## Enhanced-Sensitivity $\gamma$ - $\gamma$ Correlation Test of Time-Reversal Invariance in <sup>180</sup>Hf

B. T. Murdoch, \* C. E. Olsen, and W. A. Steyert

Los Alamos Scientific Laboratory of the University of California, Los Alamos, New Mexico 87544

and

K. S. Krane

Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720

(Received 9 October 1973)

Directional correlations of the strongly hindered 501-keV  $\gamma$  ray with subsequent 332and 215-keV  $\gamma$  rays from polarized <sup>180m</sup>Hf showed an average time-reversal-invarianceviolating asymmetry of  $(2.8 \pm 5.1) \times 10^{-4}$ , indicating no substantial effect. The phase angle between the 501-keV E3 and M2 components was  $\sin\eta = +0.048 \pm 0.087$  and the out-ofphase components of the multipole matrix elements were, respectively,  $(+1.1 \pm 2.1) \times 10^{-6}$ and  $(-2.1 \pm 3.8) \times 10^{-9}$  Weisskopf units.

The question of time-reversal invariance is basic to the understanding of the fundamental interactions governing physical processes, not only in a macroscopic (thermodynamic) sense, but particularly in a microscopic (quantum mechanical) sense. The possibility of violation of time-reversal invariance raises questions of the reversibility in time of the rate equations describing physical processes, or indeed of the occurrence of certain processes in a time-reversed frame of reference.<sup>1-3</sup> If *CPT* is invariant, evidence for such *T*-invariance violations follows from the *CP* (charge conjugation-parity) nonconservation observed in the decay of the long-lived neutral *K* meson.<sup>4</sup>

It has been well established<sup>5</sup> that appropriate selections of electromagnetic transitions in nuclei can result in magnifications of observables arising from broken symmetries in nucleon-nucleon interactions. In particular, nuclear structure effects occasionally serve to inhibit the "regular" (non-symmetry-breaking) transitions, so that, as a result, the "irregular" (symmetrybreaking) transitions are relatively enhanced and more easily observed.

Such enhancement effects have been well demonstrated with regard to violations of spatial symmetry (parity).<sup>6,7</sup> Only recently has this enhancement concept been applied to considerations of time-reversal-invariance violation (TRIV) measured in nuclear electromagnetic radiation.<sup>8,9</sup> This report describes what is apparently the first nuclear  $\gamma$ - $\gamma$  TRIV test utilizing the enhancement concept.

This test was made on the 501-keV transition in  $^{180}$ Hf (Fig. 1). The 8<sup>-</sup> intrinsic state decays to the 6<sup>+</sup> level of the ground-state rotational band with the emission of  $\gamma_1$ , with mixing ratio  $\delta(E3/M2) = 5.3$ . A parity-nonconserving *E*2 contribution has been observed.<sup>6,7</sup> Although the parity-nonconserving *E*2 matrix element is very small (only  $4 \times 10^{-10}$  times the Weisskopf single-particle estimate), the strong *K* hindrances ( $\Delta K = 8$ ) of the *M*2 and *E*3 matrix elements ( $1.3 \times 10^7$  and  $0.5 \times 10^5$ , respectively) permit the *E*2 to be easily evidenced, with  $\delta(E2/M2) = -0.04$ .

An angular correlation test of TRIV was performed by measuring the resulting asymmetry in the average value of the scalar quantity  $\langle (\mathbf{\bar{I}} \cdot \mathbf{\bar{k}} \times \mathbf{\bar{k}'}) \times \langle \mathbf{\bar{k}} \cdot \mathbf{\bar{k}'} \rangle \rangle$ .<sup>10</sup> Here  $\mathbf{\bar{k}}$  represents the momentum of  $\gamma_1$ ,  $\mathbf{\bar{k}'}$  represents the momentum of either the succeeding  $\gamma_2$  or  $\gamma_3$  (see Fig. 1), and  $\mathbf{\bar{I}}$  is the nuclear polarization. Time-reversal symmetry requires that this scalar average to zero.

