# Search for violation of time-reversal invariance in $^{169}$ Tm and $^{175}$ Lu $\gamma$ rays\*

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Directional correlations of  $\gamma$ - $\gamma$  cascades from polarized <sup>169, 175</sup>Yb were measured and a difference technique used to search for time-reversal-invariance violating (TRIV) asymmetries. For the 198-110 keV cascade in <sup>169</sup>Tm a TRIV asymmetry  $\alpha^{\text{TRIV}} = (-18 \pm 15) \times 10^{-4}$  was determined, corresponding to a phase angle between the  $E_2$  and  $M_1$  multipole matrix elements of the 198 keV radiation given by  $\sin \eta = +0.15 \pm 0.12$  with T-odd components of the E2 and M1 matrix elements equal to  $+(1.8\pm1.5)\times10^{-4}$  and  $-(6.2\pm5.0)\times10^{-3}$  of the Weisskopf estimates. respectively. Hypothetical interpretation of TRIV as a result of state mixing is discussed, giving the expectation value of the TRIV part of the interaction Hamiltonian as  $\pm 1.0 \pm 0.8$  keV. For the 283-114 keV cascade in  $^{175}$ Lu,  $C^{TRIV} = (+1.9 \pm 5.8) \times 10^{-4}$  was measured, with corresponding M2/E1 phase angle given by  $\sin\eta = -0.17 \pm 0.5$  and T-odd matrix elements equal to  $-0.05 \pm 0.16$  and  $+(2 \pm 7) \times 10^{-4}$  Weisskopf units for the M2 and E1 components, respectively. Similar measurements on the 177-131 keV cascade in <sup>169</sup>Tm yielded uncertain results due in part to a large precessional perturbation. However, treatment of TRIV in the 283 keV  $\gamma$  ray assumed due to state mixing is shown to result in the conclusion that a modestly sensitive measurement of the phase angle of the E2 with respect to the M1 matrix element would give a good test of the magnitude of the TRIV component of the relevant interaction potential.

RADIOACTIVITY <sup>169,175</sup>Yb;  $\gamma$ - $\gamma$  directional correlations, time-reversal-symmetry tests; deduced mixing ratio phase angles, 198-, 177-, and 283-keV  $\gamma$  rays; estimated *T*-odd component, Hamiltonian.

## I. INTRODUCTION

The possibility of the violation of time-reversal invariance being evidenced in nuclear radiation is of continuing interest, due to uncertainty about possible mechanisms for such an occurrence<sup>1,2</sup> and due to previous indications in K-meson decay that T violation may occur.<sup>3</sup> In a previous paper<sup>4</sup> we found, by performing a search for asymmetries in  $\gamma - \gamma$  correlations in <sup>180</sup>Hf<sup>m</sup> [provided certain assumptions were made about the source of a timereversal-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  $\gamma$  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<sup>5</sup> led us to perform a similar test on the 198-110 keV and 177-131 keV  $\gamma$ - $\gamma$  cascades in <sup>169</sup>Tm and on the 283-114 keV cascade in <sup>175</sup>Lu. <sup>175</sup>Lu is a  $\beta$ -decay daughter of <sup>175</sup>Yb while <sup>169</sup>Tm results from electron capture in <sup>169</sup>Yb. The Weisskopf hindrances of the 198, 177, and 283 keV radiation components are as high as 10<sup>6</sup>.

A difference experiment on directional correlations of the  $\gamma$ - $\gamma$  cascades mentioned above was performed to test for the presence of a time violating term of the form  $(\vec{k} \cdot \vec{k}')(\vec{l}_1 \cdot \vec{k} \times \vec{k}')$ ,<sup>1</sup> where  $\vec{l}_1$ is the spin of the polarized nuclear state from which the first radiation  $\gamma_1$  emanates, after decay from a polarized parent state  $I_0$ , and  $\vec{k}$  and  $\vec{k}'$  are the momentum vectors of the first and second successive radiations  $(\gamma_1 \text{ and } \gamma_2)$  in a cascade.

