N$ respectively (Kra 72a) occurs at a temperature of 20 mK.

For each working sample, 20 mg of natural Yb was placed in a sealed, nitrogen-filled container and irradiated with a flux of 1.4×10^{12} thermal neutrons/cm²-sec for 3 h. The activated Yb was then arc-melted into gold to 2% and rolled to a thickness of 0.2 mm. After acid etching, radiographs showed uniform distribution throughout the foil. A small piece 2 mm x 8 mm was cut from the foil and indium soldered to the copper sample holder with the principal axis vertical. (A diagram of the cryogenic portion of the apparatus was shown in Fig. V-2.) The working samples had initial ^{175}Yb activity of 5 μCi .

Two 7.6 cm x 7.6 cm NaI (Tl) detectors (7-1/2% resolution at 1.33-MeV) were positioned in the horizontal plane of distances of 5 cm for the ^{169}Tm and from 7 cm to 5 cm for the shorter-lived ^{175}Lu radiation, depending on source strength. Sn shielding of thickness 0.01 cm reduced X-ray lines in each spectrum.

Two samples were utilized for the ^{175}Lu 283-114-keV cascade, for a total of 175 hours of data accumulation. For the two ^{169}Tm cascades, measured simultaneously, one sample sufficed to provide an approximately equal amount of data time. Runs were mostly of 20 min duration, with the polarizing field rotated over a 40 sec period through 180° between runs by the automatic data collection system described elsewhere (Han 74). By this means an accumulative difference measurement of the correlation function was obtained in order again to determine the asymmetry

$$A = \frac{W(\uparrow) - W(\downarrow)}{W(\uparrow) + W(\downarrow)} \quad (\text{V-8})$$

where $W(\uparrow)$ indicates the directional correlation function measured with nuclear spin pointing up (and vice versa) and an azimuthal angle ϕ of 135° from detector 1 (detecting $\vec{k}_1$) to detector 2 (detecting $\vec{k}_2$) (Fig. V-5).

The methods of data accumulation and directional correlation were as described in more detail for the previous experiment. The spectra were accumulated for both singles and coincidences. Typical spectra are shown in Figures V-10 and V-11. To obtain good coincidence efficiency, NaI(Tl) detectors were chosen. Hence for ^{169}Tm the 198-keV (γ_1) and 177-keV (γ_1) radiations could not be fully resolved, and the SCA windows were set to include both. As a result, that coincidence spectrum gives the partially resolved 110-keV and 131-keV peaks so that the two ^{169}Tm correlation asymmetries could be measured simultaneously but separately. Typical peak to background ratios were 4 to 1 in the coincidence spectra.

Fig. V-10. Typical singles and coincidence (w/177-198-keV) γ -spectra from ^{169}Tm , using 7.6 x 7.6 cm NaI(Tl) detectors. Starred peaks are from ^{175}Lu .

Fig. V-11. Typical singles and coincidence (w/ 283 -keV) γ -spectra from ^{175}Lu , using 7.6 x 7.6 cm NaI(Tl) detectors. Unstarred peaks are from ^{169}Tm .

As before, a minicomputer based acquisition system recorded and integrated the peaks of interest in the singles and coincidence spectra (including a background region). Also, the singles peak centroid positions and SCA counts were recorded automatically for each run and corresponding asymmetries computed. The peak count asymmetries computed. The peak count asymmetries, along with the standard deviations and Poisson χ^2 test values were again automatically computed.

Residual systematic asymmetries were checked for in the aforementioned SCA and centroid measurements. Careful magnetic shielding procedures, described previously, again were used to effectively eliminate gain shifts due to magnetic interaction with the detector phototubes. Such gain shifts were consistently less than 0.02%.

Measured asymmetries

The measured asymmetries in the case of cold ($T = 15 - 20$ mK) and warm (0.2, 0.5, and 4 K) temperatures are listed in Table V-4. The measured asymmetries listed have been corrected as follows:

- (1) Corrections for systematic asymmetries were made by noting that the effect of a positive systematic asymmetry in γ_1 count rate is to reduce the measured correlation asymmetry. Small additive corrections for residual systematic asymmetries in the single channel analyzer outputs, as listed in Table V-4, were made.
- (2) Asymmetries in the background regions of the coincidence spectra were measured as listed in Table V-4. Corrections were made accordingly. These corrections had an effect of 10-20% on the measured asymmetries.

Table V-4. Summary of raw measured asymmetries and corrections needed for the correlations discussed in the text for the ^{169}Tm and ^{175}Lu experiments. Raw data is the quantity $[W(\uparrow) - W(\downarrow)]/[W(\uparrow) + W(\downarrow)]$ without corrections. Also listed are the measured systematic asymmetries in the γ_1 coincidence gating count rates and in background regions of the coincidence spectra for which the measured asymmetries have been corrected as well as for the geometrical factor $Q_2(\gamma_1)Q_2(\gamma_2)$. Cold data were taken with $T = 20$ mK while warm data were accumulated with $T = 200$ mK to 4K.

$E_{\gamma_1} - E_{\gamma_2}$ (keV)	Raw measured asymmetries $\times 10^4$		Systematic γ_1 asymmetries $\times 10^4$		Background asymmetries $\times 10^4$		Corrected asymmetries $\times 10^4$	
	Warm	Cold	Warm	Cold	Warm	Cold	Warm	Cold
198-110	+86.1 $\pm$ 3.7	+76.5 $\pm$ 3.2	+0.3 $\pm$ 0.3	+0.16 $\pm$ 0.19	+10.2 $\pm$ 12.4	+4.4 $\pm$ 12.4	+221 $\pm$ 11	+198 $\pm$ 10
177-131	+262 $\pm$ 6	+223 $\pm$ 4	+0.3 $\pm$ 0.3	+0.16 $\pm$ 0.19	+10.2 $\pm$ 12.4	+4.4 $\pm$ 12.4	+709 $\pm$ 18	+609 $\pm$ 19
283-114		+4.2 $\pm$ 3.3		+2.5 $\pm$ 0.2		+4.1 $\pm$ 3.2		+1.9 $\pm$ 5.8

- (3) Finally, corrections for representative true-to-chance ratios of 4.5:1 and 7:1 in the ^{169}Tm and ^{175}Lu measurements, respectively, were made by appropriate multiplicative factors. In the last two columns of Table V-4 are the asymmetries after the three corrections listed above and finally after correction for the standard factors $Q_2(\gamma_1)$ $Q_2(\gamma_2)$ correcting for finite angular resolution.

TRIV interpretation of data

The orientation and angular distribution parameters used in computer computations of the directional correlation functions for each of the three cases studied, both at $T = 20$ mk and at higher temperatures, are enumerated and discussed in a later section.

For the two correlations in ^{169}Tm , the measured (corrected) asymmetries $A_m^{(\text{corr})}$ at warm temperatures listed in Table V-4 were found to correspond to precessions of the intermediate states (I_2) of $2-1/2^\circ$ and 10° respectively. These would contribute the terms W' to the correlation functions. From the intermediate state half lives $t_{1/2} = 63$ psec and 0.32 nsec and from the magnetic dipole moments $\mu = 0.735 \pm 0.053 \mu_N$, and $1.32 \pm 0.065 \mu_N$, for the 118 and 139 keV states, respectively (Gun 69), one finds that these mean precession angles are consistent with an "effective" hyperfine field of $0.2 - 0.3$ MOe. Of course this result cannot be related simply to the actual hyperfine field of Tm in gold due to the disruptive effects of the preceding electron capture in ^{169}Yb coming through relatively short-lived states. However, such an interpretation of the precession does tend to support a previous measurement (Ste 74) of that hyperfine field which put an upper limit of 1 MOe on its magnitude.

198-110-keV correlation in ^{169}Tm .

An effective precession angle for the intermediate state of $(2.6 \pm 0.13)^\circ$ (deduced from the measured warm correlation symmetry) led to the prediction that the "warm" asymmetry should be 4.9×10^{-4} larger than the asymmetry measured at "cold" temperatures, in the absence of TRIV. However, this experiment resulted in a much larger difference, 22.6×10^{-4} . The difference between the prediction and measurement, including uncertainty in the precession angle, gives the result for the TRIV-related asymmetry; i.e., that contribution not accounted for by precessional perturbation is found to be given by

$$A(\text{TRIV}) = (18 \pm 15) \times 10^{-4},$$

leading to a phase of the E2 component with respect to the M1 component of one 198-keV radiation equal to

$$\sin \eta = +0.15 \pm 0.12.$$

Table V-5 summarizes the phase angle results for each cascade.

Although the preceding result does not place a highly sensitive upper limit on the magnitude of $\sin \eta$, the large hindrance factors for the M1 and E2 radiation components lead to reasonable upper limits on the out-of-phase, TRIV, components of the associated transition matrices.

The partial transition rates for the M1 and E2 components of the 198-keV transition, found by using the previously quoted values for lifetime and mixing ratio and by deducing partial lifetimes of 2.0 μsec and 20 μsec respectively for these components of the γ -radiation

Table V-5. Results of directional correlation computation for $W(\theta_1, \theta_2, \phi) = W_0 + W_T$ (not including precessional effects, for the cascades of interest in ^{169}Tm and ^{175}Lu , with $\theta_1 = \theta_2 = 90^\circ$, $\phi = 135^\circ$, with orientation parameters corresponding to $T = 20$ mK as discussed in text. W_0 is the sum of P-even, T-even terms contributing to W , and W_T is the sum of P-even, T-odd terms. Also listed are the deduced TRIV asymmetries and mixing ratio phases ($\sin \eta$ where $\delta = |\delta|e^{i\eta}$. a^{TRIV} has been deduced from the data of Table V-4 corrected for precessional effects as described in text

Cascade (keV)	W_0	W_T ($\sin \eta$)	W_T/W_0 ($\sin \eta$)	a^{TRIV} ($\times 10^{-4}$)	$\sin \eta$
198-110	1.060	-0.0128	-0.012	-18 ± 15	$+0.15 \pm 0.12$
177-131	1.066	-0.0068	-0.0064	-81 ± 26	$\geq 0^{(a)}$
283-114	1.156	-0.00134	-0.0012	$+1.9 \pm 5.8$	-0.17 ± 0.5

(a) In this case we attempted to correct for a large ($\sim 10^\circ$) precession perturbation in a way which may be inadequate, as is usually the case with such large precessions.

(also see Hag 68), are found to be: $T_e(M1) = 3.65 \times 10^5 \text{ sec}^{-1}$,
 $T_e(E2) = 3.3 \times 10^4 \text{ sec}^{-1}$. These values lead to hindrances in Weisskopf
units $H_W(M1) = 0.67 \times 10^6$ and $H_W(E2) = 0.6 \times 10^3$.

As was pointed out earlier in this chapter, it is quite useful to
evaluate the imaginary (out-of-phase) parts of the electromagnetic
matrix-elements as functions of the Weisskopf estimates. Hence we
have,

$$\frac{\text{Im} \langle 2 || M1 || 1 \rangle}{| \langle 2 || M1 || 1 \rangle_W |} = \frac{-\sin \eta_a}{\sqrt{H_W(M1)}} = -(1.0 \pm 1.5) \times 10^{-4}$$

and

$$\frac{\text{Im} \langle 2 || E2 || 1 \rangle}{| \langle 2 || E2 || 1 \rangle_W |} = +(6.2 \pm 5.0) \times 10^{-3},$$

where the denominators are the Weisskopf estimates for these transition
amplitudes (the square roots of Weisskopf transition rates).

Analysis of previous TRIV experiments on M1 + E2 transitions shows
the present null result to be of comparable sensitivity, as shown in
the final section of this chapter.

To explore further possible consequences of this TRIV measurement,
let us consider the hypothetical situation where TRIV in the 198-keV
radiation comes about as a result of TRIV mixing of the $I^\pi = 7/2^+$,
 $K = 1/2$ level at 139-keV into the 316-keV state, since this is the only
one known nearby state with equal spin and parity. (An extensive sur-
vey for γ -rays of intensity more than 10^{-4} quanta per decay is pre-
sented in Mim 70.) Following the arguments of Stey 73, if this mixing

occurs, then some of the out-of-phase radiation would be "mixed into" the observed 198-keV γ -rays. We would write for the 316-keV initial state $|1\rangle$, with the convention $|1^\pi K\rangle$, $|1\rangle = |7/2^+ 7/2\rangle + i\epsilon |7/2^+ 1/2\rangle$, where $\epsilon = \langle 2|H_{\text{TRIV}}|1\rangle / 0.177 \text{ MeV}$, $\langle 2|H_{\text{TRIV}}|1\rangle$ being the expectation value of the T-violating part of the interaction Hamiltonian, assuming $H = H_0 + iH_{\text{TRIV}}$. Writing $|1\rangle = |1e\rangle + i|1o\rangle$, $|2e\rangle + i|2o\rangle$, where e and o indicate T-even and T-odd parts, we have from Stey 73, the following expression for the TRIV effect:

$$0 = \frac{|\delta| \sin \eta}{1+\delta^2} = \epsilon \frac{\langle 2e||E2||1e\rangle \langle 2e||M1||1o\rangle - \langle 2e||M1||1e\rangle \langle 2e||E2||1o\rangle}{\langle 2e||M1||1e\rangle^2 + \langle 2e||E2||1e\rangle^2} \quad (\text{V-9})$$

This allows for an evaluation of the first order perturbation parameter ϵ and thus of $\langle 2|H_{\text{TRIV}}|1\rangle$. In order to do so, the four matrix elements in Eqn. V-9 must be computed as follows. The reduced matrix elements $\langle 2e||M1||1e\rangle$ and $\langle 2e||E2||1e\rangle$ are obtained from their definitions as square roots of partial transition probabilities for the M1 and E2 components of γ -radiation in the 198-keV transition. Hence we have

$$\langle 2e||M1||1e\rangle = [T_e(M1)]^{1/2} = 0.6 \times 10^3 \text{ sec}^{-1/2}$$

and

$$\langle 2e||E2||1e\rangle = [T_e(E2)]^{1/2} = 0.18 \times 10^3 \text{ sec}^{-1/2}.$$

The $\langle 2e||\lambda||1o\rangle$ matrix elements can be estimated by determining the partial transition probabilities for M1 and E2 γ -radiation in the 20.8-keV transition from the $7/2^+$ 139-keV state to the $5/2^+$ 118-keV

state. To do so we then correct for transition energy dependence E^3 and E^5 in the M1 and E2 components, respectively, and take δ (E2/M1) = 7.15×10^{-4} (Gun 69) and a deduced branching ratio 0.335 (Ale 63, Yen 72). The results are

$$\langle 2e || M1 || 1o \rangle = 1.10 \times 10^5 \text{sec}^{-1/2}$$

and

$$\langle 2e || E2 || 1o \rangle = 2.79 \times 10^4 \text{sec}^{-1/2}.$$

(Substantially similar values, within 15%, for these last two matrix elements result from applying values for the intrinsic parameters of the $K = 1/2$ ground state rotational band obtained from energy line fits in ^{169}Tm (Gun 69) to theoretical formulas for the reduced transition probabilities in rotational bands [Kau 68, Nat 65]).