Hafnium enriched to  $87\%^{179}$ Hf was arc melted in ferromagnetic ZrFe<sub>2</sub> to form (Zr<sub>0.95</sub>Hf<sub>0.05</sub>)Fe<sub>2</sub>. The sample was cut into needle-shaped pieces 1 ×1×7 mm<sup>3</sup> which were irradiated with thermal



FIG. 1. Decay scheme of <sup>180m</sup>Hf, showing the radiations  $\gamma_1$ ,  $\gamma_2$ , and  $\gamma_3$  discussed in the text.

neutrons. Individual radioactive samples were attached to a  ${}^{3}\text{He}{}^{-4}\text{He}$  dilution refrigerator with the principal axis vertical, and cooled to 20-35 mK. A vertical magnetic field of 5 kOe, produced by a pair of superconducting approximately Helmholtz coils, was applied to polarize the ferromagnetic host and thus the  ${}^{180m}\text{Hf}$  (the hyperfine field on Hf in ZrFe<sub>2</sub> is  $-200 \pm 20$  kOe).<sup>11</sup> The results reported are from eight separate sources, each providing about 20 h of useful data before becoming too weak.

Two 3-in.  $\times$ 3-in. 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 initial source activity at the beginning of "cold" data accumulation was 2.5  $\mu$ Ci with initial coincidence rates of 12/sec.

For each of the two detectors, full singles and coincidence spectra were recorded; the coincidence spectrum from one detector was gated  $(2\tau = 0.8 \ \mu \text{sec})$  by a  $\gamma_1$  pulse in the other detector. These data as well as those discussed below were accumulated and recorded by a minicomputer-based data acquisition system<sup>12</sup> over a 5- to 10-min period, following which the magnetic field was rotated through  $180^{\circ}$  by means of auxiliary coils for the next counting period; this cycle was continuously repeated for several hours.

Due to the proximity of detector photomultiplier tubes to the magnet field, elaborate shielding measures were necessary. The 2-cm-diam magnet coils were surrounded with a hollow soft iron cylinder 5 cm in diameter and 3 mm thick, almost closed on the ends, which was in turn wrapped with a superconducting lead foil. Additional conventional Conetic shielding was placed around the outside of the refrigerator Dewar. The resultant field exterior to the Dewar was less than 0.01 Oe.

The integrated singles counting rates, centroid positions, and the outputs of the single-channel

analyzers used for coincidence counting were continuously monitored to avoid false asymmetries which might have resulted from phototube gain shifts or other systematics. Measured gain shifts were less than 1 part in 5000 and gating count rate changes were less than 1 part in 3000.

A detailed description of the formalism necessary for analyzing directional correlations from oriented nuclei has recently been presented<sup>13</sup> with applications to time-reversal studies discussed in a subsequent work.<sup>14</sup> The correlation may be represented by the expression  $W = W_0$  $+ W_T$ , where  $W_0$  is the correlation function<sup>13</sup> of the two  $\gamma$  rays in the absence of TRIV, depending on the alignment  $(B_2, B_4, \ldots)$  of the state *I* and *not* on the polarization  $(B_1, B_3, \ldots)$ ;  $W_T$  is the TRIV term, which does depend on polarization and is maximized for the presently employed geometry  $\vec{k} \perp \vec{l}$ ,  $\vec{k'} \perp \vec{l}$ , and  $\angle(\vec{k}, \vec{k'}) = 3\pi/4$ .<sup>14</sup>

By reversing the direction of nuclear polarization, we search for asymmetries of the form  $a = [W(\mathbf{1}) - W(\mathbf{1})]/[W(\mathbf{1}) + W(\mathbf{1})] \approx W_T/W_0$ , assuming  $W_T \ll W_0$ .  $W(\mathbf{1})$  and  $W(\mathbf{1})$  indicate the measured correlation with nuclear spin up and down, respectively, and with  $\gamma_1$  in the detector at azimuthal angle 0 and  $\gamma_2$  at  $+3\pi/4$ .

Spurious contributions to  $W_T$  may arise from the perturbed angular correlation (PAC) of the intermediate state, or from a parity-nonconserving (PNC) component in  $\gamma_1$ . The case of <sup>180</sup>Hf provides a favorable situation with respect to the PAC effect, since the anisotropy of the angular correlation almost vanishes for both oriented and unoriented<sup>15</sup> <sup>180</sup>Hf. The PNC contribution may represent a serious drawback to the use of <sup>180</sup>Hf, since there is a large PNC effect on the 501-keV  $\gamma$  ray; however, this effect can be minimized by careful alignment of the source-detector geometry  $[(\vec{k} \times \vec{k}') \parallel \vec{I}]$  and may be accurately estimated from the asymmetry of the  $\gamma_1$  (501 keV) singles.