We note that this appears to be the first TRIV test on an E1 + M2 transition (the 283 keV  $\gamma$  ray) and also 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  $\gamma_1$  radiations in each case. Also, for the two  $\gamma$ - $\gamma$  cascades in <sup>169</sup>Tm, we have interpreted the TRIV results in terms of possible mixing of the  $\frac{7}{2}$ + $\left[\frac{1}{2}\right]$  level at 139 keV into the  $I_1$  spin state, which is the  $\frac{7}{2}$ + $\left[\frac{7}{2}\right]$  level at 316 keV, in order to estimate the resultant expectation value of TRIV component of the interaction Hamiltonian.

## **II. EXPERIMENTAL METHODS**

Following the formalism recently developed by Krane, Steffen, and Wheeler,<sup>6,7</sup> the directional correlation function can be expressed as  $W = W_0$ +  $W' + W_T$  where  $W_0$  is the "normal" correlation function of  $\gamma_1$  and  $\gamma_2$  (depending only on even values



FIG. 1. Simplified decay scheme of <sup>169</sup>Yb, showing the cascades

$$I_1 \xrightarrow{\gamma_1} I_2 \xrightarrow{\gamma_2} I_3$$
 and  $I_1' \xrightarrow{\gamma_1'} I_2' \xrightarrow{\gamma_2'} I_3'$  studied.

of the alignment parameter  $B_K$ ), W' is a "false TRIV" term arising from precession perturbation of the intermediate state, and  $W_T$  is the true *T*-violating contribution. The correlation function is expressed by

$$W(\theta_1, \theta_2, \Phi) = \sum_{\lambda_1, \lambda, \lambda_2} B_{\lambda_1}(I_1) A_{\lambda}^{\lambda_2 \lambda_1}(X_1) A_{\lambda_2}(X_2) H_{\lambda_1 \lambda \lambda_2}(\theta_1 \theta_2 \Phi)$$
(1)

where  $\lambda_1$  is the index of the alignment parameters of the initial state,  $\lambda$  is a positive integral index which is even when the radiation  $X_1$  (see Figs. 1 and 2) is non-parity violating and  $\lambda_2$  is the even integral for a non-parity violating  $X_2$  (for further details, see especially Ref. 7). Furthermore, the



FIG. 2. Simplified decay scheme of  $^{175}$ Lu, showing the cascade

$$I_1 \xrightarrow{\gamma_1} I_2 \xrightarrow{\gamma_2} I_3$$
 studied.

*T*-invariant terms in *W* have  $\lambda_1 + \lambda + \lambda_2$  even while the T-violating terms have that sum odd. Since in these cases no parity violating components occur,  $\lambda$  and  $\lambda_2$  are even. It follows that the Teven terms involve only even-K terms in the initial orientation parameters  $B_K$ , and only odd-K (polarization)  $B_{\kappa}$ 's can contribute to T violation. Note that for nuclei oriented along the +z axis,  $B_{\kappa}$  is negative for odd K.<sup>6</sup> The lowest order nonzero T-violating term in  $W(\theta_1, \theta_2, \Phi)$  is, in each of the three cases studied, that with  $\lambda_1 = 1$ ,  $\lambda = 2$ ,  $\lambda_2 = 2$ , which corresponds to the scalar  $(I \cdot \vec{k} \times \vec{k}')$  $(\vec{k} \cdot \vec{k}')$ . This term also dominates the *T*-violating part of W. Thus the angles  $\theta_1 = \theta_2 = \frac{1}{2}\pi$ ,  $\Phi = \frac{3}{4}\pi$  were chosen so as to maximize the leading TRIV term in the directional correlation function.

The polarization was vertical, produced by a pair of superconducting coils with reversible current source, while the correlation was performed in the horizontal plane. This is the N1N2 geometry of Ref. 6, and is illustrated in Fig. 3.

For the electromagnetic mixing ratios  $\delta((L+1)/L)$  throughout this discussion we use the convention  $\delta = |\delta| e^{i\eta}$ .

In each case the value of the phase  $\sin\eta$  in the electromagnetic mixing ratios can be evaluated by the relation

$$\frac{W_T}{W_0} = \alpha^{\text{TRIV}}, \qquad (2)$$

noting that  $\sin\eta$  is contained in the left-hand quantity. (The out-of-phase radiation components contribute only as  $\sin\eta$  departs from zero.) In this manner the values of  $\sin\eta$  listed in Table I were obtained.

In order to achieve polarization of initial states (the 32 day half-life ground state of  $^{169}$ Yb and the 101 h half-life ground state of  $^{175}$ Yb) advantage was taken of the large hyperfine magnetic field exper-



FIG. 3. Geometry of correlation, with  $\theta_1 = \theta_2 = 90^\circ$ ,  $\Phi = 135^\circ$ .