From these four reduced matrix elements, Eq. V-9 yields the result

$$\langle 2 | H_{\text{TRIV}} | 1 \rangle = (0.177 \text{ Mev}) \varepsilon = 6.83 \times 10^{-3} \text{ Mev} \sin \eta,$$

which becomes, with the value for $\sin \eta$ determined in this experiment

$$\langle 2 | H_{\text{TRIV}} | 1 \rangle = +1.0 \pm 0.8 \text{ keV},$$

thus establishing a reasonably small upper limit on this TRIV part of the interaction under the particular state mixing we have hypothesized.

One could also consider mixing of the $7/2^+ [3/2]$ state at 718.3-keV into the 316-keV level. However, insufficient available information on branching, mixing ratios, and lifetime make that impractical at present.

177-131-keV correlation in ^{169}Tm

After attempting to correct the large measured asymmetry for the effect of the 10° precession angle deduced from the warm data, we were left with the anomalous result

$$A^{(\text{TRIV})} = (81 \pm 26) \times 10^{-4},$$

indicating a possible non-zero TRIV effect. However, the large precession perturbation throws doubt upon such an interpretation, since the assumption of equal mean precession angles over the range of observation temperatures may not be correct, not to mention other physical effects which may not be understood for such a large perturbation. We note that such an asymmetry result leads to a value for the corresponding mixing ratio phase angle given by $\sin \eta = +1.3 \pm 0.4$. It seems safe only to deduce, as shown in Table V-5 that $\sin \eta \geq 0$.

Nevertheless, the following simple arguments show that a modestly sensitive measurement of $\sin \eta$ could yield a quite good TRIV measurement.

From relative intensity measurements and total conversion coefficients (Yen 72, Ale 63, Bro 67, Hat 56) along with theoretical values for $\alpha(\text{M1})$ and $\alpha(\text{E2})$ (Hag 68), we compute the experimental partial transition rates $T_e(\text{M1}) = 2.0 \times 10^5 \text{sec}^{-1}$ and $T_e(\text{E2}) = 3.8 \times 10^4 \text{sec}^{-1}$ for the 177-keV transition, with hindrances $H_W(\text{M1}) = 8.8 \times 10^5$ and $H_W(\text{E2}) = 3.1 \times 10^2$ Weisskopf units. Hence, as fractions of the single-particle Weisskopf estimates, we have

$$\frac{\text{Im} \langle 2 || \text{M1} || 1 \rangle}{|\langle 2 || \text{M1} || 1 \rangle_W|} = -1.1 \times 10^{-3} \sin \eta$$

and

$$\frac{\text{Im} \langle 2 || E2 || 1 \rangle}{|\langle 2 || E2 || 1 \rangle_W|} = +5.7 \times 10^{-2} \sin \eta,$$

so that a measurement of $|\sin \eta| < 0.1$ or better would yield a good TRIV test.

Let us now consider what would be the effect of direct mixing of the $7/2^+[1/2]$ 139-keV and $7/2^+[1/2]$ 316-keV states participating in the 177-keV transition, a mixing we consider allowed since the spins and parities are equal. Again following arguments of Stey 73, we let the 316-keV state be $|1\rangle$ and the 139-keV state be $|2\rangle$, and consider mixing of the type $|2\rangle = |2e\rangle + i\epsilon|1e\rangle$, $|1\rangle = |1e\rangle + i\epsilon|2e\rangle$, where $\epsilon = \langle 2 || H_{\text{TRIV}} || 1 \rangle / 0.177 \text{ MeV}$. With this kind of mixing, then, from first-order perturbation it is possible to show that

$$0 = \epsilon [\langle 2e || L' || 1e \rangle (\langle 2e || L || 2e \rangle - \langle 1e || L || 1e \rangle)$$

$$\frac{-\langle 2e || L || 1e \rangle (\langle 2e || L' || 2e \rangle - \langle 1e || L' || 1e \rangle)}{|\langle 2e || L || 1e \rangle|^2 + |\langle 2e || L' || 1e \rangle|^2} \quad (\text{V-10})$$

$$= \frac{\epsilon |\delta|}{1+\delta^2} \left\{ \frac{\langle 2e || L || 2e \rangle - \langle 1e || L || 1e \rangle}{\langle 2e || L || 1e \rangle} \frac{\langle 2e || L' || 2e \rangle - \langle 1e || L' || 1e \rangle}{\langle 2e || L' || 1e \rangle} \right\} \quad (\text{V-11})$$

For this case, since we are dealing with static moments, we formulate the mixing in terms of the reduced matrices of the multipole tensor operators, $\langle || M(\lambda) || \rangle$ (Nat 65) rather than the transition rate defined matrices used earlier. Eqn. V-11 can be recast in the following form:

$$\sin \eta = \varepsilon \frac{\langle 2e || M(M1) || 2e \rangle - \langle 1e || M(M1) || 1e \rangle}{\langle 2e || M(M1) || 1e \rangle} - \frac{\langle 2e || M(E2) || 2e \rangle - \langle 1e || M(E2) || 1e \rangle}{\langle 2e || M(E2) || 1e \rangle} \quad (V-12)$$

The matrices of the form $\langle 1e || M(\lambda) || 1e \rangle$ are related to observable moments by the relations (Boh 69, pp. 333, 337)

$$\mu = (4\pi/3)^{1/2} (2I + 1)^{-1/2} \langle II10 || II \rangle \langle 1K || M(M1) || 1K \rangle \quad (V-13)$$

and

$$eQ = (16\pi/5)^{1/2} (2I + 1)^{-1/2} \langle II20 || II \rangle \langle 1K || M(E2) || 1K \rangle \quad (V-14)$$

Using a value $\mu = 0.154 \pm 0.008 \mu_N$ (Nig 72) for the magnetic dipole moment of the 316-keV state and $\mu = 1.318 \pm 0.065 \mu_N$ for the 139-keV state (Gun 69) we obtain $\langle 7/2 \ 7/2 || M(M1) || 7/2 \ 7/2 \rangle = 0.24 \pm 0.01$ and $\langle 7/2 \ 1/2 || M(M1) || 7/2 \ 1/2 \rangle = 2.1 \pm 0.1 \mu_N$.

Using a value $(7.7 \pm 0.2) \times 10^2 \text{e-fm}^2$ for the intrinsic electric quadrupole moment, Q_0 , of the ground state rotational band in ^{169}Tm we assign through the relation $Q = Q_0 \frac{3K^2 - I(I+1)}{(I+1)(2I+3)}$ the following quadrupole moments:

$$Q(316) = (3.60 \pm 0.09) \times 10^2 \text{e-fm}^2$$

$$Q(139) = -(2.4 \pm 0.07) \times 10^2 \text{e-fm}^2,$$

which result in the following matrix elements, using Eqn. (V-14):

$$\langle 7/2 \ 7/2 || M(E2) || 7/2 \ 7/2 \rangle = (-4.7 \pm 0.01) \times 10^2 e\text{-fm}^2$$

$$\langle 7/2 \ 1/2 || M(E2) || 7/2 \ 1/2 \rangle = (+3.2 \pm 0.1) \times 10^2 e\text{-fm}^2.$$

The transition matrix elements are found as follows. The reduced transition probabilities $B(\lambda)$ are evaluated from the partial lifetimes of the M1 and E2 transitions $t_{1/2}(M1) = 3.5 \pm 0.6 \mu \text{ sec}$ and $t_{1/2}(E2) = 18.4 \pm 3.3 \mu \text{ sec}$, i.e. $B(M1) = [\tau(M1) \times 1.76 \times 10^{13} E^3(\text{MeV})]^{-1} \mu_N^2 = (2.0 \pm 0.3) \times 10^{-6} \mu_N^2$ and $B(E2) = [\tau(E2) \times 1.22 \times 10^9 E^5(\text{MeV})]^{-1} e^2\text{-fm}^4 = 0.18 \pm 0.03 e^2\text{-fm}^4$. From the relation

$$B(\lambda) = \frac{1}{2I_i + 1} | \langle I_f K_f || M(\lambda) || I_i K_i \rangle |^2 \quad (\text{V-15})$$

we find

$$\langle 2e || M(M1) || 1e \rangle = 4.0 \pm 0.3 \times 10^{-3} \mu_N,$$

and

$$\langle 2e || M(E2) || 1e \rangle = -(1.2 \pm 0.1) e\text{-fm}^2,$$

recalling that the E2/M1 mixing ratio is negative.

Thus, solving for ϵ in Eqn. V-12 we have

$$\epsilon = 8.8 \times 10^{-4} \sin \eta$$

so that

$$\langle 2 | H_{\text{TRIV}} | 1 \rangle = -1.6 \times 10^{-4} \text{ MeV} \times \sin \eta.$$

Hence with this mixing model, a null measurement of $\sin \eta$ of sensitivity 10^{-1} or better would yield a measurement of the T-violating part of the Hamiltonian to a sensitivity of the order of 10 eV or less, comparable to some recent work (Bli 73b).

A measurement of TRIV in the 177-keV radiation by some other method which avoided the precession problem would provide a meaningful TRIV test in this case. For example if the linear polarization $\vec{\epsilon}$ of the 177-keV γ -ray is measured, and the initial 316-keV level with spin 1 is polarized, then a search for a term of the form $(\vec{k} \cdot \vec{I}_1 \vec{x} \vec{\epsilon}) (\vec{I}_1 \cdot \vec{\epsilon}) (\vec{I}_1 \cdot \vec{k})$, could be performed (Bli 73a, Boe 68).

213-114-keV correlation in ^{175}Lu

Since ^{175}Lu does not experience any significant hyperfine field in gold, precessional effects were not important, and the corrected cold data yielded

$$A^{\text{TRIV}} = +(1.9 \pm 5.8) \times 10^{-4}.$$

Here we have no obvious candidates for state mixing, so we shall discuss only $\text{Im} \langle ||E1|| \rangle$ and $\text{Im} \langle ||M2|| \rangle$ for the 283-keV transition. The experimental values for partial transition rates in the γ -ray are $T_e(E1) = 7.7 \times 10^7 \text{sec}^{-1}$ and $T_e(M2) = 1.24 \times 10^5 \text{sec}^{-1}$, thus leading to hindrances of $H_W(E1) = 5.1 \times 10^5$ and $H_W(M2) = 10.2$ in Weisskopf units, so that

$$\frac{\text{Im} \langle 2 ||E1|| 1 \rangle}{|\langle 2 ||E1|| 1 \rangle_W|} = (+0.2 \pm 0.7) \times 10^{-3}$$

and

$$\frac{\text{Im} \langle 2 || M2 || 1 \rangle}{|\langle 2 || M2 || 1 \rangle_W|} = -0.05 \pm 0.16 .$$

So these experiments on ^{169}Tm and ^{175}Lu have resulted in a test of T-violation to 10^{-3} sensitivity in the E1 component and 10^{-1} in the M2 component of the 283-keV radiation. As remarked earlier in the introduction, this appears to be the first test by any method for TRIV components in an E1 + M2 transition.

Conclusions

The results of this experiment show that, when certain assumptions about the nature of T-violating potentials are made, TRIV can be precluded to an upper limit of 10^{-3} to 10^{-4} in the M1 + E2 198-keV transition in ^{169}Tm and in terms of a simple state mixing model to show that the T-violating part of the mixing Hamiltonian would have to be zero within about 2 keV. Similarly TRIV is ruled out to a sensitivity of $\sim 10^{-3}$ in the E1 component and $\sim 10^{-1}$ in the M2 component of the 283-keV E1 + M2 γ -radiation in ^{175}Lu . We have also argued that a reasonable measurement (10^{-1} - 10^{-3} or better) of $\sin \eta$ in the mixing ratio of the 177-keV γ -ray in ^{169}Tm by some method other than directional correlations would provide a good TRIV test.

Orientation parameters

Since certain estimates were required in arriving at orientation and angular distribution parameters to be used in computing directional correlation functions, we enumerate here the relevant parameters for each of the three cascades studied.

198-110-keV

For the 20 mK correlation, from previous work (Kra 72a, we had $B_2(I_0) U_2(I_1) G_2(\gamma_1) = B_2(I_1) = 0.046 \pm 0.004$, where $B_2(I_0)$ is an orientation parameter of the initial ^{169}Yb ground state, $U_2(I_1)$ is the deorientation parameter due to the electron capture decay of that state to I_1 and $G_2(\gamma_1)$ is the attenuation coefficient for the angular distribution of γ_1 , and $\delta_1(E2/M1) = -0.30$. Assuming $G_1/G_2 \sim 1.75$ and, by deduction from the tables of Kra 71b, that $U_1/U_2 \sim 1.25$, $B_1(I_1)/B_2(I_1) = -2$, we deduce the value $B_1(I_1) = -0.23 \pm 0.1$. $B_3(I_1)$ and $B_4(I_1)$ were set equal to zero. From the mixing ratio $\delta_2(E2/M1) = -0.17 \pm 0.03$ for the 110-keV γ -ray (Kra 72a), we have for the angular distribution of γ_2 , $A_2 = 0.667 \pm 0.046$ and $A_4 = 0.02$. For the $T = 4$ K correlation, we used $B_1(I_1) = -0.00016$, $B_2(I_1) = B_4(I_1) \equiv 0$.

177-131-keV

Since the 177-keV γ -ray originates at the 316-keV level, the orientation parameters for this case are identical to those of the preceding case, while the angular distribution coefficients for the pure E2 131-keV γ -ray are $A_2(\gamma_2) = -0.468$ and $A_4(\gamma_2) = -0.358$. For the 177-keV radiation, $\delta_1 = -0.44 \pm 0.01$ (Kra 72b).

283-114-keV

From the β -decay of the ^{175}Yb ground state to the 396-keV level (I_1) in ^{175}Lu , the deorientation parameters are $U_1 = 0.975$, $U_2 = 0.925$, $U_3 = 0.8498$, $U_4 = 0.7495$ (Kra 71b). From Kra 73a, we have for the 20 mK alignment parameters of the ^{175}Yb ground state $B_1(I_0) = -0.72$, $B_2(I_0) = 0.25$, $B_3(I_0) = -0.06$, $B_4(I_0) = 0.01$, and for the 283-keV γ -ray, $\delta_1(M2/E1) = -0.036 \pm 0.004$ (Kra 72a). No attenuation of the γ_1 angular distribution has been observed so we use for the correlation

parameters $B_1(I_1) = B_1(I_0)$ $U_1(I_1) = -0.70$ and similarly $B_2(I_1) = 0.23$, $B_3(I_1) = -0.06$, $B_4(I_1) = 0.008$ at $T = 20$ mK. At $T = 4$ K we used the values $B_1 = -0.003$, $B_2 = B_3 = B_4 \equiv 0$. For the 114-keV radiation, $\delta_2(E2/M1) = 0.45$ (Kra 72a) which gives $A_2(\gamma_2) = 0.452$, $A_4(\gamma_2) = +0.098$.

The 922-keV γ -ray of ^{184}W

Selection criteria

The 922-keV γ -ray from ^{184}W (see Fig. V-12 for level scheme) was selected for TRIV study for the following reasons:

- (1) The 922-keV radiation, resulting from a transition from the $5^- [5]$ 1285-keV state to the $4^+ [0]$ 364-keV state and the succeeding 253-keV γ -ray resulting from a further decay to the $2^+ [0]$ 111-keV state are strong and easily resolved with NaI (Tl) 7.6 cm x 7.6 cm detectors.