For the  $\gamma_1 - \gamma_2$  (and also  $\gamma_1 - \gamma_3$ ) correlation, we obtain  $W_0(T = 20 \text{ mK}) = 1.297$ ,

$$W_{T} = Q_{2}(\gamma_{1})Q_{2}(\gamma_{2})B_{1}(I)F_{2}^{21}(\gamma_{1})F_{2}(\gamma_{2})\frac{2|\delta|\sin\eta}{1+|\delta|^{2}}(-0.61\sin 2\varphi) + \dots,$$
(1)

where we assume  $(\mathbf{k} \times \mathbf{k}') \parallel \mathbf{I}$ . The various parameters are defined and discussed in Refs. 13 and 14. The dots indicate higher-order terms in the indices, the effect of which is to cancel about one third of the leading term.

The TRIV term is thus given by

$$W_{\tau} = +0.0076 \sin\eta,$$
 (2)

with  $\varphi = 3\pi/4$ ,  $|\delta| = 5.3$ , and T = 20-35 mK.

Contributions to  $W_T$  from a PAC effect may be estimated from the azimuthal shifts  $\Delta \varphi(6^+) = -0.2^\circ$ ,  $\Delta \varphi(4^+) = -1.5^\circ$ ; for the  $\gamma_1 - \gamma_3$  correlation,  $W_{PAC} \approx +4 \times 10^{-4}$  with an uncertainty of ~10% due to the uncertainty in the angular-correlation coefficients. We can also estimate the PNC contribution:  $W_{PNC} = 1.8a_{501}$ , where  $a_{501}$  is the 501-keV singles asymmetry.<sup>16</sup> After correcting the raw measured asymmetries (1) for the 501-singles asymmetry by the addition of  $(2.8 \pm 1.4) \times 10^{-4}$  and  $(2.2 \pm 0.8) \times 10^{-4}$  to the warm and cold asymmetries, respectively, (2) for a small asymmetry in the background of the coincidence spectrum by ~0.3×10<sup>-4</sup>, and (3) for the measured true-to-accidental coincidence ratio of 6.5, we have the following corrected correlation asymmetries (in units of 10<sup>-4</sup>): for  $\gamma_{-} = \gamma_{-}$ 

 $\gamma_1 - \gamma_2$ ,

 $a(\text{cold}) = 2.8 \pm 6.1, \quad a(\text{warm}) = 0.9 \pm 7.9;$ 

for  $\gamma_1 - \gamma_3$ ,

 $a(\text{cold}) = 12.0 \pm 5.5$ ,  $a(\text{warm}) = 9.1 \pm 7.6$ .

The  $\gamma_1 - \gamma_3$  correlation is subject to uncertainties associated with precession of the intermediate  $4^+$  state and the warm results suggest a possible false asymmetry. Thus, for only that correlation the warm results were subtracted from the cold results to yield  $a(\gamma_1 - \gamma_3) = (2.9 \pm 9.4) \times 10^{-4}$ . Averaged with the  $\gamma_1 - \gamma_2$  cold data (which are not expected to suffer significant precession), this yields

$$a = (+2.8 \pm 5.1) \times 10^{-4}$$

for the average asymmetry. If the  $\gamma_1 - \gamma_3$  result is not corrected by the warm data, the average result is  $a = (+7.9 \pm 4.1) \times 10^{-4}$ , indicating possible TRIV which we discount because this result includes the uncorrected, relatively less reliable  $\gamma_1 - \gamma_3$  data.<sup>14</sup>

From Eq. (2) we obtain  $\sin \eta = +0.048 \pm 0.087$ . This result may also be expressed in terms of the magnitude of the imaginary part of the multipole matrix element as

$$\operatorname{Im}\langle L\rangle = \langle L\rangle \sin\eta, \qquad (3)$$

where  $\langle L \rangle$  is the transition matrix element of multipolarity L in units of sec<sup>-1/2</sup>. From the present work, the magnitudes of any imaginary components of the M2 and E3 transition matrix elements are, respectively,  $(-1.0 \pm 1.8) \times 10^{-5}$  and  $(+1.1 \pm 1.9) \times 10^{-4}$  sec<sup>-1/2</sup>.