TABLE I. Result of directional correlation computation for  $W(\theta_1, \theta_2, \Phi) = W_0 + W_T$  (not including precessional effects) with  $\theta_1 = \theta_2 = 90^\circ$ ,  $\Phi = 135^\circ$ , for the cascades of interest with orientation parameters corresponding to T = 20 mK as discussed in text.  $W_0$  is the sum of *P*-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  $(\sin \eta \text{ where } \delta = |\delta| e^{i\eta})$ .  $\mathbf{C}^{\text{TRIV}}$ has been deduced from the data of Table I corrected for precessional effects as described in text.

Cascade (keV)	W <sub>0</sub>	$W_T$ (sin $\eta$ )	$W_T/W_0$ (sin $\eta$ )	<b>Q</b> <sup>TRIV</sup> (×10 <sup>-4</sup> )	$\sin\eta$
198-110	1.060	-0.0128	-0.012	$-18 \pm 15$	$+0.15 \pm 0.12$
177 - 131	1.066	-0.0068	-0.0064	$-81 \pm 26$	$\geq 0^{a}$
283-114	1,156	-0.001 34	-0.0012	$+1.9 \pm 5.8$	$-0.17 \pm 0.5$

<sup>a</sup> In this case we attempted to correct for a large (~10°) precession perturbation in a way which may be inadequate, resulting in  $\sin \eta = +1.3 \pm 0.4$  (see text).

ienced by Yb nuclei when dilutely alloyed into gold. This field has an effective value  $H_{\rm hf} = 1.77$  MOe up to 1.86 MOe as the external field on the sample is varied from 1 kOe to 10 kOe.<sup>8,9</sup> With our applied field of 5 kOe, we have taken  $H_{\rm hf} = +1.8$  MOe as a reasonable value. Hence significant polarization of the <sup>169,175</sup>Yb ground states (which have magnetic moments  $(-0.63 \pm 0.09)\mu_N$  and  $(+0.40 \pm 0.05)\mu_N$ , respectively)<sup>10</sup> 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  $1.4 \times 10^{12}$  thermal neutrons/cm<sup>2</sup> 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×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 is shown in Fig. 4. The working samples had initial <sup>175</sup>Yb activity of 5  $\mu$ Ci.

Two 7.6 cm ×7.6 cm NaI(Tl) detectors  $(7\frac{1}{2}\%$  resolution at 1.33 MeV) were positioned in the horizontal plane of distances of 5 cm for the <sup>169</sup>Tm and from 7 cm to 5 cm for the shorter-lived <sup>175</sup>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 <sup>175</sup>Lu 283-114 keV cascade, for a total of 175 h of data accumu-



FIG. 4. Schematic representation of experimental arrangement indicating a portion of the cryogenics, source mounting, and magnetic shielding; vertical cross section (a), horizontal cross section (b).

$E_{\gamma_1} - E_{\gamma_2}$	Raw measured asymmetries $\times 10^4$		Systematic $\gamma_1$ asymmetries $\times 10^4$		Background asymmetries ×10 <sup>4</sup>		Corrected asymmetries $\times 10^4$	
(keV)'2	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 ±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		$+4.2 \pm 3.3$		$+2.5 \pm 0.2$		$+4.1 \pm 3.2$		$+1.9 \pm 5.8$

lation. For the two <sup>189</sup>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 an automatic data collection system described elsewhere.<sup>11</sup> By this means an accumulative difference measurement of the correlation function was obtained in order to determine the asymmetry

$$a = \frac{W(\dagger) - W(\dagger)}{W(\dagger) + W(\dagger)}, \qquad (3)$$

where  $W(\dagger)$  indicates the directional correlation function measured with nuclear spin pointing up (and *vice versa*) and an azimuthal angle  $\Phi$  of 135° from detector 1 (detecting  $\mathbf{k}$ ) to detector 2 (detecting  $\mathbf{k}'$ ) (Fig. 3).