In the electron capture decay of ^{184}Re (see Fig. V-13) which has a $3^- [3]$ ground state of half-life 38 days and an $8^+ [8]$ isomeric state of 165 day half-life, the 1285-keV level of ^{184}W is predominantly populated by the decay of the isomeric state (Kra 73e). Typical singles and coincidence spectra taken with NaI(Tl) detectors and Tennelec 202BLR amplifiers are shown in Figure V-14. In the singles spectrum the strong 162-keV as well as the 292-keV and 313-keV lines from ^{183}W are resolved, as are the 216- and 793-keV ^{184}W lines. The NaI detectors do not fully resolve the 895-, 904-, and 922-keV lines from ^{184}W ; however, the 895- and 904-keV transitions do not compete in coincidence with either of the 922- or 253-keV γ -rays under investigation.

- (2) The 1285-keV level in ^{184}W can be polarized by initial polarization of the $^{184}\text{Re}^m$ state. Re is highly soluble in iron and

Fig. V-12. Detailed level diagram of ^{184}W , showing population from beta-decay of ^{184}Ta (Yat 72). Rotational and vibrational bands discussed in the text are indicated.

Fig. V-13. Simplified decay scheme of ^{184}Re , showing population of the 1285-keV level which gives rise to the 922-253-keV γ - γ cascade (Kra 73c).

Fig. V-14. Typical singles and coincidence (w/ 253-keV) γ -spectra from ^{184}W , using 7.6 x 7.6 cm NaI (Tl) detectors.

nickel (Sel 70).

In concentrations of the order of 1% at., the Re nucleus experiences hyperfine magnetic fields equal to -760 ± 15 kOe and -100 ± 2 Oe in Fe and Ni, respectively (Kon 67). [We note here a misprint in the compilation of Koster and Shirley (Kos 71.)] The hyperfine splitting $\mu H / I k$ for $^{184}\text{Re}^m$ in iron at 1% at. concentration has been measured as $|\Delta(^{184}\text{Re}^m)|_{(\text{Fe}, 1\% \text{ at.})} = 10.1 \pm 0.1 \text{ mK}$, corresponding to a magnetic moment $|\mu(^{184}\text{Re}^m)| = (2.90 \pm 0.15)\mu_N$. (Kra 73d) with Nilsson model arguments made that this magnetic moment is positive. The anticipated magnetic hyperfine splitting for Re dissolved in Ni would thus be given by

$$|\Delta(^{184}\text{Re}^m)|_{(\text{Ni}, 1\% \text{ at.})} = \frac{-100 \pm 2 \text{ kOe}}{-760 \pm 15 \text{ kOe}} \times |\Delta(^{184}\text{Re}^m)|_{(\text{Fe}, 1\% \text{ at.})}$$

or

$$|\Delta(^{184}\text{Re}^m)|_{(\text{Ni}, 1\% \text{ at.})} = 1.33 \pm 0.04 \text{ mK}.$$

At anticipated source temperature of 20 mK, $(\Delta/T) \approx 0.5$ in Fe or 0.07 in Ni, corresponding to the following anticipated sets of orientation parameters, from the tables of Krane (Kra 71b, 73a) for a spin=8 state:

For Re in Fe:

$$B_0 = 1, B_1 = -1.319, B_2 = 1.0123, B_3 = -0.5715, B_4 = 0.2544.$$

For Re in Ni:

$$B_0 = 1, B_1 = -0.3, B_2 = 0.05, B_3 = -0.005, B_4 = 0.$$

The deorientation parameters U_K for the decay from the $^{184}\text{Re}^m$ state to the 1285-keV $5^- [5]$ level in ^{184}W are partially known. From angular distribution measurements (Kra 73e), $U_2 = 0.731 \pm 0.023$ and $U_4 = -0.22 \pm 0.69$.

We can estimate U_1 by scanning a table of deorientation parameters (Kra 71b) and noting that these values for U_2 and U_4 correspond approximately to the deorientation due to a single transition from I_1 to I_2 with $(I_1, I_2, L) = (8, 6, 3), (8, 7, 3),$ or $(8, 8, 3)$ in each case with a corresponding value for U_1 from 0.91 to 0.92. So it is most likely reasonable to let $U_1 = 0.91 \pm 0.02$ for the purposes of the decay in question. Following similar reasoning, we shall let $U_3 = 0.55 \pm 0.05$.

Thus, the orientation parameters for the 1285-keV level are as follows, at 20 mK:

$$\text{in Iron: } B_0 U_0 = 1, B_1 U_1 = -1.20, B_2 U_2 = 0.74,$$

$$B_3 U_3 = -0.31, B_4 U_4 = -0.06.$$

$$\text{in Nickel: } B_0 U_0 = 1, B_1 U_1 = -0.27, B_2 U_2 = 0.036,$$

$$B_3 U_3 = -0.001, B_4 U_4 = 0.$$

These computations show that the $B_1 U_1$ values are sufficiently high at 20 mK to represent significant polarization of the 1285 mK level in both cases.

- (3) The precessional effect of the intermediate $(I_2) 4^+ [0]$ level at 364-keV was estimated as follows. The magnetic dipole moment of that level is $+1.20 \pm 0.18$ (Ger 70). The nuclear magnetic hyper-

fine field at a W site is -643 ± 13 kOe in Fe and -90 ± 25 kOe in Ni crystals at 1% at. concentration levels (Stea 73). [The Purdue Tandem Group has recently published a value -65 ± 5 kOe for the hyperfine field on W in Ni at $T = 305\text{K}$ (Gar 74) using the recoil implantation method. Similar results for this hyperfine field have been deduced by other groups. However, due primarily to the controversial nature of the transient field correction (Kra 74c) and also to the lack of correction for a possible cone angle effect (Kra 73c), the more conservative value for the hyperfine field quoted above has been used in the present computations.] The half-life of the $I = 4$, 364-keV state is 4.35×10^{-11} sec. (Kos 71). The resultant mean predicted precessional angles

$$\bar{\theta} = \frac{\mu_I H}{|I|} \frac{t_{1/2}}{\ln 2}$$

are given by

$$\begin{aligned} \bar{\theta}_{(\text{Fe})} &= \frac{(1.20 \pm 0.18) (5.05 \times 10^{-24})}{4 (1.05 \times 10^{-27})} \frac{(-643 \pm 13) \times 10^3 (4.35 \times 10^{-11})}{(0.693)} \text{ rad} \\ &= (-3.3 \pm 0.5)^\circ \end{aligned}$$

similarly in nickel, we expect

$$\bar{\theta}_{(\text{Ni})} = (-0.5 \pm 0.3)^\circ$$

While the expected mean precession angle in iron is a bit large, both predicted angles are nevertheless small enough not to rule out the possibility of making precessional perturbation corrections to obtain measured TRIV asymmetries.

(4) The E1 multipole component of the 922-keV transition is hindered by a factor of 0.7×10^{11} Weisskopf units (Kra 73d) while the E2 hindrance is about 10^6 ; the M3 hindrance is about 10^3 with respect to the Weisskopf estimate. Hence, especially for the E1 component, possible enhancement of any TRIV effect was a consideration, following the arguments presented earlier.

The 922-253 γ - γ directional correlation from ^{184}W satisfied all reasonable criteria for a good TRIV experiment by the methods outlined earlier in this chapter.

Source preparation

Following a procedure previously found successful (Kra 73e), a disc of natural W (14.4% ^{183}W) was sent to the Lawrence Berkeley Laboratory to be irradiated with 50 MeV deuterons in the beam of the 88 inch cyclotron, forming ^{184}Re through the reaction $^{183}\text{W} (d,n) ^{184}\text{Re}$. The disc was 2 inches in diam and 0.005 inches in thickness, and could be inserted directly into a target holder at the cyclotron facility. The first sample, used for the Fe host experiments had been irradiated a year earlier; the second was irradiated at irregular intervals during the second quarter of 1973, prior to being shipped to our laboratory in early June, 1973, after about 100 μAh of exposure. The deuteron beam activated an area 1 inch in diameter. However, radiographs made by placing the sample on ASA 3000 Type 47 Polaroid film sheets showed the radioactivity to be non-uniformly distributed on this circle, and to be concentrated on one side. A piece about 1/4" in diameter from the most active region was delivered to Dr. Joseph Bubernak of IASL Group CMB-1, who performed a chemical separation of the Re activity by dissolving the foil in an acid solution and precipitating ReS .

Following this, Dr. Clayton Olsen, of LASL Group CMB-8, prepared the working sample as follows: The ReS was reduced in an atmosphere H_2 at $1100^\circ C$ for about 4 hours, then the resultant Re was arc-melted with Fe, rolled into a foil of thickness 0.5 mm, and finally annealed in H_2 at $900^\circ C$ for 2 hours. Radiographs, made as described previously, showed uniform distribution of radiation throughout the sample.

Electromagnetic mixing ratios

At present, experimental evidence for the values of the M2/E1 and E3/E1 electromagnetic mixing ratios (δ_{12} and δ_{13}) is conflicting and ambiguous. Krane, Olsen, and Steyert (Kra 73a) found, by averaging two previously published electron conversion results (Gla 69, Age 68) and combining this average with their angular distribution coefficients ($A_2 = +0.317$ (11), $A_4 = +0.075$ (60) of the 922-keV γ -ray on a graphical plot of δ_{13} vs. δ_{12} , that two regions of overlap of the A_2 solution with the electron conversion results occurred. The two sets are:

$$\text{Set 1: } \delta_{12} = +0.09 \pm 0.02, \delta_{13} = +0.24 \pm 0.05$$

$$\text{Set 2: } \delta_{12} = -0.14 \pm 0.04, \delta_{13} = -0.19 \pm 0.03$$

The A_4 solution corresponding to $A_4 = +0.075 \pm 0.060$ did not overlap well in either region, but did lie closer to Set 1, with both δ 's positive (see Figure V-15).

In an almost simultaneous publication, Hübel, et al (Hub 73) reported angular distribution coefficients for this γ -ray from a nuclear orientation experiment. Their result for A_2 was in agreement with that reported by Krane, Olsen, and Steyert, but their result for

Fig. V-15. Plots of solutions for the mixing ratios δ_{12} and δ_{13} in the 922-keV γ -ray of ^{184}W , resulting from: (a) analysis of previous k-conversion coefficients α_K and measurement of angular distribution coefficients A_2 , A_4 by nuclear orientation techniques (Kra 73e), (b) simultaneous fits to electron conversion and angular correlation data, with two minima in the fitting parameter χ^2 (Can 73), (c) nuclear orientation measurements of A_2 and A_4 (Hub 73), and (d) perturbed-angular-correlation measurements (the present work) due to precession of the 4.35×10^{-11} sec 364-keV state with ^{184}W as a dilute impurity in iron at warm temperatures. It is deduced from these combined plots that the most likely set of mixing ratio solutions is that for which both are negative.

A_4 was -0.23 ± 0.10 . In order to compare this result with other mixing ratio solutions, the following procedure was followed in the present work: The expression for A_K in the case of this triply mixed transition is given by

$$\begin{aligned} A_K = & [F_K(11I_f I_i) + 2\delta_{12} F_K(12I_f I_i) + 2\delta_{13} F_K(13I_f I_i) \\ & + 2\delta_{12}\delta_{13} F_K(23I_f I_i) + \delta_{12}^2 F_K(22I_f I_i) \\ & + \delta_{13}^2 F_K(33I_f I_i)] / (1 + \delta_{12}^2 + \delta_{13}^2). \end{aligned} \quad (V-16)$$

where the mixing ratios are defined in the Krane and Steffen notation (Kra 70). For $K=4$, the relevant F-functions are, in this case, $F_4(1145) \equiv 0$, $F_4(1245) \equiv 0$, $F_4(1345) = 0.5000$, $F_4(2345) = -0.4392$, $F_4(2245) = 0.5666$, $F_4(3345) = -0.0852$, from previously mentioned tables (Kra 71b). With a Wang programmable calculator the quadratic equation solution for δ_{13} in terms of δ_{12} and A_K , resulting from Eqn. V-16, was found for $A_K = -0.23 \pm 0.10$ and for values of δ_{12} from -0.25 to 0.25 . A band, labeled (c), for this solution is shown on Figure V-15. It can be observed that overlap with the set of δ 's which are both negative is obtained. Again in a nearly simultaneous publication, Canty, et al (Can 73) performed K-electron conversion and angular correlation measurements on this γ -ray and a fit to both sets of data which they performed resulted in two minima, as depicted in Figure V-15, with minimum values for fitting $\chi^2 = 0.35$ in the positive region and $\chi^2 = 2.2$ in the negative region. Hence Canty, et al, speculated that the positive set of δ 's might be the best solution.

A band corresponding to the solutions to the present PAC data is shown on Figure V-15, labeled "PAC". From the overlapping

of the various data shown in Figure V-15, it becomes readily apparent that, except for the A_4 measurement of Krane, Olsen, and Steyert, the best region of overlapping solutions coincides with a negative set for δ_{12} , δ_{13} , as proposed by Krane, Olsen and Steyert and discussed above. Hence, we have carried out our analysis assuming the negative set is the correct one, although some consequences of assuming the set with both δ_{12} and δ_{13} positive are also discussed below. Thus, we take for the M2/E1 and E3/E1 electromagnetic mixing ratios of the 922-keV radiation $\delta_{12} = -0.14 \pm 0.04$ and $\delta_{13} = -0.19 \pm 0.03$, respectively.

The present work involved a measurement of a large precession perturbation of the 922-253-keV correlation at warm (no nuclear polarization) temperatures, for W in ferromagnetic Fe. In order to obtain a mixing ratio solution for these results, one can write the simplified Krane and Steffen correlation function (Kra 70)

$$W(\theta) = \sum_{K=0,2,4} B_K(\gamma_1) A_K(\gamma_2) Q_K(\gamma_1) Q_K(\gamma_2) P_K(\cos \theta), = \sum_{K=0,2,4} A_{KK} P_K(\cos \theta). \quad (V-17)$$

with $Q_2(\gamma_1)Q_2(\gamma_2) = 0.64$, $Q_4(\gamma_1)Q_4(\gamma_2) \approx 0.15$, $A_2 = -0.4477$, and $A_4 = -0.3044$. The orientation parameters $B_K(\gamma_1)$ (in this case for the intermediate state) are given by the relation

$$\begin{aligned} B_K = & [F_K(1154) - 2\delta_{12}F_K(1254) \\ & + 2\delta_{13}F_K(1354) - 2\delta_{12}\delta_{13}F_K(2354) \\ & + \delta_{12}^2 F_K(22I_{f_i}) + \delta_{13}^2 F_K(33I_{f_i})] / (1 + \delta_{12}^2 + \delta_{13}^2) \end{aligned} \quad (V-18)$$

where δ_{12} and δ_{13} are the M2/E1 and E3/E1 electromagnetic mixing

ratios for the 922-keV transition, in the Krane & Steffen notation.