In order to make a meaningful comparison of these results with previous work<sup>17-24</sup> we should remove (1) final-state density effects (essentially energy dependence), (2) effects of the radiation field at the origin (*L* dependence), and (3) nuclear size effects (*A* dependence). All of this can be accomplished conveniently by a direct comparison of the measured  $\text{Im}\langle L \rangle$  with the Weisskopf single-particle estimates<sup>17</sup> for the transition matrix, written  $\langle L \rangle_{W}$ . From this work we find Im $\langle M2 \rangle | \langle M2 \rangle_W |^{-1} = (-2.1 \pm 3.8) \times 10^{-9}$ , and Im $\langle E3 \rangle \times | \langle E3 \rangle_W |^{-1} = (+1.1 \pm 2.1) \times 10^{-6}$ , while from previously published<sup>17-24</sup> results on M1-E2 transitions, one finds upper limits on  $|\text{Im}\langle L \rangle | \langle L \rangle_W |^{-1}$  ranging from  $10^{-1}$  to  $10^{-4}$ . Thus it can be seen that any TRIV contribution to the  $\gamma$ -transition amplitudes is small, both for the previously studied M1-E2 transitions and especially for the presently reported M2-E3 transition.

In the present work, even in a transition where hindrance of the "regular" transition results in enhancement of a  $10^{-7}$  parity-nonconserving. presumably two-body, nucleon-nucleon interaction to produce a large (10<sup>-2</sup>) parity-nonconserving effect, no TRIV effect was observed. Parity nonconservation is thought to be associated with a two-body interaction which could be partially hindered as are normal two-body operators. But TRIV is thought to occur through three-body nucleon-nucleon interactions and through two-body transition operators.<sup>4</sup> (Normal transition operators are one-body.) In either case, these unusual TRIV operators would not be hindered in the same way as are normal or parity-nonconserving operators, and we may conclude that TRIV enhancement from hindrance could be a larger effect than the previously described PNC enhancement. It would seem a reasonable supposition that an effect of intrinsic relative magnitude  $10^{-7}$ in the nuclear Hamiltonian might be enhanced to a measurable effect in the observed radiation. Since this is not seen, the possibility of a TRIV arising from hypercharge-conserving ( $\Delta Y = 0$ , "millistrong") or electromagnetic interactions (relative intrinsic amplitude 10<sup>-3</sup>) may perhaps be discounted in favor of hypercharge-changing  $(\Delta Y = 1, \text{``milliweak,'' amplitude 10^{-8}}, \text{ or } \Delta Y = 2,$ superweak, amplitude 10<sup>-13</sup>) interactions.<sup>25</sup> Model-dependent estimates of the nuclear and electromagnetic multipole matrix elements which support these conclusions will be discussed in a more detailed description of the present work currently in preparation.<sup>26</sup> Clearly, very careful and detailed theoretical analysis of these results should be undertaken to see precisely what forms of TRIV potential can be rejected, based on the experimental results.

One of the authors (B.T.M.) expresses gratitude for the support provided by Associated Western Universities and for the hospitality of the Los Alamos Scientific Laboratory.

<sup>\*</sup>U.S. Atomic Energy Commission-Associated West-

ern University Fellow. Premanent address: Physics Department, Utah State University, Logan, Utah 84321.

<sup>1</sup>R. J. Blin-Stoyle, Fundamental Interactions and The Nucleus (North-Holland, Amsterdam, 1973).

<sup>2</sup>A. Bohr and B. R. Mottelson, Nuclear Structure (Benjamin, New York, 1969), Vol. I, Chap. 1.

<sup>3</sup>An elementary discussion is found in R. G. Sachs, Science 176, 587 (1969).

<sup>4</sup>J. H. Christenson, J. W. Cronin, V. L. Fitch, and

R. Turlay, Phys. Rev. Lett. 13, 138 (1964).

<sup>5</sup>See, for instance, the review by E. M. Henley, Annu. Rev. Nucl. Sci. 19, 367 (1969).

<sup>6</sup>P. Jenschke and P. Bock, Phys. Lett. 31B, 65 (1970). Their circular polarization results have been confirmed in several other laboratories.

<sup>7</sup>K. S. Krane, C. E. Olsen, J. R. Sites, and W. A. Steyert, Phys. Rev. C 4, 1906 (1971).

<sup>8</sup>C. F. Clement and L. Heller, Phys. Rev. Lett. 27, 545 (1971); C. F. Clement, Ann. Phys. (New York) 75, 219 (1973).

<sup>9</sup>W. A. Steyert and K. S. Krane, to be published.

<sup>10</sup>F. Boehm, in Hyperfine Structure and Nuclear Radia tions, edited by E. Matthias and D. A. Shirley (North-Holland, Amsterdam, 1968), p. 279.