The methods of data accumulation and directional correlation were as described in more detail in previous papers.<sup>4,11,12</sup> The spectra were accumulated for both singles and coincidences. To obtain good coincidence efficiency, NaI(T1) detectors had been chosen. Hence for <sup>169</sup>Tm the 198 keV ( $\gamma_1$ ) and 177 keV ( $\gamma_1$ ) radiations could not be fully resolved, and the single channel analyzer (SCA) windows were set to include both. As a result, that coincidence spectrum gives the partially resolved 110 and 131 keV peaks so that the two <sup>169</sup>Tm correlation asymmetries could be measured simultaneously but separately. Typical peak to background ratios were 4 to 1 in the coincidence spectra.

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, along with the standard deviations and Poisson  $\chi^2$  test values, were automatically computed. Residual systematic asymmetries were checked for in the aforementioned SCA and centroid measurements. Careful magnetic shielding procedures, described elsewhere,  $^{4,12}$  effectively eliminated gain shifts due to magnetic interaction with the detector phototubes. Such gain shifts were consistently less than 0.02%.

# **III. 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 II. 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  $\gamma_1$  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 II, were made.

(2) Asymmetries in the background regions of the coincidence spectra were measured as listed in Table II. Corrections were made accordingly. These corrections had an effect of 10-20% on the measured asymmetries.

(3) Finally, corrections for representative trueto-chance ratios of 4.5:1 and 7:1 in the <sup>169</sup>Tm and <sup>175</sup>Lu measurements, respectively, were made by appropriate multiplicative factors. In the last two columns of Table II are the asymmetries after the three corrections listed above and finally after correction for the standard factors  $Q_2(\gamma_1)Q_2(\gamma_2)$ for finite detector angular resolution.

#### **IV. 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 the Appendix.

For the two correlations in <sup>169</sup>Tm, the measured (corrected) asymmetries  $\mathbf{G}_{m}^{corr}$  at warm temperatures listed in Table II were found to correspond to precessions of the intermediate states  $(I_2)$  of  $2\frac{1}{2}$ 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,<sup>13</sup> respectively, 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 <sup>169</sup>Yb coming through relatively short-lived states. However, such an interpretation of the precession does tend to support a previous measurement<sup>14</sup> of that hyperfine field which put an upper limit of 1 MOe on its magnitude.

# 198-110 keV correlation in <sup>169</sup>Tm

An effective precession angle for the intermediate state of  $(2.6 \pm 0.13)^{\circ}$  (deduced from the measured warm correlation asymmetry) led to the prediction that the "warm" asymmetry should be  $4.9 \times 10^{-4}$  larger than the asymmetry measured at "cold" temperatures, in the absence of TRIV. However, this experiment resulted in a much larger difference,  $22.6 \times 10^{-4}$ . 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  $\alpha$ =  $(18 \pm 15) \times 10^{-4}$ , leading to a phase of the E2 component with respect to the M1 component of one 198 keV radiation equal to  $\sin \eta = +0.15 \pm 0.12$ .

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.

effect:

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 and 20  $\mu$ sec, respectively, for these components of the  $\gamma$  radiation,<sup>15</sup> 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<sup>16</sup>  $H_W(M1) = 0.67 \times 10^6$  and  $H_W(E2) = 0.6 \times 10^3$ .

As we pointed out in an earlier paper,<sup>4</sup> 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

$$\frac{\operatorname{Im}\langle 2 \| M1 \| 1 \rangle}{|\langle 2 \| M1 \| 1 \rangle_{w}|} = \frac{-\sin \eta_{a}}{|H_{w}(M1)|^{1/2}} = -(1.8 \pm 1.5) \times 10^{-4}$$

and

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

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<sup>12,17-23</sup> shows the present null result to be of comparable sensitivity.

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  $I^{\pi} = \frac{7}{2}^+$ ,  $K = \frac{1}{2}$  level at 139 keV into the 316 keV state, since this is the only known nearby state with equal spin and parity.<sup>24</sup> Following the arguments of Ref. 5, if this mixing occurs, then some of the out of phase radiation would be "mixed into" the observed 198 keV  $\gamma$  rays. We would write for the 316 keV initial state  $|1\rangle$ , with the convention  $|I^{\pi}K\rangle$ ,  $|1\rangle = |\frac{7}{2}\rangle + i\epsilon |\frac{7}{2} + \frac{1}{2}\rangle$ , 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  $H = H_0 + iH_{TRIV}$ . 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 Ref. 5, the following expression for the TRIV

$$\mathfrak{O} = \frac{|\delta| \sin\eta}{1+\delta^{2}} \\
= \epsilon \left[ \frac{\langle 2e \| E2 \| 1e \rangle \langle 2e \| M1 \| 1o \rangle - \langle 2e \| M1 \| 1e \rangle \langle 2e \| E2 \| 1o \rangle}{\langle 2e \| M1 \| 2e \rangle^{2} + \langle 2e \| E2 \| 1e \rangle^{2}} \right].$$
(4)