With the F_2 and F_4 values from the same tables, we find the following formula for A_{22} and A_{44} :

$$A_{22} = [-0.0570 - 2\delta_{12}(-0.21684) + 2\delta_{13}(-0.19980) + \delta_{12}^2(-0.08163) + \delta_{13}^2(-0.12980) - 2\delta_{12}\delta_{13}(-0.02633)] / (1 + \delta_{12}^2 + \delta_{13}^2) \quad (V-19)$$

and

$$A_{44} = [2\delta_{13}(-0.0780) + \delta_{12}^2(-0.0084) + \delta_{13}^2(-0.00447) - 2\delta_{12}\delta_{13}(-0.02742)] / (1 + \delta_{12}^2 + \delta_{13}^2) \quad (V-20)$$

For the expected precession angle of $\Delta\phi = -3.3 \pm 0.5^\circ$ (clockwise) as discussed above, the correlation function would be perturbed an amount

$$\begin{aligned} \Delta W(\Delta\phi) &= W(\phi - \Delta\phi) - W(\phi + \Delta\phi) = (1 + \delta_{12}^2 + \delta_{13}^2)^{-1} A_{22} \\ &\quad \{P_2[\cos(\phi - \Delta\phi)] - P_2[\cos(\phi + \Delta\phi)]\} \\ &\quad + A_{44} \{P_4[\cos(\phi - \Delta\phi)] - P_4[\cos(\phi + \Delta\phi)]\} . \end{aligned} \quad (V-21)$$

In order to be certain of obtaining a realistic solution, we let $\Delta\phi = -4^\circ$, so we have, with $\phi = 135^\circ$

$$\begin{aligned} \Delta W &= (1 + \delta_{12}^2 + \delta_{13}^2)^{-1} \{A_{22} [P_2(\cos 139^\circ) - P_2(\cos 131^\circ)] \\ &\quad + A_{44} [P_4(\cos 139^\circ) - P_4(\cos 131^\circ)]\} \end{aligned} \quad (V-22)$$

which reduces to

$$\begin{aligned} \Delta W = & [-0.00954 + 2\delta_{12} (0.04528) + 2\delta_{13} (-0.04244) \\ & + 2\delta_{12}\delta_{13} (0.00789) + \delta_{12}^2 (-0.01817) \\ & + \delta_{13}^3 (-0.02750)] / (1 + \delta_{12}^2 + \delta_{13}^2) . \end{aligned} \quad (V-23)$$

With our programmable calculator, Eqn. V-23 was solved for δ_{13} for the case with $\Delta W = (-51 \pm 13) \times 10^{-4}$ (the experimental value), with δ_{12} in steps of 0.05 from -0.25 to +0.25.

Directional correlation function computations

For the orientation parameters at $T = 20$ mK of the 1285-keV level in ^{184}W , the values of $B_{K'K}^U$ enumerated earlier, were used. For the 253-keV γ -ray which is pure E2, resulting in a transition from the $4^+[0]$ 364-keV level to the $2^+[0]$ 111-keV state (two ground state $K=0$ rotational band levels), the angular distribution parameter values required are $A_2 = F_2(2224) = -0.4477$ and $A_4 = F_4(4424) = -0.3044$.

Results of the correlation computation are listed in detail in Tables V-6 and V-7 for the central values of mixing ratios and correlation parameters. Ranges of uncertainty for W and W_T were estimated by inserting appropriate values of δ_{12} , δ_{13} and the correlation parameters over their ranges of uncertainty. In this case of triple mixing, dependence of W_T upon $\sin \eta_{12}$ and $\sin \eta_{13}$ were separated by computing W_T in each case with either $\sin \eta_{12}$ or $\sin \eta_{13}$ set identically equal to zero, as discussed in Chapter IV. The results of this procedure were as follows, for typical 20 mK data temperature

Table V-6. Directional correlation function terms for Fe host, ^{184}W 922-253-keV $\gamma\text{-}\gamma$ cascade, with $\delta_{12} = -0.14$, $\delta_{13} = -0.19$. The F's are the generalized F-functions described in text, and the AG's are the generalized A functions appropriate to triple mixing. L1, L, L2 listings stand for λ_1 , λ , λ_2 . The last column is the product of B, AG, A, and H. W is the sum of these products for $\theta_1 = \theta_2 = 90^\circ$, $\phi = 135^\circ$. The P-even T-odd terms (of the two types discussed in text) are listed separately. B represents the B_{λ_1} 's, the orientation parameters for the 1285-keV level at $T = 20$ mK. Detector geometry factors are not included

L1	L	L2	F(L1)	F(L1+1)	F(L1+1)	F(L1+2)	F(L1+2)	F(L1+2)	A	M	PRD
0	0	0	1.0000	0.0000	1.0000	0.0000	0.0000	1.0000	1.0000	1.0000	1.0000
0	2	2	.1595	-.7568	.2849	.6973	.4530	1.0000	-.4477	.2500	-.0131
0	4	4	0.0000	0.0000	.1937	.1703	.0979	1.0000	-.3044	-.0662	-.0155
2	0	2	2.1005	0.0000	1.5754	0.0000	.4742	.7390	1.0000	-.2236	-.1514
2	2	2	.2944	-.9309	-.0000	.3619	.0176	.7390	1.0000	-.2236	-.1229
2	4	4	-.2511	.9324	-.2690	.3753	-.5401	.7390	1.0000	-.1336	-.0154
2	2	2	1.477	-.0563	.5275	1.0249	.2083	.7390	1.0000	-.1002	.0000
2	4	4	0.0000	0.0000	.4545	.3775	.0297	.7390	1.0000	-.1345	-.0074
2	6	6	0.0000	0.0000	-.2683	-.2096	-.1076	.7390	1.0000	-.1419	-.0104
4	0	4	2.3032	0.0000	0.0000	0.0000	.4404	.7390	1.0000	-.0316	-.0081
4	2	2	.6521	-1.3751	.3789	0.0000	-.1113	.7390	1.0000	-.1250	.0024
4	4	4	-.2727	1.0760	-.3083	.2619	-.6884	.7390	1.0000	-.0712	-.0080
4	6	6	0.0000	0.0000	.5195	.3362	.0399	.7390	1.0000	-.0712	.0041
4	2	2	0.0000	0.0000	-.5666	.5000	-.4392	.7390	1.0000	-.0750	-.0024
4	4	4	0.0000	0.0000	-.6088	.5174	.0726	.7390	1.0000	-.0750	.0032
4	6	6	0.0000	0.0000	.4432	.2450	-.8509	.7390	1.0000	-.1143	-.0001
4	2	2	0.0000	0.0000	0.0000	0.0000	.7574	.7390	1.0000	-.0750	-.0001
4	4	4	0.0000	0.0000	0.0000	0.0000	.5463	.7390	1.0000	-.0750	-.0001

WITHETA1= 50,THETA2= 50,PHI=135) = 1.00491

P-EVEN-T-ODD

L1	L	L2	F(L1)	F(L1+1)	F(L1+2)	B	AG	A	M	PRD
1	2	2	.1954	.0356	-.4501	1.2000	.1602	-.4477	.6124	-.0427
1	4	4	0.0000	.1274	-.0604	1.2000	.0153	-.3044	.1398	-.0004
3	2	2	-.6213	-.5483	.6055	.3100	-.1903	-.4477	.2004	.0053
3	4	4	.4308	-.2535	-1.2214	.3100	.4529	-.3044	-.2362	.0101
3	6	6	0.0000	.2408	-.1958	.3100	.0504	-.4477	-.3169	.0024
3	2	2	0.0000	-.3625	.1856	.3100	-.0495	-.3044	-.3169	-.0007
3	4	4	0.0000	.0356	-.4501	1.2000	-.0500	-.4477	.6124	.0165
3	6	6	0.0000	.1274	-.0604	1.2000	.0004	-.3044	.1398	-.0001
3	2	2	-.6213	-.5483	.6055	.3100	.1372	-.4477	.2004	-.0024
3	4	4	.4308	-.2535	-1.2214	.3100	-.1270	-.3044	-.2362	.0024
3	6	6	0.0000	.2408	-.1958	.3100	.0121	-.4477	-.3169	.0005
3	2	2	0.0000	-.3625	.1856	.3100	-.0183	-.3044	-.3169	-.0003

WITHETA1= 90,THETA2= 90,PHI=135) = -.03622

SIN ETA2 = 0

L1	L	L2	F(L1)	F(L1+1)	F(L1+2)	B	AG	A	M	PRD
1	2	2	.1954	.0356	-.4501	1.2000	-.0500	-.4477	.6124	.0165
1	4	4	0.0000	.1274	-.0604	1.2000	.0004	-.3044	.1398	-.0001
3	2	2	-.6213	-.5483	.6055	.3100	.1372	-.4477	.2004	-.0024
3	4	4	.4308	-.2535	-1.2214	.3100	-.1270	-.3044	-.2362	.0024
3	6	6	0.0000	.2408	-.1958	.3100	.0121	-.4477	-.3169	.0005
3	2	2	0.0000	-.3625	.1856	.3100	-.0183	-.3044	-.3169	-.0003

WITHETA1= 90,THETA2= 90,PHI=135) = .00977

Table V-7. Directional correlation function components for the 922-253-keV γ - γ cascade from ¹⁸⁴W at T = 20 nK, with a nickel polarizing host. The notation on this computer output is identical to that of Table V-6

P-EVEN-T-O-EVEN			I1 = 9.0	I2 = 4.0	L = 1	O2L =	-1.0	KPS =	0.00	O2L2 =	-1.0		
L1	L	L2	FILL1	FILL-1	FIL-1L-1	FILL-2	FIL-1L-2	FIL-2L-2	0	A0	A	H	SHAN
0	0	0	1.0000	0.0000	1.0000	0.0000	0.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000
0	2	2	.1525	-.7560	.7949	.0973	-.0719	.4530	1.0000	.1170	-.4477	.7500	-.0131
0	4	4	0.0000	0.0000	1.0000	.1937	.1700	-.0000	.0973	1.0000	-.0000	-.4477	-.0131
2	0	2	2.1005	0.0000	1.5754	0.0000	0.0000	.0742	.0300	2.0000	-.0477	.0074	.0074
2	2	0	.2946	-.0300	-.0000	.0619	-.0477	.0170	.0300	.3125	1.0000	-.4000	-.0000
2	2	2	-.2511	.0924	-.0000	-.0201	.3752	-.0001	.0300	-.3427	-.4477	-.1336	-.0000
2	2	4	.1477	-.0563	.0000	1.0000	.0777	.0000	.0300	.0000	-.3000	-.1000	.0000
2	4	2	0.0000	0.0000	.0563	-.3775	-.0000	.0000	.0300	-.1700	-.4477	-.1336	-.0000
2	4	4	0.0000	0.0000	-.2563	-.2000	.7363	-.1070	.0300	.1400	-.3000	-.4610	.0000
2	6	4	0.0000	0.0000	0.0000	0.0000	0.0000	.4400	.0300	.0101	-.3000	.0310	-.0000

WTHETA1 = 1.7372742 = 90.000(135) = .07720

P-EVEN-T-ODD

L1	L	L2	FILL-1)	F(L-1L-2)	FILL-2)	0	A0	A	H	SHAN
			SIN CTA1 = 0							
1	2	2	.1000	.0350	-.4501	.2700	-.1000	-.4477	.0120	.0010
1	4	4	0.0000	.1274	-.0000	.2700	-.0131	-.3000	.1300	.0002
3	2	2	-.0213	-.5403	.0000	-.0100	.1000	-.4477	.0000	.0002
3	4	4	.0300	-.2535	-.1000	-.0100	-.4477	-.3000	-.2302	.0002
3	6	4	0.0000	.2000	-.1000	-.0100	-.4477	-.3000	-.2302	.0002
3	6	6	0.0000	-.3025	.1000	-.0100	-.4477	-.3000	-.2302	.0002

WTHETA1 = 0.017742 = 90.000(135) = .01250

SIN CTA2 = 0

1	2	2	.1000	.0350	-.4501	.2700	-.1000	-.4477	.0120	.0010
1	4	4	0.0000	.1274	-.0000	.2700	-.0131	-.3000	.1300	.0002
3	2	2	-.0213	-.5403	.0000	-.0100	.1000	-.4477	.0000	.0002
3	4	4	.0300	-.2535	-.1000	-.0100	.1000	-.4477	-.3000	.0002
3	6	4	0.0000	.2000	-.1000	-.0100	-.4477	-.3000	-.2302	.0002
3	6	6	0.0000	-.3025	.1000	-.0100	-.4477	-.3000	-.2302	.0002

WTHETA1 = 0.017742 = 90.000(135) = -.00300

Host = Fe, $T = 20$ mK, $\phi = 135^\circ$

$$W_O = 1.00 \quad (4)$$

$$W_T = [-0.0092 \quad (27) \sin \eta_{12} + 0.036 \quad (5) \sin \eta_{13}]$$

$$T \geq 2K:$$

Host = Ni, $T = 20$ mK, $\phi = 135^\circ$,

$$W_O = 0.9771 \quad (85)$$

$$W_T = [-0.0038 \quad (11) \sin \eta_{12} + 0.0121 \quad (18) \sin \eta_{13}].$$

The effects of the anticipated precession angles, as discussed previously, were deduced from computations of the correlation functions at $\phi = 135^\circ \pm 4^\circ$ and $135^\circ \pm 0.5^\circ$ for the Fe and Ni hosts, respectively. This data is listed in Tables V-8 and V-9.

Experimental details

The geometrical angles θ_1 , θ_2 , ϕ as well as the electronics system, were identical to that previously described for TRIV experiments on Hf, Tm and Lu. Again the two 7.6 cm x 7.6 cm NaI (Tl) detectors were used, in this case with filtering consisting of 5 mils tin and 5 mils lead foil, to reduce the counting of low energy X-rays and X-rays below 200-keV in energy. In Figure V-14 is indicated the SCA window setting (on the 253-keV line). Figure V-14 also shows a typical coincidence spectrum (with $2\tau = 300$ nsec). It should also be noted that the 793-keV peak extends into the 922-keV peak (the one of interest) in the coincidence spectrum, due to the resolution of the NaI(Tl) crystal, so that an appropriate correction must be made.