<sup>11</sup>H. J. Körner, F. E. Wagner, and B. D. Dunlap, Phys. Rev. Lett. 27, 1593 (1971).

<sup>12</sup>L. E. Handy and W. A. Stevert, to be published.

<sup>13</sup>K. S. Krane, R. M. Steffen, and R. M. Wheeler,

Nucl. Data, Sect. A 11, 351 (1973).

<sup>14</sup>K. S. Krane, University of California Lawrence Berkeley Laboratory Report No. LBL-1686, 1973 (unpublished).

<sup>15</sup>E. Bodenstedt, H. J. Körner, E. Gerdau, J. Radeloff, C. Günther, and G. Strube, Z. Phys. 165, 57 (1961).

<sup>16</sup>For  $\gamma$ -ray energies as high as 501 keV, we do not expect any significant false asymmetry from internal conversion interference.

<sup>17</sup>See Ref. 2, pp. 382 and 389.

<sup>18</sup>J. Eichler, Nucl. Phys. <u>A120</u>, 535 (1968).

<sup>19</sup>J. Kajfosz, J. Kopecky, and J. Honzatko, Nucl. Phys. A120, 225 (1968).  $\frac{20}{20}$  M. Garrell, H. Frauenfelder, D. Ganek, and D. C.

Sutton, Phys. Rev. 187, 1410 (1969).

<sup>21</sup>O. C. Kistner, Phys. Rev. Lett. <u>19</u>, 872 (1967).

<sup>22</sup>R. B. Perkins and E. K. Ritter, Phys. Rev. <u>174</u>,

1426 (1968).

<sup>23</sup>M. J. Holmes, W. D. Hamilton, and R. A. Fox, Nucl. Phys. A199, 401 (1973).

<sup>24</sup>M. Atac, B. Chrisman, P. Debrunner, and H. Frauenfelder, Phys. Rev. Lett. 20, 691 (1968).

<sup>25</sup>See Ref. 1, p. 16, for further discussion of these interactions.

<sup>26</sup>In particular, we will show that the matrix element of the TRIV part of the nuclear Hamiltonian between the 8", K=2 octupole state expected at about 2.5 MeV and the 8<sup>-</sup> isomeric state is less than 10<sup>-6</sup> MeV.

New K = 0 Bands and Two-Phonon Gamma Vibrations in <sup>188</sup>Os and <sup>190</sup>Os<sup>+</sup>

Harbans L. Sharma and Norton M. Hintz

J. H. Williams Laboratory, University of Minnesota, \* Minneapolis, Minnesota 55455 (Received 24 August 1973)

The level structure of <sup>188</sup>Os and <sup>190</sup>Os has been investigated by the (p,t) reaction at 19.0-MeV proton energy. New 0<sup>+</sup> states were seen at 1480 and 1705 keV in <sup>188</sup>Os and at 1551 and 1734 keV in <sup>190</sup>Os. Additional evidence is presented for describing the lowest excited K=0 and K=4 bands as two-phonon  $\gamma$  vibrations.

Important information has been obtained from (t,p) and (p,t) reactions for the soft nuclei around N = 88 where there occurs a rapid change in the average nuclear shape from spherical to axially symmetric prolate as the neutron number increases. The most interesting experimental result is the strong population of excited  $0^+$  states in the residual nucleus which have average shapes similar to that of the target ground state. Thus deformed excited  $0^+$  states are seen in  $^{148}$ Sm and  $^{150}$ Sm in (*p*,*t*) reactions<sup>1</sup> and a spherical excited 0<sup>+</sup> state is seen in <sup>152</sup>Sm in the (t,p)reaction.<sup>2</sup> In the shape transition region, the usual tendency for pair correlations to force the two-particle transfer strength to the ground state

is inhibited by the small shape overlap of the Aand  $A \pm 2$  ground states as has been calculated by Takemasa, Sakagami, and Sano.<sup>3</sup> This results in the excited  $0^+$  strength being a maximum ( $\approx 100\%$  of the ground-state strength) between N = 88 and 90 where the variation of the quadrupole deformation of the ground state  $(\beta)$  with neutron number is greatest, and where excited states of the residual nucleus exist which are similar in shape to the target ground state (shape isomeric states). In order to study this effect in the somewhat different and more gradual shape transition region around W, Os, and Pt, we have studied (p,t) reactions on <sup>190</sup>Os and <sup>192</sup>Os. For these nuclei the parameter  $\beta$  is changing much more