This allows for an evaluation of the first order perturbation parameter  $\epsilon$  and thus of  $\langle 2 | H_{\text{TRIV}} | 1 \rangle$ . In order to do so, the four matrix elements in Eq. (4) must be computed as follows. The reduced matrix elements  $\langle 2e \parallel M1 \parallel 1e \rangle$  and  $\langle 2e \parallel E2 \parallel 1e \rangle$  are obtained from their definitions as square roots of partial transition probabilities for the *M*1 and *E*2 components of  $\gamma$  radiation in the 198 keV transition. Hence we have

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

and

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

The  $\langle 2e \| \lambda \|$  10 matrix elements can be estimated by determining the partial transition probabilities for M1 and E2  $\gamma$  radiation in the 20.8 keV transition from the  $\frac{1}{2}$  139 keV state to the  $\frac{5}{2}$  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  $\delta^2(E2/M1)$ = 7.15 × 10<sup>-4</sup> (Ref. 13) and a deduced branching ratio 0.335.<sup>25,26</sup> The results are

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

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  $K = \frac{1}{2}$  ground state rotational band obtained from energy line fits in <sup>169</sup>Tm <sup>13</sup> to theoretical formulas for the reduced transition probabilities in rotational bands.<sup>27, 28</sup>)

From these four reduced matrix elements, Eq. (4) yields the result

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

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  $|\frac{7}{2} + \frac{3}{2}\rangle$ 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 <sup>169</sup>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  $\alpha^{\text{TRIV}} = -(81 \pm 26) \times 10^{-4}$ , indicating a possible nonzero 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 I, that  $\sin \eta \ge 0$ .

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<sup>25, 26, 29, 30</sup> along with theoretical values for  $\alpha(M1)$  and  $\alpha(E2)$ ,<sup>15</sup> we compute the experimental partial transition rates  $T_e(M1)$ =  $2.0 \times 10^5 \text{ sec}^{-1}$  and  $T_e(E2) = 3.8 \times 10^4 \text{ sec}^{-1}$  for the 177 keV transition, with hindrances  $H_W(M1)$ =  $8.8 \times 10^5$  and  $H_W(E2) = 3.1 \times 10^2$  Weisskopf units. Hence, as fractions of the single-particle Weisskopf estimates, we have

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

and

$$\frac{\operatorname{Im}\langle 2 \| E2 \| 1 \rangle}{\langle 2 \| E2 \| 1 \rangle_{W}} = (-1.1 \times 10^{-3}) \sin \eta ,$$

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  $|\frac{7}{2}+\frac{1}{2}\rangle$  139 keV and  $|\frac{7}{2}+\frac{1}{2}\rangle$ 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 Ref. 5, 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  $\epsilon = \langle 2 |H_{\text{TRIV}} |1\rangle / 0.177$  MeV. With this kind of mixing, then, from first-order

perturbation it is possible to show that

$$\mathfrak{O} = \epsilon \frac{\left[\langle 2e \parallel L' \parallel 1e \rangle \left(\langle 2e \parallel L \parallel 2e \rangle - \langle 1e \parallel L \parallel 1e \rangle \right) - \langle 2e \parallel L \parallel 1e \rangle \left(\langle 2e \parallel L' \parallel 2e \rangle - \langle 1e \parallel L' \parallel 1e \rangle \right]}{\left|\langle 2e \parallel L \parallel 1e \rangle\right|^2 + \left|\langle 2e \parallel L' \parallel 1e \rangle\right|^2}$$
(5)

$$=\frac{\epsilon|\delta|}{1+\delta^2} \left[ \frac{\langle 2e \parallel L \parallel 2e \rangle - \langle 1e \parallel L \parallel 1e \rangle}{\langle 2e \parallel L \parallel 1e \rangle} - \frac{\langle 2e \parallel L' \parallel 2e \rangle - \langle 1e \parallel L' \parallel 1 \rangle}{\langle 2e \parallel L' \parallel 1e \rangle} \right].$$
(6)