Integration peaks for the Nova program were set appropriately on the several peaks. Asymmetries in two background regions were deter-

Table V-8. Predicted precessional asymmetries, $A(\text{pred}, \text{Fe})$, for the 922-253-keV correlation from ^{184}W , Fe polarizing host, with $\theta_1 = \theta_2 = 90^\circ$, $\phi = 135^\circ$, with a precession of the intermediate state equivalent to $\Delta\phi = -3.3 \pm 0.5^\circ$

T	δ_{12}	δ_{13}	$W(\phi=131^\circ)$	$W(135^\circ)$	$W(139^\circ)$	W_T	A
20 mk	-0.14	-0.16	0.98951	0.98771	0.99024	$+0.00489\sin\eta_1 - 0.03081\sin\eta_2$	+0.00236
warm			0.98054	0.97381	0.96801		-0.00412
20 mk	-0.14	-0.22	1.01780	1.01975	1.02216	$+0.00963\sin\eta_1 - 0.0414\sin\eta_2$	+0.00137
warm			0.98206	0.97893	0.97706		-0.00163
20 mk	-0.10	-0.19	1.03343	1.03548	1.03789	$+0.00701\sin\eta_1 - 0.03660\sin\eta_2$	+0.00138
warm			0.98589	0.98367	0.98247		-0.0011
20 mk	-0.18	-0.19	0.97510	0.97523	0.97579	$+0.01241\sin\eta_1 - 0.03574\sin\eta_2$	+0.00022
warm			0.97688	0.96935	0.96297		-0.00459

Results: , for $\Delta\phi = -3.3 \pm 0.5^\circ$,

$$A(\text{pred}, \text{Fe}, T = 20 \text{ mk}) = (+10.6 \pm 10.7) \times 10^{-4}$$

$$A(\text{pred}, \text{Fe}, T > 200 \text{ mk}) = (-23.6 \pm 18.7) \times 10^{-4}$$

$$W_T = (0.0097 \pm 0.0027) \sin\eta_1 - (0.0361 \pm 0.0053) \sin\eta_2$$

Table V-9. Predicted precessional asymmetries, $A(\text{pred}, \text{Ni})$, for the 922-253-keV correlation from ^{184}W , Ni polarizing host, with $\theta_1 = \theta_2 = 90^\circ$, $\phi = 135^\circ$, with a precession of the intermediate state equivalent to $\Delta\phi = 0.52$

T	δ_{12}	δ_{13}	$W(\phi=134.5^\circ)$	$W(135^\circ)$	$W(135.5^\circ)$	W_T	A
20 mk	-0.14	-0.16	0.97481	0.97405	0.97331	$+0.00372\sin\eta_1 - 0.01025\sin\eta_2$	$+0.00051$
warm			0.97460	0.97381	0.97303		$+0.00054$
20 mk	-0.14	-0.22	0.98064	0.98034	0.98006	$+0.00352\sin\eta_1 - 0.01386\sin\eta_2$	$+0.00197$
warm			0.97926	0.97893	0.97863		$+0.00021$
20 mk	-0.10	-0.19	0.98585	0.98565	0.98547	$+0.00276\sin\eta_1 - 0.01278\sin\eta_2$	$+0.00013$
warm			0.98390	0.98367	0.98347		$+0.00015$
20 mk	-0.18	-0.19	0.96991	0.96907	0.96825	$+0.00407\sin\eta_1 - 0.01238\sin\eta_2$	$+0.00057$
warm			0.97023	0.96935	0.96848		$+0.00060$

Results: $A(\text{predicted}, \text{Ni}, T = 20 \text{ mk}) = (3.5 \pm 3.0) \times 10^{-4}$
 $A(\text{predicted}, \text{Ni}, T > 200 \text{ mk}) = (3.8 \pm 2.3) \times 10^{-4}$
 $W_T = (+0.0038 \pm 0.0011)\sin\eta_1 - (0.0121 \pm 0.0018)\sin\eta_2.$

mined. One consisted of the portion of the spectrum including channels 1470 through 1530 in ADC1 and channels 1480 through 1540 in ADC2 (just below the 538-keV coincidence peak). The other covered a region just above the 642-keV coincidence peak, channels 1750 - 1810 in ADC1 and channels 1760-1820 in ADC2.

The maximum current in the vertical magnet was ± 5 amps, while that in the horizontal was 10 amps, producing fields of ± 2.7 and 2.9 kOe, respectively.

For the Ni host experiments only, new Tennelec 202 BLR amplifiers were used and the coincidence resolving time 2τ was reduced to 100 nsec.

Resultant asymmetries and corrections

Fe host

Warm data. The raw overall average asymmetry for the 922-keV coincidence peak was $(-33.5 \pm 8) \times 10^{-4}$, with background to true peak ratio $R = 0.099$ and the measured background asymmetry was $A_B = (-34 \pm 14) \times 10^{-4}$. Applying a background correction, we obtain $A_p = (-33.4 \pm 8.9) \times 10^{-4}$. Subtracting from this the systematic 253-keV singles asymmetry $(-3 \pm 0.5) \times 10^{-4}$ and multiplying by a factor of 1.4 to correct for true-to-chance we obtain the corrected experimental result

$$A = (-51.1 \pm 12.5) \times 10^{-4}.$$

This relatively large effect at warm temperatures was used to help distinguish the proper mixing ratio values and will be interpreted below in terms of a precessional perturbation.

Cold (T=20 mk) data. The corrected experimental result for the 922-keV coincidence peak asymmetry was given by

$$A = (-32.1 \pm 12.5) \times 10^{-4}.$$

This result is analyzed below in terms of TRIV contribution.

Ni host

The smaller perturbation precession made the Ni host more likely useful and thus a more detailed discussion of the data correction follows.

- (a) Residual systematic asymmetry. Systematics are in this case best diagnosed by subtracting the single-channel-analyzer asymmetry factor, which is that in SCA 1 averaged with the negative of that in SCA 2, and equal to $(+0.4 \pm 0.4) \times 10^{-4}$ at warm temperatures and $(0.79 \pm 0.21) \times 10^{-4}$ at 20 mk. The rationale for such correction has been justified in previous sections. The intermediate results after this correction are $A_{(\text{warm})} = (+2.3 \pm 5.6) \times 10^{-4}$, and $A_{(\text{cold})} = (2.1 \pm 3.1) \times 10^{-4}$.
- (b) Background asymmetry. The total width of the 793-keV peak is 160 channels, the upper 30 of which are under the 922-keV peak, with a corresponding fraction of about 0.027 of the total counts lying in that region. The measured count rate in the 793 peak is about 1.75 cts/sec total. For a correction, we take a factor $B_1 = 1.75 \text{ cts/sec} \times 0.027 = 0.05 \text{ cts/sec}$, with the count rate in the "true" peak of interest given by $P = 4.3 \text{ cts/sec}$. So the ratio $R = B_1/P = 0.01$. This leads to the following asymmetry values, corrected for

the 793-keV peak: $A_{(\text{warm})} = (1.96 \pm 5.66) \times 10^{-4}$, $A_{(\text{cold})} = (-2.87 \pm 3.08) \times 10^{-4}$. Next we correct for the measured asymmetry in the 100 channels wide background region, with $B'_2 = 1.5 \text{ cts/sec/100 channels} = 0.015 \text{ cts/sec channels}$ and $P' = (4.3 \text{ cts/sec}) \div 36 \text{ channels} = 0.12 \text{ cts/sec.}$, so that the ratio $R_2 = B'_2 = 0.126$, which leads to the background corrected values $A_{(\text{warm})} = (0.93 \pm 7.38) \times 10^{-4}$, $A_{(\text{cold})} = (-2.88 \pm 3.7) \times 10^{-4}$.

- (c) True-to-chance ratio. A true-to-chance ratio $T/c = 2.6$ was measured in the 922-keV coincidence peak, with a corresponding correction factor 1.39.

Computation of TRIV contributions

In order to estimate TRIV asymmetry contributions, it is necessary to compute and correct for the precession perturbation term W' as in the previous cases.

Iron host

As summarized in Table V-8, for the negative set of mixing ratios and a precession angle $\Delta\phi = (-3.3 \pm 0.5)^\circ$, the predicted asymmetry at warm temperatures is $A_{(\text{pred,Fe,warm})} = (-23.6 \pm 18.7) \times 10^{-4}$, while the measured result was $(-51.1 \pm 12.5) \times 10^{-4}$. The average of the predicted and experimental values can be taken as $(-37 \pm 33) \times 10^{-4}$. Hence a reasonable adjustment to predicted asymmetries is to multiply the predicted asymmetry by the ratio $37/24 = 1.6$ and the predicted uncertainty by $33/19 = 1.7$. (A similar analysis for the positive set of mixing ratios leads to a predicted warm asymmetry of $(-96 \pm 17) \times 10^{-4}$, which is not in as good agreement with our experimental result.)

From Table V-8, the predicted asymmetry at $T = 20$ mk turns out to be $A_{(\text{pred}, \text{Fe}, 20 \text{ mk})} = (10.6 \pm 10.7) \times 10^{-4}$ which we correct to $(18.0 \pm 17.1) \times 10^{-4}$ while we measured $A_{(\text{meas.}, \text{Fe}, 20 \text{ mk})} = -32.1 \pm 12.5) \times 10^{-4}$. Thus the TRIV contribution should be the difference, i.e. $A_{(\text{TRIV}, \text{Fe})} = [(-32.1 \pm 12.5) - (18.0 \pm 17.7)] \times 10^{-4} = (-50.1 \pm 21.2) \times 10^{-4}$. Recalling now the P-even, T-odd contribution to the correlation function was:

$W_T = (0.0097 \pm 0.0027) \sin \eta_{12} - (0.0361 \pm 0.0053) \sin \eta_{13}$, and the "normal" part $W_O = 1.00 \pm 0.04$ (for the negative set of mixing ratios), we can now investigate possible limits on the phases $\sin \eta_{12}$ and $\sin \eta_{13}$. From Eqn. IV-31, we have only one equation in two unknowns, so a complete solution is not possible. We thus look at two possible cases.

$$\text{If } \sin \eta_{12} = 0, \text{ then } \sin \eta_{13} = \frac{(-50 \pm 21) \times 10^{-4}}{0.64} \\ \times \frac{(1.00 \pm 0.04)}{-0.036 \pm 0.0053} = 0.22 \pm 0.10.$$

On the other hand, if $\sin \eta_{13} = 0$, then

$$\sin \eta_{12} = \frac{-50 \pm 21}{0.64} \times \frac{1.00 \pm 0.04}{0.0097 \pm 0.0027} \times 10^{-4} = -0.81 \pm 0.41$$

Ni host

Here the warm measured asymmetry, $A_{(\text{meas}, \text{Ni}, \text{warm})} = +1.3 \pm 10.3 \times 10^{-4}$ clearly agrees with the predicted value $A_{(\text{pred}, \text{Ni}, \text{warm})} = (+3.8 \pm 7.3) \times 10^{-4}$, a useful check on systematics, etc.

The cold measured asymmetry $A_{(\text{meas}, \text{Ni}, 20 \text{ mk})} = (-4.0 \pm 5.1) \times 10^{-4}$ minus the predicted result $A_{(\text{pred}, \text{Ni}, 20 \text{ mk})} = (+3.5 \pm 3.0) \times 10^{-4}$ gives $A_{(\text{TRIV}, \text{Ni})} = (-7.5 \pm 5.9) \times 10^{-4}$.

From the form of W_T for this case we obtain, similarly to the iron case, these two (of many) possible solutions:

$$\sin \eta_{12} = 0, \sin \eta_{13} = +0.095 \pm 0.089;$$

$$\sin \eta_{13} = 0, \sin \eta_{12} = -0.30 \pm 0.32.$$

Since the Ni host data are considered inherently more reliable due to the smaller precession perturbation, we shall use the Ni results in carrying out the calculations of the following sections, rather than combining the data of the two experiments.

Implications of TRIV results

Transition matrices

The half-life of the 1285-keV state is 8.33 μ sec and the Weisskopf hindrance of the E1 component of the 922-keV γ -radiation from that state is 10^{11} (Gla 69), from an E1 partial half-life equal to $(2.8 \pm 0.3) \times 10^{-5}$ sec.

Assuming the negative set of mixing ratios ($\delta_{12} = -0.14$, $\delta_{13} = -0.19$) we get for the partial half-life of the M_2 component

$$\begin{aligned} \tau_{1/2} (M2) &= \tau_{1/2} (E1) \times \frac{1 + \delta_{12}^2 + \delta_{13}^2}{\delta_{12}^2} & (V-24) \\ &= 1.48 \times 10^{-3} \text{ sec.} \end{aligned}$$

thus the $M2$ hindrance is given by

$$H_W (M2) = \frac{\tau_{1/2} (M2)}{\tau_{1/2} (M2) |_W} \approx \frac{1.48 \times 10^{-3}}{10^{-9}} \approx 10^6.$$

similarly for the E3 component,

$$\tau_{1/2} (E3) = (2.8 \times 10^{-5} \text{ sec}) \frac{1.06}{0.04} (0.75 \times 10^{-3}) \text{ sec.}$$

$$\text{and } H_W (E3) \approx \frac{0.75 \times 10^{-3}}{10^{-6}} \approx 10^3 .$$

The imaginary parts of transition matrices in these two typical cases are now easily derivable.

Case I: Recall $H_W (E1) \approx 10^{11}$

$$(\sin \eta_{12} = 0)$$

$$\begin{aligned} \frac{\text{Im} \langle 2 || E1 || 1 \rangle}{|\langle 2 || E1 || 1 \rangle_W|} &= \frac{-\sin \eta_{13}}{\sqrt{H_W(E1)}} \\ &= \frac{+0.095 \pm 0.089}{3.2 \times 10^5} = (2 \pm 2) \times 10^{-7} \text{ sec}^{-1/2} \end{aligned}$$

$$\text{and } \frac{|\text{Im} \langle 2 || E3 || 1 \rangle_W|}{\langle 2 || E3 || 1 \rangle_W} = \frac{-\sin \eta_{13}}{33} = (2.9 \pm 2.7) \times 10^{-3} \text{ sec}^{-1/2}$$

Case II: ($\sin \eta_{13} = 0$)

$$\begin{aligned} \frac{\text{Im} \langle 2 || E1 || 1 \rangle}{|\langle 2 || E1 || 1 \rangle_W|} &= \frac{-\sin \eta_{12}}{\sqrt{H_W(E1)}} = \frac{+0.30 \pm 0.32}{3.2 \times 10^5} = (+0.9 \pm 1.0) \\ &\times 10^{-6} \text{ sec}^{-1/2} \end{aligned}$$

and

$$\frac{\text{Im} \langle 2 || M2 || 1 \rangle}{|\langle 2 || M2 || 1 \rangle_W|} = \frac{+\sin \eta_{12}}{\sqrt{H_W(M2)}} = (-0.3 \pm 0.3) \times 10^{-3} \text{ sec}^{-1/2}$$

Possible level mixing

Looking at the states I_1 , I_2 , I_3 , involved in the transition, we seek states which could mix into either I_1 (5^- [5], 1285-keV) or I_2 (4^+ [0], 364-keV): i.e. we need to survey any states of equal spin and parity to either of those two levels (Steig 73).

Two candidates for mixing into I_2 are the 4^+ [2] 1135-keV γ -vibrational band level and the 4^+ [3] 1537-keV level, which is considered to be a member of a $K^\pi = 3^+$ rotational band based at 1425-keV. The members are the levels at 1425-, 1537-, and 1677-keV (Yat 72). (See Figure V-12.) These are both mixtures into I_2 , so the perturbation formalism would be the same in either case.

In addition there is an apparent $K = 2^-$ octupole rotational band based at 1130-keV, with $I = 3^-$ and 4^- excitations at 1221- and 1345.5-keV (Yat 72). This predicts the existence of an excitation state with $I = 5^-$ at an energy predictable by the rotational model. In the pure rotational model (where the nuclear matter is considered to be an irrotational fluid) the excitation energy E^* above the intrinsic energy of the lowest lying level of a band can be reasonably well predicted by the well-known relation

$$E^* = \frac{(I(I+1) - K^2)\hbar^2}{2 I_{\text{eff}}} + \frac{\hbar^2 K^2}{2 I_{\text{eff}}^{[3]}} \quad (\text{V-25})$$

for spin values below 10 (Mar. 70, Vol. II, pg. 1304), where I_{eff} is the effective moment of inertia about the symmetry axis and $I_{\text{eff}}^{[3]}$ is the component of moment of inertia about an axis perpendicular to the symmetry axis.