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,  $\langle || \mathfrak{M}(\lambda) || \rangle$ ,<sup>31</sup> rather than the transition rate defined matrices used earlier. Equation (6) can be recast in the following form:

$$\sin \eta = \epsilon \left\{ \frac{\langle 2e \| \mathfrak{M}(M1) \| 2e \rangle - \langle 1e \| \mathfrak{M}(M1) \| 1e \rangle}{\langle 2e \| \mathfrak{M}(M1) \| 1e \rangle} - \frac{\langle 2e \| \mathfrak{M}(E2) \| 2e \rangle - \langle 1e \| \mathfrak{M}(E2) \| 1e \rangle}{\langle 2e \| \mathfrak{M}(E2) \| 1e \rangle} \right\}.$$
(7)

The matrices of the form  $\langle 1e \| \mathfrak{M}(\lambda) \| 1e \rangle$  are related to observable moments by the relations<sup>32</sup>

$$\mu = (\frac{4}{3}\pi)^{1/2} (2I+1)^{-1/2} \langle II10 | II \rangle \langle IK || \mathfrak{M}(M1) || IK \rangle$$
(8)

and

$$eQ = \left(\frac{16}{5}\pi\right)^{1/2} (2I+1)^{-1/2} \langle II20 | II \rangle \langle IK || \Im (E2) | IK \rangle.$$
(9)

Using a value  $\mu = (0.154 \pm 0.008)\mu_N^{33}$  for the magnetic dipole moment of the 316 keV state and  $\mu = (1.318 \pm 0.065)\mu_N$  for the 139 keV state<sup>13</sup> we obtain  $\langle \frac{7}{2} \frac{7}{2} \| \mathfrak{M}(M1) \| \frac{7}{2} \frac{7}{2} \rangle = 0.24 \pm 0.01$  and  $\langle \frac{7}{2} \frac{1}{2} \| \mathfrak{M}(M1) \| \frac{7}{2} \frac{1}{2} \rangle = (2.1 \pm 0.1)\mu_N$ .

Using a value  $(7.7 \pm 0.2) \times 10^2 \ e \ fm^2$  for the intrinsic electric quadrupole moment  $Q_0$  of the ground state rotational band in <sup>169</sup>Tm, we assign through the relation

$$Q = Q_0 \frac{3K^2 - I(I+1)}{(I+1)(2I+3)}$$

the following quadrupole moments:

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

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

which result in the following matrix elements, using Eq. (9):

$$\left\langle \frac{7}{2} \frac{7}{2} \| \mathfrak{M}(E2) \| \frac{7}{2} \frac{7}{2} \right\rangle = (-4.7 \pm 0.1) \times 10^2 \ e \ \mathrm{fm}^2$$

$$\left\langle \frac{7}{2} \frac{1}{2} \| \mathfrak{M}(E2) \| \frac{7}{2} \frac{1}{2} \right\rangle = (+3.2 \pm 0.1) \times 10^2 \ e \ \mathrm{fm}^2$$
.

The transition matrix elements are found as follows. The reduced transition probabilities  $B(\lambda)$  are evaluated from the partial lifetimes of the *M*1 and *E*2 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.,

$$B(M1) = [\tau(M1)(1.76 \times 10^{13})E^3 \text{ (MeV)}]^{-1}\mu_N^2$$
$$= (2.0 \pm 0.3) \times 10^{-6}\mu_N^2$$

and

$$B(E2) = [\tau(E2)(1.22 \times 10^9)E^5 (\text{MeV})]^{-1} e^2 \text{fm}^4$$
  
= 0.18 ± 0.03  $e^2 \text{fm}^4$ .

From the relation

$$B(\lambda) = \frac{1}{2I_i + 1} |\langle I_f K_f \| \Re(\lambda) \| I_i K_i \rangle|^2$$
(10)

we find

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

and

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

recalling that the E2/M1 mixing ratio is negative. Thus, solving for  $\epsilon$  in Eq. (7) we have

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

so that

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

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.<sup>34</sup>

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  $\gamma$  ray is measured, and the initial 316 keV level with spin *I* is polarized, then a search for a term of the form  $(\mathbf{k} \cdot \mathbf{I}_1 \times \vec{\epsilon})(\mathbf{I}_1 \cdot \mathbf{k})$ , could be performed.<sup>1</sup>

# 283-114 keV correlation in <sup>175</sup>Lu

Since <sup>175</sup>Lu does not experience any significant hyperfine field in gold, precessional effects were not important, and the corrected cold data yielded directly

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

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  $\gamma$  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

$$\frac{\mathrm{Im}\langle 2||M2||1\rangle}{|\langle 2||M2||1\rangle_{W}|} = -0.05 \pm 0.16.$$

So the present work has 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.