The known excitation energies of the $I^\pi = 3^-$ and 4^- states can be used to solve for I_{eff} and $I_{\text{eff}}^{[3]}$ and thus to predict the excitation energy of the expected $I = 5^-$ level; i.e.

$$E^*(3^-) = 91\text{-keV} = \frac{4h^2}{I_{\text{eff}}} + \frac{2h^2}{I_{\text{eff}}^{[3]}}$$

$$E^*(4^-) = 215.5\text{-keV} = \frac{8h^2}{I_{\text{eff}}} + \frac{2h^2}{I_{\text{eff}}^{[3]}}$$

from which we find

$$I_{\text{eff}} = 0.0321 h^2/\text{keV}$$

$$I_{\text{eff}} = -0.0595h^2/\text{keV},$$

and thus

$$E^*(5^-) = 1/2 h^2 \frac{5(6) - 2^2}{I_{\text{eff}}} + \frac{2^2}{I_{\text{eff}}^{[3]}} = 306.6\text{-keV},$$

and

$$\begin{aligned} E(5^-)[2] &= E_0(2^- \text{ band}) + E^*(5) = 1130\text{-keV} + 306.6 \\ &= 1436.6\text{-keV} \end{aligned}$$

thus we expect a $5^-[2]$ level at 1437-keV, on the basis of a purely rotational model.

A more meaningful prediction of the excitation energy of the $5^-[2]$ level expected can be made by using the rigid octupole rotor model for even-even nuclei of Davidson (Dav 62). In this model, the deformation is described by two parameters; ζ , the octupole shape parameter,

and η , the octupole asymmetry parameter, for which the moments of inertia can be expressed thus:

$$I_1 = 4B_3 \zeta^2 (\sin^2 \eta + 1/2 \cdot 15 \sin \eta \cos \eta + \frac{3}{2} \cos^2 \eta), \quad (V-26)$$

$$I_2 = 4B_3 \zeta^2 (\sin^2 \eta - 1/2 \cdot 15 \sin \eta \cos \eta + 3 \cos^2 \eta), \quad (V-27)$$

$$\text{and } I_3 = 4B_3 \zeta^2 \sin^2 \eta \quad (V-28)$$

An eigenvalue diagram for the quantity $E(\hbar^2/8B_3\rho)^{-1}$ vs. η is shown in Fig. V-16.

From the ratio of excitation energies of the 3^- and 4^- levels alone the 2^- level of the octupole band in question, can be deduced from Figure V-15 that $\eta \approx 85^\circ$ for this band, assuming a pure octupole rotator model. This value of η gives the correct spin sequences as well. Thus the next excitation level should have $I = 5^-$ and have band excitation energy predicted by $\frac{E(5^-) - E(2^-)}{E(4^-) - E(2^-)} = \frac{27 - 7}{19 - 7} = \frac{20}{12}$ so

$$E^*(5^-) \approx \frac{5}{3} (215.5) = 359\text{-keV}$$

and

$$E_{\text{pred}}(5^-) \approx 1130 + 359 = 1490\text{-keV}.$$

Mixing of $4^+[2]$ 1134-keV γ -vibrational level into I_2 . A similar perturbation calculation to that used in the ^{180}Hf analysis (following Stey 73) yields for the two cases being discussed.

Fig. V-16. Energy eigenvalues for a pure, asymmetric, octupole rotator (Davidson octupole rotor model), plotted against the asymmetry parameter η for values of angular momentum $I \leq 8$ (from Dav 62).

Case I ($\sin \eta_{12} = 0$):

$$\begin{aligned} \rho_{13} &= \frac{|\delta_{13}| \sin \eta_{13}}{1 + \delta_{12}^2 + \delta_{13}^2} \\ &= \epsilon_{13} \frac{\langle 2e || E1 || 1e \rangle \langle 2o || E3 || 1e \rangle - \langle 2e || E3 || 1e \rangle \langle 2o || E1 || 1e \rangle}{\langle 2e || E1 || 1e \rangle^2 + \langle 2e || M2 || 1e \rangle^2 + \langle 2e || E3 || 1e \rangle^2} \quad (V-29) \end{aligned}$$

and

Case II ($\sin \eta_{13} = 0$):

$$\begin{aligned} \rho_{12} &= \frac{|\delta_{12}| \sin \eta_{12}}{1 + \delta_{12}^2 + \delta_{13}^2} \\ &= \epsilon_{12} \frac{\langle 2e || E1 || 1e \rangle \langle 2o || M2 || 1e \rangle - \langle 2e || M2 || 1e \rangle \langle 2o || E1 || 1e \rangle}{\langle 2e || E1 || 1e \rangle^2 + \langle 2e || M2 || 1e \rangle^2 + \langle 2e || E3 || 1e \rangle^2} \quad (V-30) \end{aligned}$$

The reduced matrix elements $\langle 2e || E1 || 1e \rangle$, $\langle 2e || M2 || 1e \rangle$, and $\langle 2e || E3 || 1e \rangle$ are square roots of the partial transition rates for the E1, M2, and E3 components of the 922-keV radiation or, using the previously quoted partial half-lives, we have

$$\langle 2e || E1 || 1e \rangle = [(0.693)/(2.8 \pm 0.3 \times 10^{-5} \text{ sec})]^{1/2}$$

$$= (1.57 \pm 0.08) \times 10^2 \text{ sec}^{-1/2},$$

$$\langle 2e || M2 || 1e \rangle = -[0.693/(1.48 \pm 0.83 \times 10^{-3})]^{1/2}$$

$$= -(2.2 \pm 0.6) \times 10^1 \text{ sec}^{-1/2},$$

and

$$\langle 2e || E3 || 1e \rangle = -[0.693/(0.75 \pm 0.24 \times 10^{-3})]^{1/2}$$

$$= -(3.0 \pm 0.5) \times 10^1 \text{ sec}^{-1/2}.$$

In order to utilize Eqns. V-29 and V-30 it is necessary to evaluate the three reduced matrix elements of the form $\langle 2e || L || 1e \rangle$ for this proposed perturbation. In this case these matrix elements will represent transitions between the $5^- [5]$ level at 1285-keV (I_2) and the $4^+ [2]$ 1134-keV level, with the energy difference adjusted to 922-keV.

A γ -ray of energy 151.5-keV resulting from a transition between these two states has been observed (Yat 72), although the electromagnetic mixing ratios have not been determined. The relative intensity of the 151.5-keV γ -ray is 0.22 ± 0.04 compared to 44.4 ± 1.1 for the 922-keV line. Thus the γ -partial half-life may be determined by comparison to the $(2.8 \pm 0.3 \times 10^{-5}) \text{ sec}$ γ -partial half-life for the 922-keV γ -ray (Gla 69) by the ratio of intensities; i.e.

$$t_{1/2\gamma}(151.5\text{-keV}) = (5.65 \pm 0.21) \times 10^{-3} \text{ sec.},$$

with corresponding partial transition rate $T_\gamma(151.5\text{-keV}) = (1.23 \pm 0.05) \times 10^2 \text{ sec}^{-1}$.

For $|\Delta K| = 3$, the survey of Löbner (Lob 68) shows that the expected K- hindrances (H_K) for the E1, M2, and E3 components of such a transition would be $10^{9 \pm 1}$, 10^4 , and $10^{3 \pm 1}$, respectively.

As the 151.5-keV transition is the only γ -ray resulting from a transition between the K=5 band and the γ -vibrational band, it is not presently possible to deduce theoretical reduced transition probabilities for the various multipolarity components of this γ -ray by compari-

son, using rotational-vibrational band predictions. Thus, we must resort to E1: M2: E3 ratios using the Weisskopf single-particle estimates and the predicted hindrances mentioned in the preceding paragraph.

The Weisskopf single-particle estimates for the partial transition rates for such a transition, with energy difference adjusted to $E = 0.922$ MeV and $A = 184$ are given by (Boh 69)

$$T_w(E1) = (1.02 \times 10^{14}) E^3 A^{2/3} = 2.58 \times 10^{15} \text{ sec}^{-1}$$

$$T_w(M2) = (2.23 \times 10^7) E^5 A^{2/3} = 4.81 \times 10^8 \text{ sec}^{-1}$$

$$T_w(E3) = 33.7 E^7 A^2 = 6.46 \times 10^5 \text{ sec}^{-1}$$

If the ratios of the expected K-hindrances are included then we have the ratios of predicted transition rates

$$(T(E1)/T(M2)/T(E3))_{\text{predicted}} = 1/(1.9 \times 10^{-2 \pm 1})/(7.7 \times 10^{-6 \pm 1}).$$

The E1 component clearly predominates, with predicted mixing ratios $|\delta'_{12}|^2 \approx 2 \times 10^{-2 \pm 1}$ and $|\delta'_{13}|^2 \approx 8 \times 10^{-6 \pm 1}$. From the γ -partial transition rate for the 151.5-keV γ -ray, we compute an expected E1 rate with energy adjusted to 922-keV, assuming an E^3 dependence, since the E1 component should predominate; i.e.

$$\begin{aligned} \langle 20 || E1 || 1e \rangle &\approx [T(151.5\text{-keV})(922/151.5)^3]^{1/2} \\ &= (1.66 \pm 0.01) \times 10^2 \text{ sec}^{-1/2} \end{aligned}$$

For the M2 and E3 components, we use the mixing ratios $|\delta'_{12}|^2$ and $|\delta'_{13}|^2$ thus:

$$\langle 2e || M_2 || 1e \rangle \approx \frac{\langle 2e || E_1 || 1e \rangle |\delta_{12}^e|^2}{1 + |\delta_{12}^e|^2 + |\delta_{13}^e|^2} = -3.4 \times 10^{\pm 1} \text{sec}^{-1/2}$$

and similarly

$$\langle 2e || E_3 || 1e \rangle \approx -1.4 \times 10^{-3 \pm 1} \text{sec}^{-1/2}.$$

Now the perturbation parameters ϵ_{13} and ϵ_{12} in Eqns. V-29 and V-30 may be evaluated:

$$\theta_{13} = \epsilon_{13} \frac{(1.57 \pm 0.08) \times 10^2 (-1.4 \times 10^{-3 \pm 1}) - (-3.0 \pm 0.5) \times 10^1 (1.66 \pm 0.01) \times 10^3}{[(1.57 \pm 0.08) \times 10^2]^2 + [2.2 \pm 0.6] \times 10^1]^2 + [(3.0 \pm 0.5) \times 10^1]^2}$$

The quantity in brackets is equal to $(5 \times 10^3) / [(2.6 \pm 0.3) \times 10^4] = (1.9 \pm 0.6) \times 10^{-1}$. Thus for Case I ($\sin \eta_{12} \equiv 0$)

$$\begin{aligned} \epsilon_{13} &= |\delta_{13}| \sin \eta_{13} / [(1 + \delta_{12}^2 + \delta_{13}^2) (1.9 \pm 0.6) \times 10^{-1}] \\ &= (0.19 \pm 0.03) (0.095 \pm 0.089) / (1.06) (0.19 \pm 0.06) = 0.09 \pm 0.09, \end{aligned}$$

so that the imaginary component of H producing such mixing is now given by

$$\langle 2 | H_{\text{TRIV}} | 1 \rangle_{\text{Case I}} = \epsilon_{13} (1134 - 364) \text{ keV} = 69 \pm 69 \text{ keV}.$$

For Case II, with $\sin \eta_{12} = 0$, Eqn. V-30 becomes

$$\begin{aligned} \theta_{12} &= \epsilon_{12} \frac{(1.57 \pm 0.08) \times 10^2 (-3.4 \times 10^1) - (-2.2 \pm 0.6) \times 10^1 (1.66 \pm 0.01) \times 10^2}{(2.6 \pm 0.3) \times 10^4} \\ &= \epsilon_{12} (0.13 \pm 0.04), \end{aligned}$$

so that

$$\begin{aligned}\epsilon_{12} &= |\delta_{12}| \sin \eta_{12} / [(1 + \delta_{12}^2 + \delta_{13}^2) (0.13 \pm 0.04)] \\ &= (0.14 \pm 0.04) (-0.30 \pm 0.32) / (1.06) (0.13 \pm 0.04) \\ &= -0.30 \pm 0.34,\end{aligned}$$

and in this case the TRIV mixing interaction may be estimated as

$$\langle 2 | H_{\text{TRIV}} | 1 \rangle \sim (-0.30 \pm 0.34) (177 \text{ keV}) \sim -53 \pm 60 \text{ keV}.$$

Considering the results of Cases I and II, we can tentatively put an upper limit of about 100 keV on the magnitude of the matrix element $\langle 2 | H_{\text{TRIV}} | 1 \rangle$ for this type of level mixing.

Mixing of $4^+[3]$ 1538-keV rotational level into I_2 . If we hypothesize mixing of the $4^+[3]$ level at 1538-keV into I_2 , perturbation relations used will be identical to those of the previous case. However, the matrix elements $\langle 20 || L || 1e \rangle$ must be reevaluated.

There have been no observed transitions from the $K = 3$ band of which the 1538-keV level is a member to either the 5^- or 6^- levels of the $K = 5$ band based at 1285-keV. Hence no data presently exists upon which estimates of a transition rate from the $4^+[3]$ 1538-keV level to the $5^-[5]$ 1285-keV level could be based.

Theoretical studies of interband transitions have been pursued in limited cases (Dav 62, Dav 68). However, the number of transitions available for analysis is quite limited, so that such theoretical work has not been entirely conclusive. Consequently, we shall elect to follow a simplified procedure which may hopefully yield order of magnitude estimates.

The expected K-hindrances for such a transition with $|\Delta K| = 2$ would be $10^{6\pm2}$, $10^{2\pm1}$, and $10^{-2\pm1}$ for the E1, M2, and E3 components, respectively.

Adjusting the Weisskopf single-particle estimated transition rates (again with $E = 922$ keV) by the expected K-hindrances, we have for the matrix elements of interest

$$\langle 20 || E1 || 1e \rangle = (2.58 \times 10^{9\pm2} \text{sec}^{-1})^{1/2} = 5 \times 10^4 \text{sec}^{-1/2},$$

$$\langle 20 || M2 || 1e \rangle = -(4.81 \times 10^{6\pm1} \text{sec}^{-1})^{1/2} = -2 \times 10^{3\pm1} \text{sec}^{-1/2},$$

and

$$\langle 20 || E3 || 1e \rangle = -(6.46 \times 10^{7\pm1} \text{sec}^{-1})^{1/2} = -8 \times 10^{3\pm1} \text{sec}^{-1/2}.$$

Following a procedure identical to the preceding mixing case, we obtain, from Eqn. V-29

$$O_{13} \approx \epsilon_{13} (1 \times 10^{1\pm1})$$

$$\epsilon_{13} \approx 1.7 \times 10^{-3\pm1}$$

so that, with $E = 1173$ keV,

$$\langle 2 || H_{\text{TRIV}} || 1 \rangle_{\text{Case I}} = 1994 \times 10^{-3\pm1} \text{ keV} < 20 \text{ keV}$$

and for the second solution, from Eqn. V-30,

$$O_{12} \approx \epsilon_{12} (4 \times 10^{1\pm1}),$$

$$\epsilon_{12} \approx (1 \pm 1) \times 10^{-3\pm1},$$

and

$$\langle 2 | H_{\text{TRIV}} | 1 \rangle_{\text{Case II}} \approx 12 \times 10^{-1 \pm 1} \text{ keV} < 12 \text{ keV}.$$

Combining the Case I and Case II results, 20 keV is a likely upper limit on the magnitude of amplitude of a mixing interaction for this second possible level mixing situation.

Mixing of $5^- [2]$ 1490-keV octupole level into I_1 . Again a similar perturbation approach is used, except that in Eqns. V-29 and V-30, the odd-even matrix elements take on the form $\langle 2e || L || 1o \rangle$.

We will estimate the partial transition rate for γ -radiation from the $5^- [2]$ state at a predicted energy of 1490 keV to I_2 ($4^+ [0]$, 364 keV), and then as before make an energy correction for $E = 922$ keV (rather than $1437 - 364 = 1073$ keV). We note that where the degree of K-forbiddens $|k_1 - k_f| = L$, this forbiddenness would be 1, 0, and -1 for the E1, M2, and E3 components of such a transition. From the survey of hindrance factors of Löbner (Lob 68), for $|\Delta K| = 2$ corresponding hindrances relative to the single particle estimate would be $H_W(E1) \approx 10^{6 \pm 2}$, $H_W(M2) \approx 10^{2 \pm 2}$, $H_W(E3) \approx 10^{-2 \pm 1}$, just as in the preceding case.

γ -transitions from the 2^- , 1130-keV state to I_2 , the 364-keV state ($E_\gamma = 769.8$ -keV) (M2) and from the 3^- , 1221-keV state (E3) to I_2 ($E_\gamma = 857.2$ -keV) have been observed (Yat 72).

This negative-parity band has also been partially studied by Canty, et al, (Can 73), who also computed the reduced transition probably B(E3) for the transition from the 3^- level to the ground state, obtaining $B(E3, 0^+ \rightarrow 3^-) = (25 \pm 5) \times 10^{-2} e^2 b^3$.

The magnitudes of the even-odd matrix elements will be identical to those of the previous mixing case discussed, i.e.

$$\langle 2e | |E1| | 1o \rangle \approx 5 \times 10^{4 \pm 1} \text{sec}^{-1/2},$$

$$\langle 2e | |M2| | 1o \rangle \approx -2 \times 10^{3 \pm 1} \text{sec}^{-1/2},$$

and

$$\langle 2e | |E3| | 1o \rangle \approx -8 \times 10^{3 \pm 1} \text{sec}^{-1/2},$$

so that the results for ϵ_{12} and ϵ_{13} will be the same as in the previous heme mixing and, with $\Delta E = (1490-1285) \text{ keV} = 205 \text{ keV}$,

$$\begin{aligned} \langle 2 | H_{\text{TRIV}} | \rangle_{\text{Case I}} &= \epsilon_{13} \Delta E \approx (1.7 \times 10^{-3 \pm 1}) (205 \text{ keV}) \\ &\approx 0.35 \times 10^{\pm 1} \text{ eV} < 3.5 \text{ eV} \end{aligned}$$

and

$$\begin{aligned} \langle 2 | H_{\text{TRIV}} | \rangle_{\text{Case II}} &= \epsilon_{12} \Delta E \approx (1 \pm 1 \times 10^{-3 \pm 1}) (205 \text{ keV}) \\ &\approx 0.2 \times 10^{\pm 1} \text{ eV} < 2 \text{ eV}. \end{aligned}$$

Hence it appears that for this proposed level mixing, the T-violating interaction associated with the mixing would be less than 4 eV in amplitude. This lower value appears because of the proximity of the energies of the two states which could mix.

Summary of Principal Results

In Table V-10 are summarized, for each correlation studied, the measured TRIV asymmetry, the corresponding relative phase angles for components of different multipolarity in the electromagnetic mixing ratios for the first γ -ray, and the values of the out-of-phase (TRIV) or imaginary parts of the transition matrices resulting from hindrance considerations. For the triply-mixed 922-keV radiation from ^{184}W , two possible solutions are presented as discussed in the description of that particular experiment.

Comparison with previous results

Analyses of the results of known previous TRIV experiments on γ -decay are presented in Table V-11 (adapted from Kra 74b). Of particular interest are the sizes of the imaginary, or TRIV, components of the reduced transition matrix elements, when hindrance considerations are taken into account. On this basis, from Table V-11, previous tests for TRIV have produced sensitivities from 10^{-1} to 10^{-5} , while from Table V-10, the present work has produced TRIV tests of sensitivity 10^{-8} and 10^{-6} in $^{180}\text{Hf}^m$, and has shown that the TRIV component of the E1 reduced transition matrix in the 922-keV radiation from ^{184}W may be so small as 10^{-7} times the Weisskopf estimate. The other TRIV tests are at least as sensitive as most previous work, on the basis of hindrance.

Table V-10. Summary of key TRIV results of the present program of experiments

Isotope	γ_1 energy, (keV)	$a_{\text{TRIV}} \times 10^4$	$\sin \eta$	Imaginary (TRIV) components of transition matrices, relative to Weisskopf estimates
$^{180}\text{Hf}^m$	501	P-even: 2.8 ± 5.1	$+0.048 \pm 0.087$	$\text{Im} \langle M2 \rangle_W = (4.2 \pm 7.5) \times 10^{-9}$ $\text{Im} \langle E3 \rangle_W = (1.1 \pm 1.9) \times 10^{-6}$
		P-odd: 11.3 ± 9.4		$\text{Im} \langle E2 \rangle / \text{Re} \langle E2 \rangle = -0.69 \pm 0.57$
^{169}Tm	198	-18 ± 15	$+0.15 \pm 0.12$	$\text{Im} \langle M1 \rangle_W = -(1.8 \pm 1.5) \times 10^{-4}$ $\text{Im} \langle E2 \rangle_W = +(6.2 \pm 5.0) \times 10^{-3}$
	177(a)	-81 ± 26	≥ 0	$\text{Im} \langle M1 \rangle_W = -1.1 \times 10^{-3} \sin \eta$ $\text{Im} \langle E2 \rangle_W = +5.7 \times 10^{-2} \sin \eta$
^{175}Lu	283	$+1.9 \pm 5.8$	-0.17 ± 0.5	$\text{Im} \langle E1 \rangle_W = +(0.2 \pm 0.7) \times 10^{-3}$ $\text{Im} \langle M2 \rangle_W = -(0.5 \pm 1.6) \times 10^{-1}$

Table V-10, continued

Isotope	γ_1 energy, keV	$a_{\text{TRIV}} \times 10^4$	$\sin \eta$	Imaginary (TRIV) components of transition matrices, relative to Weisskopf estimates
^{184}W	922	Ni host: -7.5 ± 5.9	$\sin \eta_{12} = 0$ $\sin \eta_{13} = 0.095 \pm 0.089$	$\text{Im} \langle E1 \rangle_W = -(2 \pm 2) \times 10^{-7}$ $\text{Im} \langle M2 \rangle_W = 0$, by definition (Case I) $\text{Im} \langle E3 \rangle_W = +(3 \pm 3) \times 10^{-3}$
			$\sin \eta_{12} = -0.3 \pm 0.3$ $\sin \eta_{13} = 0$	$\text{Im} \langle E1 \rangle_W = +(0.9 \pm 1.0) \times 10^{-6}$ $\text{Im} \langle M2 \rangle_W = -(0.3 \pm 0.3) \times 10^{-3}$ (Case II) $\text{Im} \langle E3 \rangle_W = 0$, by definition
		Fe host: $-50.1 \pm 21.2^{(b)}$	$\sin \eta_{12} = 0$ $\sin \eta_{13} = 0.22 \pm 0.10$	(Case I)
			$\sin \eta_{12} = -0.8 \pm 0.4$ $\sin \eta_{13} = 0$	(Case II)

(a) A large perturbation precession (10°) for which sufficiently accurate correction unlikely, makes this result dubious.

(b) A perturbation precession of 3.6° for the Fe host results, six times larger than for the Ni host data, makes the measured asymmetries inherently less reliable in those results.

Table V-11. Summary of previous studies of T-invariance in γ -decay (adapted from Kra 74b)

Nucleus	Method ^(a)	δ ^(b)	$\sin \eta$ (units of 10^{-3})	$H_W(L)$ ^(c)	$H_W(L+1)$ ^(c)	$\text{Im} \langle L \rangle_W$	$\text{Im} \langle L+1 \rangle_W$	Reference
^{36}Cl	n $-\gamma$	+0.21	0.8 ± 2.3	(0.26)	(0.1)	6×10^{-3}	1×10^{-2}	Eic 68
^{49}Ti	n $-\gamma$	+0.053	17 ± 25	(21)	(0.1)	1×10^{-2}	1×10^{-1}	Kaj 68
^{56}Fe	$\beta - \gamma$	+0.23	4 ± 26	(2.5)	(0.1)	2×10^{-2}	1×10^{-1}	Gar 69
^{99}Ru	M $-\vec{\epsilon}$	-1.6	1.0 ± 1.7	6×10^3	2×10^{-2}	4×10^{-5}	2×10^{-2}	Kis 67
^{106}Pd	$\beta - \gamma$	+0.21	4 ± 18	(3.5)	(0.1)	1×10^{-2}	6×10^{-2}	Per 68
^{192}Pt	$\vec{I} - \gamma$	-2.1	4 ± 5	(5×10^2)	(0.1)	5×10^{-4}	3×10^{-2}	Hol 73
^{193}Ir	M $-\vec{\epsilon}$	+0.56	1.1 ± 3.8	1100	5×10^{-2}	2×10^{-4}	3×10^{-2}	Ata 68

(a) Method used to achieve polarization of initial level (β : previous beta-decay, $\vec{I}$: low-temperature orientation, M: Mössbauer scattering, n: polarized neutron capture) and to observe final orientation (γ : angular disorientation of subsequent γ -ray, $\vec{\epsilon}$: γ -ray linear polarization).

(b) E2/M1 for all these previous cases.

(c) Hindrance of γ -ray transition probability relative to Weisskopf estimates; values in parentheses are estimated by assuming the E2 transition to be enhanced by a factor of 10.

APPENDIX A

THE ^3He - ^4He DILUTION REFRIGERATOR

Introduction

The use of 15-35 mK temperatures available by cooling small radioactive sources with a ^3He - ^4He dilution refrigerator was vital to the nuclear physics experiments described in this dissertation. In this Appendix is provided a summary of the history, physical basis, and techniques of dilution refrigerator technology, along with a brief description of the system actually used.

Properties of liquid He

Let us review the physical properties of liquid ^4He and ^3He which are relevant to an understanding of the cooling system in question.

The boiling point of ^4He at 1 atmosphere pressure is 4.216 K. Liquid helium as received from commercial sources is at about 4 K. It can conveniently be cooled by pumping to reduce the vapor pressure. For example, at 2 K, the equilibrium vapor pressure is 23.8 mm Hg; and at 1.4 K, it is 2.2 mm Hg. Such pressures are easily obtainable with reasonable pumping speeds. It is necessary to surround and precool a dilution refrigerator with a ^4He bath. (A table of vapor pressures, latent heats of vaporization, and molar volumes for ^3He and ^4He may be found in Kel 69, page 112). For example, in the present apparatus, 20

liters of liquid helium in a nitrogen-bath-surrounded dewar can easily be pumped down to a few millimeters Hg within a few hours with a Kinney pump model KOH-130 (Kinney Vacuum Division, New York Air Brake Co., Boston, Mass.) having a nominal rated capacity of 60 liters/sec for air at STP.

The entropy of liquid ^4He decreases very rapidly below 1K, so that below 500 mK, the entropy is essentially zero (Kel 69, page 356).

The specific heat of liquid ^3He rises very steeply as T decreases below 50 mK, reaching a maximum at about 2 mK (Kel 69, page 190).

Pure liquid ^4He changes phase below $T = 2.2\text{K}$ (the "lambda point") and becomes a superfluid (HeII), with well-known physical properties.

^3He - ^4He mixtures

Natural helium is only 0.0013% ^3He . Purified ^3He is available through commercial sources (e.g. Linde) in gaseous form.

The behavior of mixtures of liquid ^3He and ^4He at low temperatures has been of continuing interest to low temperature theorists. Landau and Pomeranchuk in 1948 (Lan 48) proposed a theory based on treating the ^3He atoms as free quasi-particles with energy $\epsilon = -E_3 + p^2/2m$ to derive hypothetical cooling in a dilution process. Heer and Daunt (Hee 51) and London (Lon 51) considered the ^3He atoms as ideal gas particles when mixed with ^4He in a dilute solution to produce cooling during the dilution process.

This analysis can be justified in at least a qualitative way by considering the previously quoted property of the dependence of the

entropy of ^4He below 1 K; i.e. that the entropy curve is virtually flat below 500 mK. Hence small amounts of heat absorbed into the mixture do not go into the ^4He system, but must go only into the ^3He , and thermodynamically the ^3He system is "independent" with respect to small amounts of heat transfer in that temperature region below 500 mK.

A way of analyzing the mixing process is to consider that, since ^3He has smaller mass, the zero-point energy as derived by London (see Kel 69, page 26) to be larger than for ^4He , resulting in comparatively larger molar volume and vapor pressure for ^3He . Hence introducing ^3He as an impurity into ^4He will cause a local expansion against the attractive Van der Waals potential and thus absorption of heat.

The λ -temperature decreases smoothly with increasing percentage of ^3He (at saturated vapor pressure) down to $T = 0.8$ K, with about 65% concentration of ^3He . (This is referred to as the "critical point" as shown in the phase diagram of Fig. A-1.) Walters and Fairbank verified by a nuclear magnetic resonance technique (Wal 56) the separation of liquid helium into two distinct equilibrium phases in terms of ^3He concentration as T decreases toward zero. At 20 mK for example, two phases, one with 6% ^3He concentration, the other 99% ^3He coexist. Clearly then layering would be expected in a normal gravitational potential, with the lighter mixture floating on top.

The final crucial thermodynamic property is the absorptive heat of mixing of about 54 joules/mole as derived from specific heat data by DeBruyn Ouboter, et al. (DeB 60). Hence the primary objective of the

Fig. A-1. Phase diagram of liquid helium at saturated vapor pressure (from Edw 70).

dilution cooling method seems clear; i.e. to precool a mixing chamber containing ^3He -rich helium down below 800 mK to induce phase separation with a phase interface in the mixing chamber, and to circulate ^3He through the chamber so as to provide continuous heat absorption.

Dilution refrigerator scheme

According to D. O. Edwards (Edw 70), the first working refrigerator was constructed at Leiden (Das 65) and achieved a continuous temperature of 220 mK. Other refrigerators were constructed at about the same time in Russia, Harwell and Manchester. The refrigerator in present use at IASL is a revised version of one first designed and constructed by W. A. Steyert in 1967. The present version has achieved temperatures as low as 13 mK.