#### **V. 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 <sup>169</sup>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 ~ $10^{-3}$  in the *E*1 component and ~ $10^{-1}$  in the *M*2 component of the 283 keV *E*1 + *M*2  $\gamma$  radiation in <sup>175</sup>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  $\gamma$  ray in <sup>169</sup>Tm by some method other than directional correlations would provide a good TRIV test.

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## APPENDIX

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<sup>10</sup> 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 <sup>169</sup>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  $\gamma_1$ , and  $\delta_1(E2/M1) = -0.30$ . Assuming  $G_1/G_2 \sim 1.75$  and, by deduction from the tables of Ref. 35, 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  $\gamma$  ray<sup>10</sup> we have for the angular distribution of  $\gamma_2$ ,  $A_2$  $= 0.667 \pm 0.046$  and  $A_4 = 0.02$ . For the T = 4 K correlation, we used  $B_1(I_1) = -0.00016$ ,  $B_2(I_1) = B_3(I_1)$  $=B_{A}(I_{1})\equiv 0.$ 

#### 177-131 keV

Since the 177 keV  $\gamma$  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  $\gamma$  ray are  $A_2(\gamma_2) = -0.468$  and  $A_4(\gamma_2) = -0.358$ . For the 177 keV radiation,  $\delta_1 = -0.44 \pm 0.01$ .<sup>10</sup>

## 283-114 keV

From the  $\beta$  decay of the <sup>175</sup>Yb ground state to the 396 keV level  $(I_1)$  in <sup>175</sup>Lu, the deorientation parameters are  $U_1 = 0.975$ ,  $U_2 = 0.925$ ,  $U_3 = 0.8498$ ,  $U_4 = 0.7495$ .<sup>35</sup> From Ref. 7 we have for the 20 mK alignment parameters of the <sup>175</sup>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  $\gamma$  ray,  $\delta_1(M2/E1)$  $-0.036 \pm 0.004$ .<sup>10</sup> No attenuation of the  $\gamma_1$  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,  $\delta_2(E2/M1) = 0.45$ ,<sup>10</sup> which gives  $A_2(\gamma_2)$ = -0.452,  $A_4(\gamma_2) = +0.098$ .

- <sup>2</sup>C. F. Clement and L. Heller, Phys. Rev. Lett. <u>27</u>, 545 (1971);
   C. F. Clement, Ann. Phys. (N. Y.) <u>75</u>, 219 (1973).
- <sup>3</sup>J. H. Christenson, J. W. Cronin, V. L. Fitch, and R. Turlay, Phys. Rev. Lett. <u>13</u>, 138 (1964).
- <sup>4</sup>B. T. Murdoch, C. E. Olsen, W. A. Steyert, and K. S. Krane, Phys. Rev. Lett. <u>31</u>, 1514 (1973).
- <sup>5</sup>W. A. Steyert and K. S. Krane, Phys. Lett. <u>47B</u>, 294 (1973).
- <sup>6</sup>K. S. Krane, R. M. Steffen, and R. M. Wheeler, Nucl.

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<sup>&</sup>lt;sup>1</sup>R. J. Blin-Stoyle, Fundamental Interactions and the Nucleus (North-Holland, Amsterdam, 1973); F. Boehm, in Hyperfine Structure and Nuclear Radiations (North-Holland, Amsterdam, 1968), p. 280.

Data A11, 351 (1973).