A typical flow diagram of a dilution refrigerator is shown in Fig. A-2. Initially the whole apparatus, except for an external ^3He pumping system, is cooled to about 1 K by a pumped helium bath. At that point the ^3He is introduced from storage tanks and the circulating pump is turned on. The initial effect is to cool the mixture by reducing the vapor pressure. As phase separation occurs the incoming ^3He is first cooled by passing through a long coiled stainless steel tube in the ^4He bath. This brings the ^3He from essentially room temperature to about 1 K. A high impedance is inserted at this point to control flow into the first heat exchange unit, and to maintain a pressure differential between the input line and the refrigerator. The first heat exchanger unit consists of concentric flow via stainless steel tubing around the

Fig. A-2. Block flow diagram of typical dilution refrigerator (from Rad 71).

still input line, thus cooling the incoming ^3He to ~ 200 mK. The incoming liquid now passes through a series of five heat exchangers before entering the top of the mixing chamber. These heat exchangers consist of disc-shaped divided closed compartments separated by walls of sintered 99.9% pure copper powder, which provides large contact area. After passing through the mixing chamber, the cold ^3He passes through the three heat exchangers, absorbs heat from the inflow, and goes up to the still. By means of an electrical resistance heating coil wrapped around the still, energy is inserted, maintaining vaporization of the ^3He (which has the higher vapor pressure). The vapor is removed by the circulation pumping system, consisting of an oil diffusion pump and a sealed mechanical pump.

The temperatures of the mixing chamber and last heat exchanger are monitored by means of an a.c. resistance bridge measurement of 100 ohm Speer resistors attached thereto. The bridge is described in Stey 68.

Several thorough reviews of dilution refrigerator theory and technology are available (Whe 68, Whe 71, Rad 71). J. C. Wheatley has led extensive research efforts in this area and has, through the SHE Corporation, made complete cooling systems available commercially in the U.S. Another commercial source is Oxford Instruments in Great Britain.

The cooling capacity of a dilution refrigerator is theoretically given by $\dot{Q} = \dot{n}_3 (94T_{MC}^2 - 12T_N^2)$ Watt-sec/mole- K^2 , where $\dot{n}_3$ is the ^3He circulation rate in moles/sec, T_{MC} is mixing chamber fluid temperature, and T_N is the temperature of the fluid coming into the mixing chamber from

the last heat exchanger.

For the refrigerator used in these experiments, the sample temperature has been determined experimentally to be related to heat load by the relation $T(\text{mK}) \approx 14 + 3(\dot{Q}[\text{erg/sec}])^{1/2}$, so that a heat load of ≈ 1 erg/sec results in a temperature of 17 mK. This heat load can come from the radioactivity of the sample, from hysteresis heating due to magnetic field rotations and fluctuations, etc.

APPENDIX B

THERMOMETRY METHODS

In the temperature range from 1 K down to 10 mK, ordinary temperature measuring devices, such as thermocouples, with which we are familiar become too insensitive to small changes to be of any use in making reasonably accurate measurements.

For the nuclear polarization experiments of this work temperatures must in some cases be known to within about 2 mK. For example, in the $^{180}\text{Hf}^m$ PT-experiment of Chapter V, the resulting phase angle measurement was dependent on the second orientation parameter B_2 . For the 7.9 mK hyperfine splitting and spin $I = 8$ level being polarized in that case, $B_2 = 0.91$ when $T = 18$ mK, and $B_2 = 0.6$ when $T = 26$ mK, so that a decrease of only 8 mK increases B_2 -terms by 50%. (This is an extreme example; the B_1 terms of the P-even, T-odd experiments were much less temperature sensitive).

Let us review four important techniques for temperature determinations at ultralow temperatures, involving both solid state and nuclear physics experiments.

Resistance Thermometry

Although temperature determination by resistance measurement is only reliable at temperatures above about 50 mK, nevertheless this

technique serves to give readings at temperatures above those at which orientation techniques, for example, might be used. In particular, in the present work, resistance thermometry is used to determine the pre-cooling of the refrigerator before it is turned on, and as a double check that temperatures determined by more precise orientation techniques are at least "in the right ball park." In some cases, where more precise temperatures were not required, resistance measurements sufficed.

Many materials, such as carbon, do not conduct well at low temperatures. Graphite, for instance, is used as a thermal insulator in constructing parts of a refrigeration system. Certain commercially available carbon resistors, such as the 100 Ω , 1/2 watt variety made by Speer (Speer Carbon Co., Bradford, Pennsylvania) have sufficient sensitivity and repeatability to be useful in thermometry even down to 20 mK with an a.c. resistance bridge. The bridge used in these experiments is described in Ste 68. We found it difficult, if not practically impossible, to eliminate stray r.f. entering the numerous long leads into the dewar from heating the resistors, making readings below 50 mK unreliable, even with extensive shielding measures. The temperatures of the dilution refrigerator mixing chamber and last heat exchanger were determined in this manner.

The resistance vs. temperature, as determined by nuclear orientation of ^{54}Mn and ^{60}Co is plotted as a calibration curve.

Calibrations and characteristic temperature curves for various commercially available resistors frequently appear in various journals

such as Review of Scientific Instruments.

Nuclear Orientation Thermometry

With the rather recent advent of continuous millikelvin temperatures available for nuclear orientation experiments, and with many well known nuclear moments and magnetic hyperfine field values, it seems natural to use the angular distribution of a nuclear γ -ray as an indication of sample temperature, eliminating all possibility of deception due to the likes of poor thermal contact between refrigerator and sample, etc.

As reviewed in Chapter III, the measured angular distribution of a γ -ray is given by

$$W(\theta) = \sum_K B_K U_K A_K O_K P_K(\cos \theta) \quad (B-1)$$

where the only temperature dependent factor is B_K . Thus the accuracy of the measurement depends upon the sensitivity of B_K to temperature T . This sensitivity depends on the ratio of the particular hyperfine splitting (Δ) to T and on the spin involved; (as discussed in Chapter III). A standard method is to measure the γ -count rate at 90° vs. 0° with respect to the polarization axis, and then compare the ratio of the two count rates to a plot of the predicted $W(0^\circ)$ and $W(90^\circ)$ vs. T . The 835 keV line of ^{54}Mn , with the Mn in an iron foil, has been found very sensitive and easy to use (Sit 71, Ste 72). The Mn is obtained in water solution, painted onto the iron, cooked in H_2 at about 1000°C , and acid etched to remove surface contamination and undissolved Mn. This foil

can be indium-soldered to a sample. Alternatively, if the sample is in iron, the ^{54}Mn may be added directly. The hyperfine splitting of the ^{54}Mn ground state is 9 mK. The 1173 keV γ -ray resulting from the decay of ^{60}Co is also useful (Sta 72).

As an example of the use of this technique in the present work, direct measurements of 0° - 90° counting rate anisotropy in the 332 keV γ -ray from $^{180}\text{Hf}^m$ were regularly made to determine the sample temperature during the "P-T" experiment of Chapter V. In this case, the γ -ray is of E2 multipolarity, with angular distribution described by $W(\theta) = 1 + Q_2 B_2 U_2 A_2 P_2(\cos \theta) + Q_4 B_4 U_4 A_4 P_4(\cos \theta)$. The deorientation parameters U_2 and U_4 for the 6^+ 641 keV level could be computed by taking into account branching from the initially polarized 5.5 hour, 1142-keV level through the 501- and 57-444-keV radiations (see Fig. V-1), with branching ratios 14% and 86%, respectively.

Taking into account the E3/M2 mixing ratio $\delta = 5.5$, the partial deorientation parameter due to the 501 keV radiation will be (with the notation $U_k(I_i I_f L)$, and using the tables of Kra 71b) $U_2^{501}(86) = [U_2(862) + (5.5)^2 U_2(863)] [(1 + (5.5)^2)]^{-1} = 0.7937$.

Similarly, $U_4^{501}(85) = 0.3823$. For the 57-444 keV cascade, the product of the two individual deorientations is taken, i.e. $U_2^{57-444} = U_2^{57}(881) U_2^{444}(862) = 0.9069$ and $U_4^{57-444} = 0.7121$. Thus the overall deorientation parameters needed are given by $U_2 = 0.14 U_2^{501} + 0.86 U_2^{57-444} = 0.8910$ and similarly $U_4 = 0.6659$.

Finding appropriate values for the remaining parameters as discussed in Chapter IV and inserting values of Q_2 and Q_4 appropriate to the detector used to measure the counting rate asymmetries thus enables a plot of $W(0^\circ)$ and $W(90^\circ)$ vs. T . For example, with 3" x 3" NaI (Tl) detectors at a distance of 7 cm, the anticipated ratio of $W(90^\circ)$ to $W(0^\circ)$ at 30 mK is 1.38. This method yielded temperature measurements accurate to about 2 mK.

Other Methods

Several magnetic susceptibility techniques are important. Measurement of the susceptibility of a paramagnetic salt such as cerium magnesium nitrate (CMN) (which obeys Curie's Law, $XT = \text{constant}$, nearly perfectly at ultralow temperatures) is very useful down to about 10 mK. Similar measurements of nuclear spin magnetic susceptibilities, as in metallic copper, are even better due to the smaller magnetic moments compared to the electron dipole moment. The nuclear susceptibility obeys Curie's Law down to even lower temperatures in similar applied induction field strengths. These measurements of susceptibility may be made by determining mutual inductances between a coil wrapped around the sample and another coil, or by using superconducting magnetometers such as a "SQUID."

The nuclear susceptibility may be measured by resonance methods, which often have the advantage of greater sensitivity. The sample is placed in a magnetic field H_0 and the magnetization precesses at the Larmor frequency ω_0 . An r-f modulated magnetic field B_1 is applied per-

pendicular to B_0 , producing a modulating magnetization which can be described by a complex susceptibility $\chi = \chi' - i\chi''$, where χ' represents the component in phase (dispersive) with B_1 and χ'' represents the component out of phase (absorptive) with B . The resonance in power absorption occurs when $\omega = \omega_0$. The susceptibility is deduced from the strength of the absorption peak, since the power absorbed is proportional to χ'' . More details of susceptibility methods may be found in Sym 72 and Wey 70. Several commercial instruments for measurement of temperatures in the 10-100 mK range, using susceptibility techniques, are now available, some with instantaneous digital readout.

Mössbauer nuclear resonance may also be used, whereby relative populations of nuclear magnetic hyperfine states in ^{57}Fe , for example, may be determined, thus allowing deduction of temperature due to the Maxwell-Boltzmann Law. Taylor (Tay 72) has achieved good agreement with temperatures derived from nuclear orientation measurements down to 27 mK.

A review of the ultralow temperature thermometry methods mentioned here, along with discussions of several other techniques in not so common usage, is presented in Wey 70. The series of volumes referred in the citations, Stey 72, Sym 72, and Tay 72, provides a wealth of information on all aspects of thermometry.

APPENDIX C

THE DATA ACQUISITION SYSTEM

The functions performed by the minicomputer-based data system used in the present work were described in the description of the ^{180}Hf experiments in Chapter V. Here shall be described more of how these functions are accomplished in terms of signals and the individual electronic units which are part of the system. The interface system was collaboratively designed by Handy and Steyert (Han 74).

First let us review the functional operation briefly, referring to the block diagram of Fig. V-3.

The detectors were in this case 3" by 3" NaI(Tl) scintillation crystals with integrated photomultiplier tubes, the pulse signals of which were amplified by Victoreen Model PIP-400 preamplifiers, to which a modification consisting of the addition of a 390 pf capacitor in parallel with the 470 k Ω resistor between the anode and the positive high voltage supply was made to increase the pulse fall time to about 100 nsec. (This was necessary in order to meet input signal requirements of the integrating spectrum amplifiers used to process the pulses.)

The signals from the amplifiers went to two single-channel analyzers, one of which was set to a "particular" peak for gating, the other set to trigger on the entire spectrum, with coincidences being performed

between the "particular peak" from one detector and the entire spectrum from the other, with an effectively dual coincidence system, as seen in the diagram. The divide-by-ten circuit, especially constructed for these experiments by the author, using TTL integrated circuit modules, consisted of a decade counter triggering a monostable flip-flop so that every tenth count caused a pulse of adjustable length to be generated and sent through an OR gate with a coincidence pulse. (This unit was added to the interiors of SCA modules.) This resultant signal served as a trigger for a Geoscience 8040 analog-to-digital converter operated in the coincidence mode. The $\div 10$ output signal was adjusted in length (about 400 nsec) so as to overlap with the later-arriving coincidence signal. At various times different coincidence units were used, all standard commercial NIM-bin modules, with typical resolving times $2\tau = 200-500$ nsec. The motivation for using the divide-by-ten circuit was to increase coincidence counting efficiency by reducing dead time in the ADC's due to processing the high rate singles counts.

The Nova computer handles data by means of 16 bit in-out data lines, plus six device selection lines and nineteen control lines to devices from the processor, making up a 50 connector bus. All in-out devices are connected to this bus, although provisions for disabling all but the selected device must be made. The central processor selects which device is to be activated, except in the case of the "data channel" for which a device, here an ADC, controls data input.

In the data channel mode, the device output causes a corresponding

memory location to be incremented by one, corresponding to counting one pulse of a particular height.

An important part of the interface is an interrupt priority system for the three possible ADC units. The basic diagram for this system is shown in Fig. C-1. The signal marked $\overline{RQENB}$ appears once during each processing cycle and, if one of the ADC units has sent out a $\overline{DATA\ READY}$ level (a positive d.c. level), one of the three flip-flops will correspondingly be triggered by the leading edge of $\overline{RQENB}$, sending a data channel request interrupt pulse to the computer and disabling inputs to the computer from the other two ADC's. The computer responds with a data channel input signal, enabling data input from the ADC with the highest priority. Priority in the cases of simultaneous ADC requests goes to the unit closest to the interface, which is in this case ADC 1.

The interfaces between the ADC's and the Nova minicomputer were designed to save processing time by avoiding the use of program control to choose memory storage locations for the coincidence spectral data. Coincidence spectra are automatically stored in a different section of memory than singles (non-coincidence) spectra. The interface is designed to increment by a pre-selected fixed amount an ADC output when accompanied by a coincidence pulse. A diagram of this portion of the interface is shown in Fig. C-2. The basic operation is as follows. An ADC receives a pulse and begins to process it, sending out a Dead Time signal. If this pulse corresponds to a coincidence count, a coincidence (1 or 2) pulse will arrive slightly later, and the 1.5 μ sec monostable

Fig. C-1. ADC interrupt priority circuit (from Han 74).

Fig. C-2. ADC-computer data interface (from Han 74).

on the Dead Time signal will allow coincidences to trigger a flip-flop which activates a set of four OR-gate inputs on the programmable panel. The other inputs come from a 0.2 μ sec monostable triggered by the Data Accept signal from the central processor. Switches and jumpers allow setting this pulse to increment the ADC output in the 2^7 to 2^{11} bits as shown, since these four inputs are added to the four most significant bits of the ADC output in the 4-bit binary adder. At the proper time, the final AND gates put the sum onto the Nova in-out bus. The basic location of the data block in memory is program assigned, and the portion of the data block assigned to a particular ADC is program addressed for data retrieval and display.

If a memory location overflows when being incremented by ADC output, the number of that memory location is entered into the first available location in an overflow block, under program control. At the end of a data accumulation period, the overflow block is scanned for those overflow entries. The number of multiple entries for any single location correspond to the number of times overflow occurred.

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