- <sup>7</sup>K. S. Krane, Lawrence Berkeley Laboratory Report No. LNL-1686, 1973. (Available National Technical Information Service, U.S. Department of Commerce, 5258 Port Royal Road, Springfield, Virginia 22150).
- <sup>8</sup>D. Spanjaard, R. A. Fox, J. D. March, and N. J. Stone, in *Hyperfine Interactions in Excited Nuclei*, edited by G. Goldring and R. Kalish (Gordon and Breach, New York, 1971), p. 113.
- <sup>9</sup>D. Spanjaard, J. D. Marsh, and N. J. Stone, J. Phys. F <u>3</u>, 1243 (1973).
- <sup>10</sup>K. S. Krane, C. E. Olsen, and W. A. Steyert, Nucl. Phys. <u>A197</u>, 352 (1972).
- <sup>11</sup>L. E. Handy and W. A. Steyert, IEEE Trans. on Nucl. Sci. NS-21, 857 (1974).
- <sup>12</sup>K. S. Krane, B. T. Murdoch, and W. A. Steyert, Phys. Rev. C 10, 840 (1974).
- <sup>13</sup>C. Günther, H. Hübel, A. Kluge, K. Krien, and H. Toschinski, Nucl. Phys. A123, 386 (1969).
- <sup>14</sup>W. A. Stevert and K. S. Krane, unpublished.
- <sup>15</sup>Also using electron conversion tables of R. S. Hager and E. C. Seltzev, Nucl. Data A4, 1 (1968).
- <sup>16</sup>A. Bohr and B. R. Mottelson, *Nuclear Structure* (Benjamin, New York, 1969), Vol. 1, pp. 382 and 389.
- <sup>17</sup>J. Eichler, Nucl. Phys. <u>A120</u>, 535 (1968).
- <sup>18</sup>J. Kajfosz, J. Kopecky, and J. Honzato, Nucl. Phys. <u>A120</u>, 225 (1968).
- <sup>19</sup>M. Garrell, H. Frauenfelder, D. Ganek, and D. C. Sutton, Phys. Rev. <u>187</u>, 1410 (1969).
- <sup>20</sup>O. C. Kistner, Phys. Rev. Lett. <u>19</u>, 872 (1967).

- <sup>21</sup>R. B. Perkins and E. K. Ritter, Phys. Rev. <u>174</u>, 1426 (1968).
- <sup>22</sup>M. J. Holmes, W. D. Hamilton, and R. A. Fox, Nucl. Phys. <u>A199</u>, 401 (1973).
- <sup>23</sup>M. Atac, B. Chrisman, P. Debrunner, and H. Frauenfelder, Phys. Rev. Lett. 20, 691 (1968).
- <sup>24</sup>An extensive survey for γ rays of intensity 10<sup>-4</sup> or more quanta/decay from levels in this region is presented by A. N. Miminoshvili, V. V. Marav'eva, and A. A. Sorokin, Yad. Fiz. <u>10</u>, 201 (1969) [transl.: Sov. J. Nucl. Phys. <u>10</u>, 113 (1970)].
- <sup>25</sup>S. K. Yen, D. L. Salie, and E. Tomchuk, Can. J. Phys. 50, 2348 (1972).
- <sup>26</sup>P. Alexander and F. Boehm, Nucl. Phys. <u>46</u>, 108 (1963).
- <sup>27</sup>O. Nathan and S. G. Nilsson, in Alpha-, Beta-, and Gamma-Ray Spectroscopy, edited by K. Siegbahn (North-Holland, Amsterdam, 1965), p. 653.
- <sup>28</sup>E. N. Kaufmann, J. D. Bowman, and S. K. Bhattacheryya, Nucl. Phys. <u>A119</u>, 417 (1968).
- <sup>29</sup>J. E. Brown and E. N. Hatch, Nucl. Instrum. Methods <u>47</u>, 185 (1967).
- <sup>30</sup>E. N. Hatch, F. Boehm, P. Marmier, and J. W. M.
- Dumond, Phys. Rev. <u>104</u>, 745 (1956).
- <sup>31</sup>See Ref. 27, p. 651.
- <sup>32</sup>See Ref. 16, pp. 333 and 337.
- <sup>33</sup>A. K. Nigam and R. Bhattacharyya, Nucl. Phys. <u>A181</u>, 298 (1972).
- <sup>34</sup>R. J. Blin-Stoyle and F. A. Bezerra Coutinho, Nucl. Phys. <u>A211</u>, 157 (1973).
- <sup>35</sup>K. S. Krane, Nucl. Data <u>A11</u>, 407 (1973).



FIG. 4. Schematic representation of experimental arrangement indicating a portion of the cryogenics, source mounting, and magnetic shielding; vertical cross section (a), horizontal cross section (b).